MODIFICATION OF SURFACE ROUGHNESS AND AREA OF FeCrAl SUBSTRATE FOR CATALYTIC CONVERTER USING ULTRASONIC TREATMENT

Y. Putrasari¹, P. Untoro², S. Hasan³, N. Huda³, D. Sebayang¹

¹Advanced Manufacturing and Materials Centre (AMMC), Faculty of Mechanical and Manufacturing Engineering, University Tun Hussein Onn Malaysia (UTHM)
Parit Raja, Batu Pahat 86400, Malaysia
y.putrasari@gmail.com; sulaiman@uthm.edu.my; darwin@uthm.edu.my
²Centre for Technology of Nuclear Industry Materials - National Nuclear Energy Agency (BATAN)
Kawasan Puspiptek Gd. 71, Serpong
Tangerang 15314, Indonesia
untoro@batan.go.id
³Research Centre for Electrical Power and Mechatronics - Indonesian Institute of Sciences (LIPI)
Komp. LIPI Bandung, Jl Sangkuriang, Gd 20, Lt 2, Bandung
Jawa Barat 40135, Indonesia
naili.huda@lipi.go.id

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Abstract
Kekasaran dan luas permukaan memainkan peranan penting dalam depositions dan reaksi katalitis pada substrat catalytic converter. Tujuan makalah ini adalah untuk menunjukkan modifikasi kekasaran dan luas permukaan substrat FeCrAl untuk catalytic converter menggunakan metode ultrasonik. Metode dilakukan dengan memproses FeCrAl dalam piranti ultrasonic cleaning bath 35 kHz dalam waktu 10 menit. Kekasaran permukaan, morfologi, dan komponen kimia dari substrat catalytic converter FeCrAl setelah proses ultrasonik dianalisis menggunakan atomic force microscope (AFM) dan diamati dengan scanning electron microscope (SEM) dikombinasikan dengan energy dispersive x-ray spectroscopy (EDS). Perlakuan ultrasonik dibantu dengan serbuk Al₂O₃ berhasil meningkatkan kekasaran dan luas permukaan FeCrAl lebih baik daripada serbuk SiC.

Kata kunci: Permukaan, Perlakuan, Kekasaran, FeCrAl, Substrat, Catalytic, Converter, Ultrasonic.

Surface roughness and area play important role especially in deposition and reaction of the catalyst in the catalytic converter substrate. The aim of this paper is to show the modification of surface roughness and area of FeCrAl substrate for catalytic converter using ultrasonic method. The method was conducted by agitating the FeCrAl in 10 minutes 35 kHz ultrasonic cleaning bath. The surface roughness, morphology, and chemical components of FeCrAl catalytic converter substrate after ultrasonic treatment were analyzed using atomic force microscope (AFM) and examined with scanning electron microscope (SEM) in combination with energy dispersive X-ray spectroscopy (EDS). The ultrasonic treatment assisted with Al₂O₃ powders successfully increased the roughness and surface area of FeCrAl better than SiC powders.

Keywords: Surface, Treatment, Roughness, FeCrAl, Substrate, Catalytic, Converter, Ultrasonic.

I. INTRODUCTION

FeCrAl has been used for fabrication of high temperatures applications components: heating elements, fire grid and honeycomb (substrate) catalytic converters for automotive exhausts. The surface area of substrate plays important role in deposition and reaction of the catalyst [1,2,3]. The honeycomb manufactured is of the efforts to increase the amount of surface area available to support the catalyst, and therefore is often called a "catalyst support" [3,4]. Other is to create washcoat, when developed on the substrate, forms a rough, irregular surface, which has a far greater surface area compared to the flat one, which then gives the converter substrate a larger surface area, and therefore more possible places for active precious metal sites [3,4,5,6]. Mellali [7] studied the influence of substrate roughness and temperature on the adhesion/cohesion of alumina coatings, when starting with a cold substrate (<100°C) the adhesion/cohesion increases almost linear with the substrate roughness. Henke [8] has used surface roughness measurements to estimate surface area on
substrates. Surfaces can be roughened or smoothened using various techniques including chemical deposition, grinding, polishing, and chemical etching [9]. The innovative approach for surface modification is ultrasonic treatment. Panin [10] studied the effect of ultrasonic treatment on mechanical behavior of titanium and steel specimens. The ultrasonic treatment used by Panin was done by exciting ultrasonic oscillations within a treating tool. The oscillation amplitude and frequency of waveguide that used were 15 µm and 24 kHz. Zhang [11] used ultrasonic horn type with the frequency and power of the ultrasound were set constant at 25 kHz and 100 W respectively, for surface treatment of magnesium hydroxide to improve its dispersion in organic phase. The transducer of ultrasound with frequency of 20 kHz was applied by Liu [12] to the aramid/epoxy composites when it was just pulled off from the resin bath to enhance its adhesion. Paniwnyk [13] has already shown that electronics materials can be surface modified using ultrasound in water medium. Paniwnyk, suggested that a rough, debris free surface, is important for optimal adhesion and that ultrasound does achieve surface modification at low temperatures of 40°C in chemical free, green and environmentally friendly deionised water. In our previous effort [14,15,16], the influence of ultrasonic assisted with SiC or Al₂O₃ powders on FeCrAl substrate to high temperature resistance of NiO catalyst development has been discussed. The ultrasonic technique successfully increased the high temperature resistance of FeCrAl substrate and homogeneity of nickel electroplating layer. However, the roughness and surface area of FeCrAl after ultrasonic treatment still needed special investigation. Therefore, in this study, the investigations of modification of surface roughness and area of FeCrAl substrate for catalytic converter by ultrasonic treatment were conducted. The discussions in this paper are focused on three following issues.

Roughness, as a measure of surface topography commonly occurs in the form of scratches, digs, pits, dust particles, polishing marks on optical surfaces, machining marks on machined surfaces, granularity or crystallites in films deposited on surfaces, undulations left by chemical etching or electropolishing, or marks left by rollers on sheet stock [8]. In this research, ultrasonic approach assisted with SiC and/or Al₂O₃ powders was expected to tune the surface roughness of FeCrAl substrate. The surface roughness was created to obtain the good quality for the next catalyst coating process on the FeCrAl substrate as explained in our previous papers [14-16]. Surface roughness was related to coating adhesion/cohesion of materials [7].

Ultrasonic wave or sound will generate cavitation bubbles in a liquid system [18]. Then, the explosion of the cavitation bubbles will produce a jet flash energy that can damage the surface of a solid material. In this work, slurring of SiC or Al₂O₃ powders in ultrasonic media liquid was expected to assist the process of surface destruction. It is estimated that the particles of the powders driven by the jet flash energy would hit the surface of the material (FeCrAl). Therefore, the irregular surface roughness occurs. Due to the higher hardness property of SiC, it was expected that ultrasonic treatment with SiC would produce higher roughness to FeCrAl.

Atomic force microscopy (AFM), is one member of a family of techniques that provides images of surface topography by mechanically moving a probe across the sample to detect the contours of the surface and enables one to detect surface morphology, nanoscale structures and molecular and atomic scale lattices [8]. Refer to literature [8], the AFM was chosen for the analysis of surface roughness and estimation of surface area of fused silica and glass substrates. This work used similar approach to calculate the surface area of FeCrAl substrate. The approach assumed that all morphology grains were nodules on FeCrAl substrate in half of sphere form. Then, the mean area taken from AFM was assumed as area of circles with diameter same as the sphere. The half of sphere surface area was then calculated using sphere surface area formula.

## II. EXPERIMENT

### A. Materials

The FeCrAl foils (Aluchrom Yhf) with 0.1 mm thickness were used as substrate in this experiment. The chemicals components of this foil are presented in Table 1.

| Ni | Cr | Fe | C  | Mn | Si  | Al | Zr | Y  | Hf | N  |
|----|----|----|----|----|-----|----|----|----|----|----|
| min | -  | 19.0 | bal | -  | -   | -  | -  | -  | -  | -  |
| max | 0.3 | 22.0 | 0.05 | 0.50 | 0.50 | 6.5 | 0.07 | 0.10 | 0.10 | 0.01 |
The substrate was prepared by cutting FeCrAl foils into 1 cm x 2 cm. The water and methanol were applied as medium for ultrasonic treatment. The SiC and Al₂O₃ powders with particle size ≤ 90 μm were provided to assist the ultrasonic treatment. These materials were supplied by Syarikat Sainfik Bersatu, Sdn. Bhd.

B. Ultrasonic treatment

The ultrasonic cleaning bath Laborette 17 was applied to the FeCrAl foils treatment. The voltage 230 V/1~, input power 2 x 240 W/period, frequency 50-60 Hz and the ultrasound frequency 35 kHz are the technical data. The bath was fully filled with water due to ultrasonic irradiation media. The specimens were put into a beaker which consists of methanol mixed with SiC and/or Al₂O₃ powders as liquid solution. The 0.2 mg/ml concentration of solution could be made by slurring 20 mg SiC or Al₂O₃ powders into 100 ml methanol. The beaker filled with slurry and FeCrAl foils then immersed into the ultrasonic bath. The FeCrAl then sonicated for 10 minutes. The illustration of ultrasonic treatment was presented in Figure 1. The specimens then dried in atmospheric condition.

C. Surface characterization

Atomic force microscope (AFM) images of the FeCrAl, FeCrAl under ultrasonic treatment with SiC, and Al₂O₃ were collected using a XE-100 Park Systems atomic force microscope (Park Systems Corp). The effect of ultrasonic on the FeCrAl substrate was examined by imaging a region on the sample before and after the ultrasonic treatment. The roughness measurement was taken using horizontal straight line mode. The roughness profiles were presented to clarify the roughness phenomenon caused by ultrasonic treatment on FeCrAl surface. The mean roughness (Ra) was resulted from a random 100 μm² scan area of specimens. The Ra of each specimen was presented as the reported roughness. The 3D images from AFM were displayed to analyze the topography of each specimen. The grain area (μm²) as 2D image was measured in AFM analysis to calculate the total surface area of specimen. The scanned surface areas of specimen were calculated by sphere surface area approach. The microstructure and chemical components on surface of FeCrAl before and after ultrasonic treatment were investigated using JEOL scanning electron microscopy (SEM) model JSM-6380LA in combination with energy dispersive X-ray spectroscopy (EDS).

III. RESULT AND DISCUSSION

A. Influence of Ultrasonic Treatment on Surface Roughness of FeCrAl Substrate

Figure 2 shows the atomic force microscope (AFM) roughness profile of the FeCrAl (a) untreated, (b) ultrasonic treatment with SiC, and (c) ultrasonic treatment with Al₂O₃. The profiles were taken by AFM random scanning from 10 μm long area of FeCrAl. From these profiles, it can be seen that the highest, medium, and lowest gap between peaks and valley occurred on FeCrAl untreated, treatment with Al₂O₃ and with SiC, respectively. The means roughness of these FeCrAl presented in Table 2.

![Figure 1. Schematic Diagram of ultrasonic treatment; (1) Methanol; (2) Specimen; (3) Beaker; (4) Water; (5) Bath; (6) Ultrasonic source.](image)

![Figure 2. Roughness profile of FeCrAl a) untreated, b) ultrasonic treatment with SiC, c) ultrasonic treatment with Al₂O₃.](image)
straight line method on random position of 10 µm x 10 µm AFM image. The most completed results were observed using the AFM 3D image. The 3D image for representing the visualization of different topography from each specimen was displayed on Figure 3.

![AFM Images](image)

**Figure 3.** 3D observation on FeCrAl a) untreated, b) ultrasonic treatment with SiC, c) ultrasonic treatment with Al₂O₃.

| Materials                          | Mean Roughness, Ra (nm) |
|------------------------------------|-------------------------|
| FeCrAl untreated                   | 31.409                  |
| FeCrAl ultrasonic treatment with SiC | 15.790                  |
| FeCrAl ultrasonic treatment with Al₂O₃ | 34.470                  |

In contrast with the expected result, according to AFM roughness test results; profile, mean roughness, and 3D topography images, the FeCrAl treatment with Al₂O₃ has the highest surface roughness compared to the others. It can be estimated that the roughness of FeCrAl resulted from treatment using ultrasonic approach, assisted with SiC and/or Al₂O₃ powders depend on the particle size and homogeneity of the powders.

**B. The Influence of Ultrasonic Treatment on FeCrAl Surface Area**

Figure 4 shows the grain area of FeCrAl which were (a) untreated, (b) ultrasonic treatment with SiC, (c) ultrasonic treatment Al₂O₃ were taken using AFM, respectively. These images resulted from 10 µm x 10 µm random scanning area of each specimen. The grain numbers on FeCrAl ultrasonic treatment with SiC was the highest, followed by FeCrAl untreated, and the lowest was FeCrAl ultrasonic treatment with Al₂O₃. Whereas the higher grain area was FeCrAl ultrasonic treatment with Al₂O₃, then FeCrAl untreated, and the smallest was FeCrAl ultrasonic treatment with SiC. The mean grain area and grain numbers from AFM examination of FeCrAl is presented in Table 3.

| Materials                          | Grain numbers | Mean area, (µm²) | Total surface area, (µm²) |
|------------------------------------|---------------|------------------|---------------------------|
| FeCrAl untreated                   | 172           | 5.412 x 10⁻¹     | 10.82 x 10⁻¹               |
| FeCrAl ultrasonic treatment with SiC | 183           | 5.058 x 10⁻¹     | 10.12 x 10⁻¹               |
| FeCrAl ultrasonic treatment with Al₂O₃ | 167           | 5.599 x 10⁻¹     | 11.20 x 10⁻¹               |
FeCrAl treatment with Al₂O₃ resulted the highest total surface area.

The surface area acts as the main role on the catalyst reaction effective. In order to accommodate the catalyst in significant amounts, substrate must be provided with a high surface area. Twigg [19] suggested that the design of substrate must provide a maximum superficial surface area that can be presented to the exhaust gas, as it is upon this surface that the catalytic coating is applied, and on which the pollutant and reactant gases must impinge in order to react.

The AFM technique results (Table 2 and 4) show that the FeCrAl ultrasonic treatment with Al₂O₃ was rougher than with SiC and the surface area followed linearly. It estimated that the roughness of FeCrAl resulted from treatment using ultrasonic approach, assisted with SiC and/or Al₂O₃ powders also depend on the particle size and homogeneity of the powders.

C. The Influence of Ultrasonic Treatment on Surface Morphology of FeCrAl and Its Chemical Composition

Figure 5 reveals SEM images and EDS results of FeCrAl substrates which were (a) untreated, (b) ultrasonic treatment with SiC, and (c) ultrasonic treatment with Al₂O₃ powders, respectively. The morphology of the FeCrAl untreated substrate forms a line texture along the roller manufacturing process direction. On the other hand, the morphology of the FeCrAl substrate treatment with Al₂O₃ and/or SiC showed many nodules, and a number of dimples were formed. The different textures of FeCrAl surface obviously describe the different surface roughness, and have been clarified using AFM in the discussion before.

The SiC grains also observable on the FeCrAl ultrasonic treatment with SiC powders (Figure 5.b) and Al₂O₃ grain on the FeCrAl ultrasonic treatment with Al₂O₃ powders (Figure 5.c). To clarify the chemical components adhered on each FeCrAl the EDS technique was implemented.

The arrow-sign at figure 5.a shows the standard FeCrAl foils chemicals component. While on FeCrAl ultrasonic treatment with SiC powders (Figure 5b), the arrow-sign indicates SiC grains. Meanwhile, on FeCrAl ultrasonic treatment with Al₂O₃ powders (Figure 5.c), the point with arrow-sign indicates the Al₂O₃ grains. It can be an evidence to the influenced of ultrasonic treatment on chemical composition of FeCrAl surface, although the percentage of each chemicals is not displayed.

The Al₂O₃ and SiC powders stick on FeCrAl after ultrasonic treatment process was a proof that

Figure 4. Grain area of FeCrAl a) untreated, b) ultrasonic treatment with SiC; c) ultrasonic treatment with Al₂O₃.
the jet flush energy of ultrasonic irradiation promoted the powders to bond on the substrate. It is estimated the amount of the adhered powders will increase with the increase of the ultrasonic treatment duration. Then if the specimen is oxidized, the $\text{Al}_2\text{O}_3$ and SiO will be formed on FeCrAl substrate. It was known that $\text{Al}_2\text{O}_3$ and SiO are include in the type of higher surface area catalyst carrier [3].

![Figure 5. Morphology and EDS of FeCrAl](image)

- (a) untreated
- (b) ultrasonic treatment with SiC
- (c) ultrasonic treatment with $\text{Al}_2\text{O}_3$
IV. CONCLUSION
The surface treatment using ultrasonic assisted with Al₂O₃ powders for 10 minutes increased the surface roughness of FeCrAl substrate better than SiC powders. The greater surface area of FeCrAl substrate was achieved using 10 minutes ultrasonic treatment assisted with Al₂O₃ powders, compared to one assisted with SiC powders. The Al₂O₃ and SiC powders were successfully attached on FeCrAl substrate after ultrasonic treatment process. These achievements will be useful for catalytic converter production using FeCrAl as a substrate.

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REFERENCES
[1]. Bruck, R., Emitec GmbH, Lohmar. "Development Status of Metal Substrate catalysts" in Material Aspects in Automotive Catalytic Converters, Bode, H., Wiley-VCH Verlag GmbH &Co. KgaA, 2002, pp. 18-30.

[2]. Nicholls, J. R., and Quadakkers, W. J. "Materials Issues Relevant to the Development of Future Metal Foil Automotive Catalytic Converters" in Material Aspects in Automotive Catalytic Converters, Hans Bode, Wiley-VCH Verlag GmbH &Co. KgaA, 2002, pp 31-48.

[3]. Heck, R. M. et al, Catalytic Air Pollution Control Commercial Technology 3rd ed, John Wiley & Sons, Inc., 2009.

[4]. Bharali, P., "Automotive Exhaust Catalysis", N. E. Quest, 3, Issue 4, pp 40-43, 2010.

[5]. Sun, H., et al, "Preparation of well-adhered γ-Al₂O₃ washcoat on metallic wire mesh monoliths by electrophoretic deposition", Applied Surface Science, 253, pp. 3303-3310, 2007.

[6]. Whu, X., et al, "Influence of an aluminized intermediate layer on the adhesion of a γ-Al₂O₃ washcoat on FeCrAl", Surface and Coatings Technology, 190, pp. 434-439, 2005.

[7]. Mellali, M., et al., "Influence of substrate roughness and temperature on the adhesion/cohesion of alumina coatings", Surface and Coatings Technology, 81, pp. 275-286, 1996.

[8]. Henke, L., et al., "An AFM determination of the effects on surface roughness caused by cleaning of fused silica and glass substrates in the process", Biosensors and Bioelectronics, 17, pp. 547-555, 2002.

[9]. Ortel, E., et al., "Influence of steel substrate roughness on morphology and mesostructure of TiO₂ porous layers produced by template-assisted dip coating", Microporous and Mesoporous Materials, 127, pp. 17–24, 2010.

[10]. Panin, A.V., et al., "The effect of ultrasonic treatment on mechanical behavior of titanium and steel specimens", Theoretical and Applied Fracture Mechanics, 41, pp. 163–172, 2004.

[11]. Zhang, F., et al., "Surface treatment of magnesium hydroxide to improve its dispersion in organic phase by the ultrasonic technique", Applied Surface Science, 253, pp. 7393–7397, 2007.

[12]. Liu, L., et al., "Ultrasonic treatment of aramid fiber surface and its effect on the interface of aramid/epoxy composites", Applied Surface Science, 254, pp. 2594–2599, 2008.

[13]. Paniwnyk, L., Cobley, A., "Ultrasonic Surface Modification of Electronics Material", Physics Procedia, 3, pp. 1103–1108, 2010.

[14]. Sebayang, D., et al., "Influence of difference deposition technique of nickel on the FeCrAl metallic monolith" in Proceedings of the Malaysian Metallurgical Conference '09 (MMC'09), UniMap Perlis Malaysia, 2009.

[15]. Sebayang, D., et al., "Effect of pretreatment using ultrasonic technique with SiC or Al₂O₃ on high temperature oxidation behavior of the FeCrAl" in Proceeding of the 14th International Conference on Applied Mechanics and Mechanical Engineering AMME-14, Egypt, 25-27 May, Military Technical College Cairo, 2010.

[16]. Sebayang, D. Et al., "NiO development on FeCrAl substrate for catalytic converter ultrasonic and nickel electroplating methods", To be published in the 2010 International Conference on Material and Manufacturing Technology (ICMMT 2010), Chongqing China, 17-19 September
2010, Advanced Materials Research Journal.

[17]. _______ "Aluchrom Yhf Material Data Sheet", (2008), No. 4049 March 2008". Germany: ThyssenKrupp VDM.

[18]. Saez, V. and Mason, T.J., "Sonoelectrochemical synthesis of nanoparticle", Molecules, pp. 1420-3049, 2009.

[19]. Twigg, M.V. and Webster, D.E. "Metal and Coated Metal Catalysts" in Structured Catalysts and Reactors 2nd ed, Cybulski, A., Moulijn, J.A. Taylor & Francis Group, USA, 2006