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Effect of pretreatment using ultrasonic technique with Sic or Al_2O_3 on high temperature oxidation behavior of the FeCrAl

By

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Abstract:

High surface area with catalyst was the most required in the catalytic converter due to the harmful gas emission reduction effectively. Therefore, the aim of this paper is to show the effect of pretreatment using ultrasonic technique with SiC or Al_2O_3 on high temperature oxidation behavior of the FeCrAl. The pretreatment conducted by agitating the FeCrAl in the methanol solution consisting of SiC or Al_2O_3 followed with ultrasonic process. Nickel electroplating used for deposited nickel as catalyst application. Oxidation test then were carried out and the morphology of oxidized specimens was examined using scanning electron microscope (SEM) in combination with energy dispersive X-ray spectroscopy (EDS). The pretreatment using ultrasonic technique with SiC on FeCrAl continued nickel electroplating exhibited the highest oxidation resistance compared to Al_2O_3. The pretreatment using ultrasonic with SiC or Al_2O_3 provides roughness on FeCrAl surface which is influenced to the nickel electroplating homogenity and stability. The pretreatment using ultrasonic with SiC shows the nickel electroplating deposition more homogeneous and stable so that better high temperature oxidation resistance.

Keywords:

Pretreatment, SiC, Al_2O_3, Ultrasonic, Oxidation, FeCrAl.

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1. Introduction:

FeCrAl foils become the most popular material than ceramics in the fabrication of catalytic converter monolith due to it advantages for high temperature resistance. It also easy manufactured as honeycomb substrate with thinner walls, so that obtains a lower pressure drop and higher contact surface area [1]. In order to form catalytic converter monolith, FeCrAl was shaped through corrugate processing and spiral processing. Literature [2] and [3] mentioned that the corrugated and spiral shaped tools have been successfully designed and developed for manufacturing the hand-made full scale catalytic converter.

Next problem is how to develop washcoat on the substrate. The washcoat was known as a kind of ceramics layer (oxide layer) which has specific surface area and serves a support for catalyst materials [4]. Although the FeCrAl substrate foil contains Al that could form oxide layer (Al_2O_3) with simple high temperature treatment, but, many investigation was done to solve this problem. In previous investigation, alumina washcoat was deposited by precipitation method, impregnation method [5] and dip-coating method [6].

Ultrasonic wave or sound will generate cavitation bubbles in a liquid system. The explosion of the cavitation bubbles will produce a jet flash energy that can damage the surface of a solid material [7]. Dissolution of SiC or Al_2O_3 powders in liquid, are expected to assist in the process of surface destruction. The particles of powders can be driven by the jet flash energy and hit the surface of the material (FeCrAl). Thus, the irregular surface roughness occurs. In addition, these particles are also expected to stick into the surface of the material, which can be improved the washcoat development due to the oxide formed.

The focus of this paper presents the effect of pretreatment using ultrasonic technique with SiC or Al_2O_3 on high temperature oxidation behavior of the FeCrAl plating with nickel. The nickel plating conducted to deposited nickel as catalyst on FeCrAl substrate [8]. The early oxidation test of FeCrAl and structure of oxide layer growth after oxidation test were analyzed.

2. Experimental procedure:

2.1 Material

FeCrAl foils with 0.1 mm thickness supplied by Tyssen Krupp VDM. The substrates were cut in 2 cm X 1 cm. Three processes were conducted to the substrate i.e. ultrasonic for pretreatment, nickel electroplating, and oxidation test.

2.2 Ultrasonic pretreatment

The FeCrAl foils were treated ultrasonically using Laborette 17 ultrasonic cleaning bath with: Voltage 230 V/1~, input power 2 x 240 W/period, frequency 50-60 Hz and the ultrasound frequency 35 kHz. The bath was filled fully water. The specimens were placed into a beaker with methanol and SiC or Al_2O_3 as liquid solution. The concentration of solution made 0.2 mg/ml or by dissolve 20 mg SiC or Al_2O_3 into 100 ml methanol. The specimens then were processed into the ultrasonic bath for 0.5 hour. The illustration of ultrasound pretreatment was presented in Figure 1.

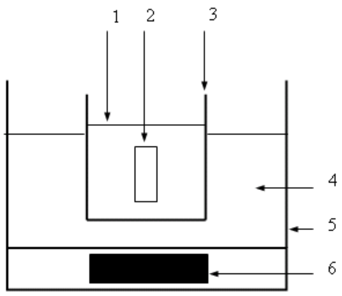

Figure (1): Schematic diagram of ultrasound pretreatment process
(1) Methanol; (2) Specimen; (3) Beaker; (4) Water; (5) Bath; (6) Ultrasonic source

Nickel electroplating used for depositing nickel as catalyst on the FeCrAl substrate. The process refers to previous project [8].

2.3 Oxidation test

The early oxidation test conducted as the catalytic converter application approach for the cold start condition, the engine running not more than 8 hours [9]. The specimens were cut in 2 mm x 2 mm. The specimens were carried out to oxidation test using thermal gravimetric analysis (TGA; Pyris Diamond, Perkin Elmer) at 1000^0C for 360 minutes. The TGA measures the weight change of a material upon heating in a highly sensitive balance. The weight changes in every minute were recorded. The graph of weight gain versus time was used to present the results. Then the kinetics reaction value of the oxidation process was obtained. For clarification, the oxide layer developed in this early oxidation test then were observed using JEOL scanning electron microscopy (SEM) model JSM-6380LA in combination with energy dispersive X-ray spectroscopy (EDS). The experimental condition of pretreatment using ultrasonic with SiC or Al_2O_3 is summarized in Table 1.

Table (1): Experimental conditions

Sample (FeCrAl)	Ultrasonic pretreatment		Nickel electroplating [8]				Oxidation test	
	Diluted materials	Time (h)	Current density (A/dm^2)	PH	Temp (^{o}C)	Time (h)	Temp (^{o}C)	Time (min)
1	SiC	0.5	3	2.5-4.5	40	0.5	1000	380
2	Al_2O_3	0.5	3	2.5-4.5	40	0.5	1000	380

3. Results:

3.1 Oxidation test.

Figure 2 shows the TGA curves of FeCrAl foils pretreatment using ultrasonic with SiC and Al_2O_3. The weight of FeCrAl foils increased rapidly on the beginning of the oxidation process at approximately first 50 minutes. It means that the oxide layer was formed on the

FeCrAl substrate. The weight of specimen pretreatment using ultrasonic with SiC decreased until minus at 100 minutes to 250 minutes. The graph attributed to the flake off phenomenon (spallation) of the oxide layer in great amounts and increase after reaches at 250 minutes. It indicated that the oxide layer start formed again. Differ with FeCrAl foils pretreatment using ultrasonic with SiC, the weight gain of specimen pretreatment using ultrasonic with Al_2O_3 decreased at about 150 minutes until 250 minutes. However, the graph did not reach minus point. It indicated that the oxide layer has lower spallation. But there was equal condition for FeCrAl foils pretreatment using ultrasonic with Al_2O_3 with the other treatment (SiC). The pretreatment using ultrasonic with Al_2O_3 increased after heated for 250 minutes. It concludes that the weight gain of specimen pretreatment using ultrasonic with Al_2O_3 greater than with SiC.

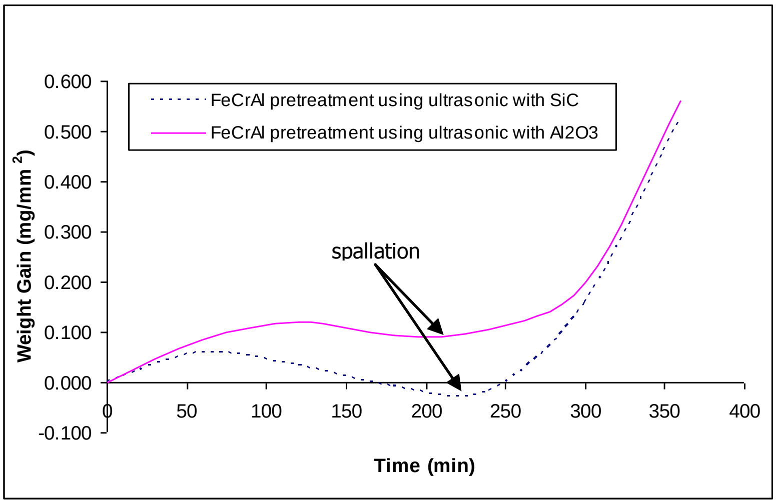

Figure (2): TG of FeCrAl pretreatment using ultrasonic with SiC or Al_2O_3

3.2 The parabolic rate constant (kp) of the kinetics of oxidation.

The parabolic rate constant (K_p) used to predict the time to failure of the FeCrAl materials [10]. The formation rate of an oxide scale, growing on the surface of a FeCrAl foils at the beginning of the oxidation test agrees with Wagner theory. At high temperature oxide films thicken according to the parabolic rate law, $x^2 \alpha t$ and the mechanism by which thickening proceeds has been explained by Wagner [11-12]. The parabolic growth equation of the film thickness with time occurs:

$$x^2 = K_p t \quad (1)$$

Where x is the layer thickness or the weight gain; t is the oxidation treatment time; K_p is parabolic rate constant. In this study the weight gain (x) resulted from mass gain per unit surface area of specimens $\left(\frac{\Delta W}{A}\right)$ [12]. Then we can write an equation to be:

$$\left(\frac{\Delta W}{A}\right)^2 = K_p t \quad (2)$$

K_p was obtained from the slope of a linear regression-fitted line of $\left(\frac{\Delta W}{A}\right)^2$ vs t plot. Figure 3 shows the nature of the fit of the parabolic rate law of the early oxidation test of FeCrAl foils pretreatment using ultrasonic with SiC or Al_2O_3 at 1000^0C. The analysis conducted on the first 60 minutes. The parabolic rate constants obtained from the present experiments are listed in Table 2. FeCrAl foils pretreatment using ultrasonic with SiC has the lower K_p than the FeCrAl foils pretreatment using ultrasonic with Al_2O_3 at 1000^0C of oxidation test. The low of K_p value indicated the long time to failure of FeCrAl substrate [10].

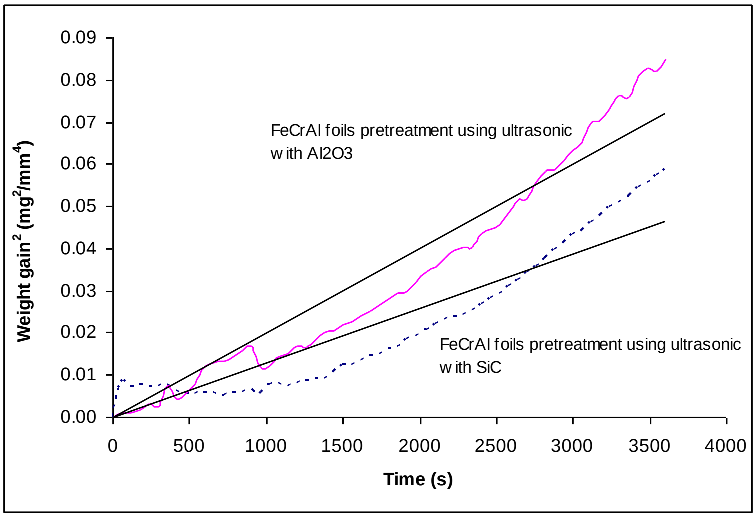

Figure (3): $\left(\frac{\Delta W}{A}\right)^2$ vs time plotted for oxidation of FeCrAl pretreatment using ultrasonic with SiC or Al_2O_3

Table (2): Parabolic rate constants for FeCrAl pretreatment using ultrasonic with SiC or Al_2O_3

Materials	kp x 10^{-3} $mg^2mm^{-4}s^{-1}$ (1000^oC)
FeCrAl foils pretreatment using ultrasonic with SiC	868.8
FeCrAl foils pretreatment using ultrasonic with Al_2O_3	934.2

3.3 Morphology of Al_2O_3 washcoat on the FeCrAl substrate.

Figure 4. summarized the observations of the cross sectional morphology using scanning electron microscope examinations (SEM) of the FeCrAl foils pretreatment using ultrasonic with SiC and Al_2O_3 powder, after oxidation test. It can be seen that cross section of the FeCrAl foils pretreatment using ultrasonic with SiC and Al_2O_3 consist of four layers. The first layer from bottom to top is FeCrAl substrate followed by Al_2O_3 as second layer and the third is nickel layer, while the fourth layer is nickel oxide layer, respectively.

The nickel electroplating on FeCrAl pretreatment using ultrasonic with SiC showed more homogeneous and stable than Al_2O_3. So that good density of nickel layer was achieved. It possible influenced by high surface roughness resulted from the FeCrAl substrate pretreatment using ultrasonic with SiC. The mean roughness measurement of FeCrAl without pretreatment, pretreatment using ultrasonic with SiC, and Al_2O_3 are 0.13µm, 0.23µm, and 0.17µm respectively. The higher roughness of FeCrAl pretreatment using ultrasonic with SiC possible influenced by random particle size and it hardness properties.

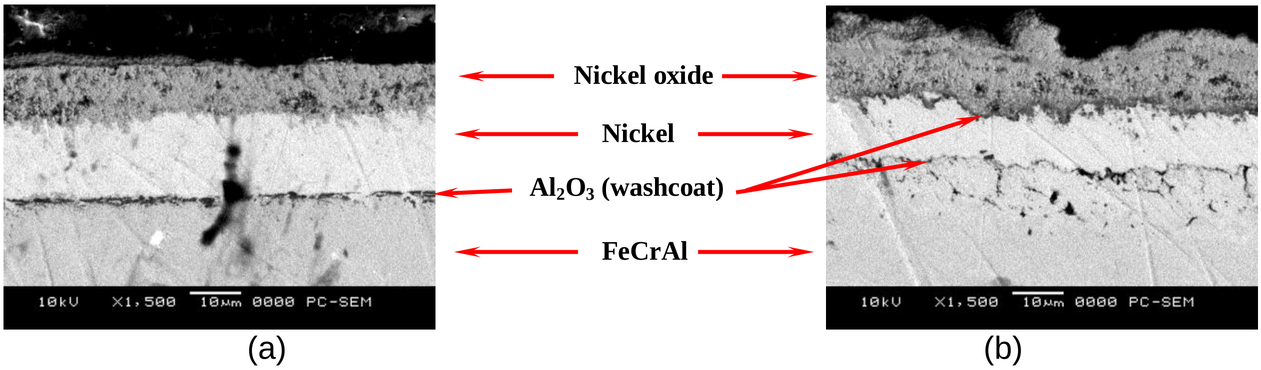

Figure (4): On cross sectional morphology of FeCrAl pretreatment using ultrasonic with (a) SiC and (b) Al_2O_3

3.4 Location of Al_2O_3 on the FeCrAl.

The mapping application of SEM and EDS are implemented for clarifying the position of chemicals component that appeared on the crossection of FeCrAl after oxidation test. The picture presented at figure 5 for pretreatment using ultrasonic with SiC powder and 6 for Al_2O_3 powder. Both of them show the existing of Al_2O_3 layer (washcoat). The Al_2O_3 layer was exist very clear and look like as a united (layer) in the FeCrAl pretreatment using ultrasonic with SiC. It was existed between FeCrAl substrate and nickel layer. While, Al_2O_3 layer exist not clear (scale) and look like separated in many area of FeCrAl pretreatment using ultrasonic with Al_2O_3. There was some phenomenon on the mapping observation. The first phenomenon was the expected SiO not appeared on the FeCrAl substrate, even though on pretreatment using ultrasonic with SiC (Figure 5). The second was Al_2O_3 scale also exists on the surface of nickel layer (Figure 6). It is possible the Al diffuse through the nickel layer then obtained the Al_2O_3 layer on the nickel layer surface.

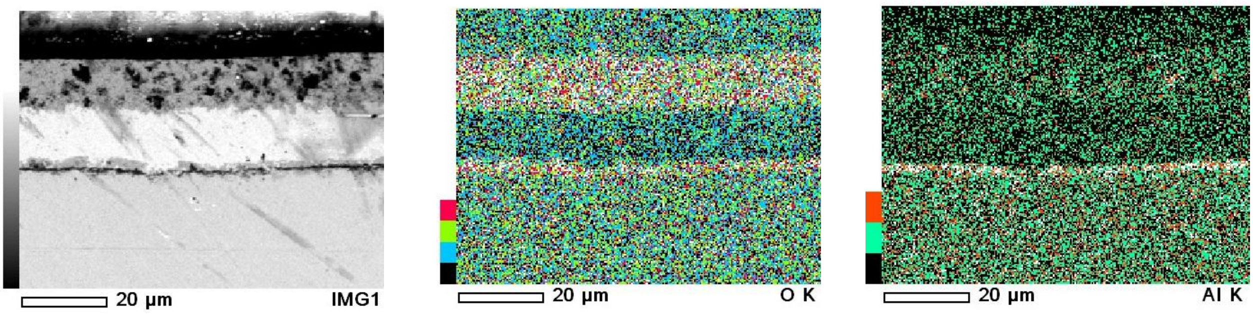

Figure (5): Mapping SEM and EDX observation for Al_2O_3 layer of FeCrAl pretreatment using ultrasonic with SiC

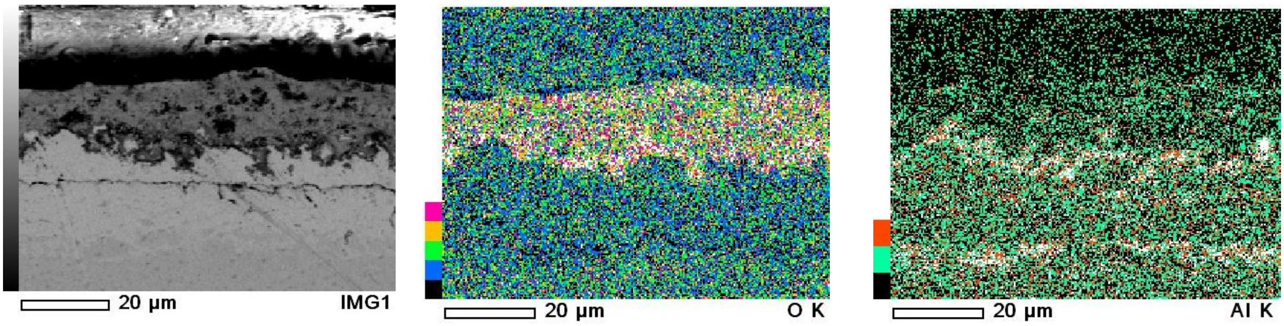

Figure (6): Mapping SEM and EDX observation for Al_2O_3 layer of FeCrAl pretreatment using ultrasonic with Al_2O_3

4. Conclusions:

The pretreatment using ultrasonic technique with SiC obtained the lower parabolic rate constant (K_p) than with Al_2O_3. The pretreatment using ultrasonic with SiC powder resulted higher surface roughness compared to Al_2O_3. It is possible influenced by particle size and it hardness properties. The higher surface roughness of FeCrAl pretreatment using ultrasonic with SiC formed the nickel electroplating deposited more homogeneous and stable. The FeCrAl pretreatment using ultrasonic with SiC resulted better high temperature oxidation resistance compared with Al_2O_3.

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