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

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Abstrak

Kekasaran dan luas permukaan memainkan peranan penting terutama dalam deposisi dan reaksi katalis 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 Al_2O_3 berhasil meningkatkan kekasaran dan luas permukaan FeCrAl lebih baik daripada serbuk SiC.

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

Abstract

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 Al_2O_3 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 honey-comb 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 Al_2O_3 powders on FeCrAl substrate to high temperature resistance and 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 Al_2O_3 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 Al_2O_3 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.

Table 1.
Chemical components of Aluchrom Yhf (wt-%) [17].

	Ni	Cr	Fe	C	Mn	Si	Al	Zr	Y	Hf	N
min	-	19.0	bal	-	-	-	5.5	-	-	-	-
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 Al_2O_3 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/I~, 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 Al_2O_3 powders as liquid solution. The 0.2 mg/ml concentration of solution could be made by slurring 20 mg SiC or Al_2O_3 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.

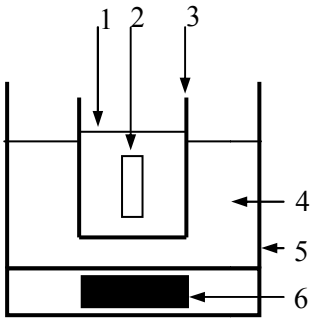


Figure 1. Schematic Diagram of ultrasonic treatment; (1) Methanol; (2) Specimen; (3) Beaker; (4) Water; (5) Bath; (6) Ultrasonic source.

C. Surface characterization

Atomic force microscope (AFM) images of the FeCrAl, FeCrAl under ultrasonic treatment with SiC, and Al_2O_3 were collected using a XE-100 Park Systems atomic force microscope (Park Systems Corp). The effect of ultrasonic on the FeCrAl substrate was examined using the result from 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 (µm^2) 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 Al_2O_3. 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 Al_2O_3 and with SiC, respectively. The means roughness of these FeCrAl presented in Table 2.

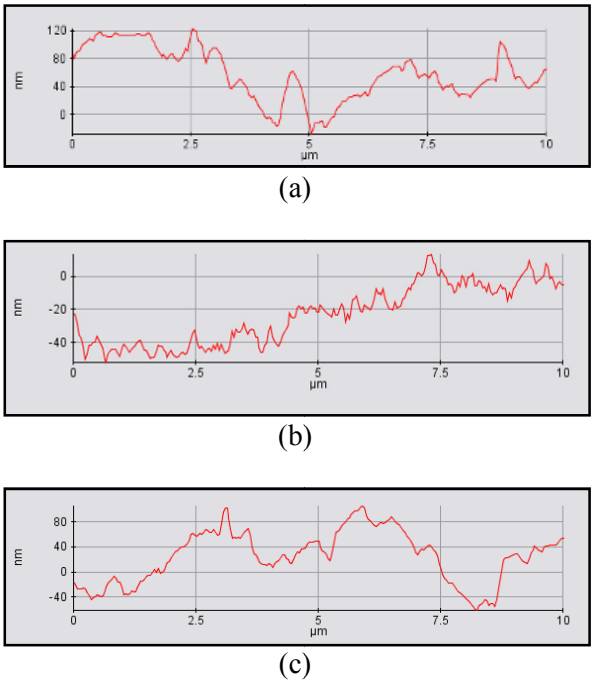


Figure 2. Roughness profile of FeCrAl a) untreated, b) ultrasonic treatment with SiC, c) ultrasonic treatment with Al_2O_3.

The mean roughness of each specimen was resulted from roughness analysis using horizontal

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.

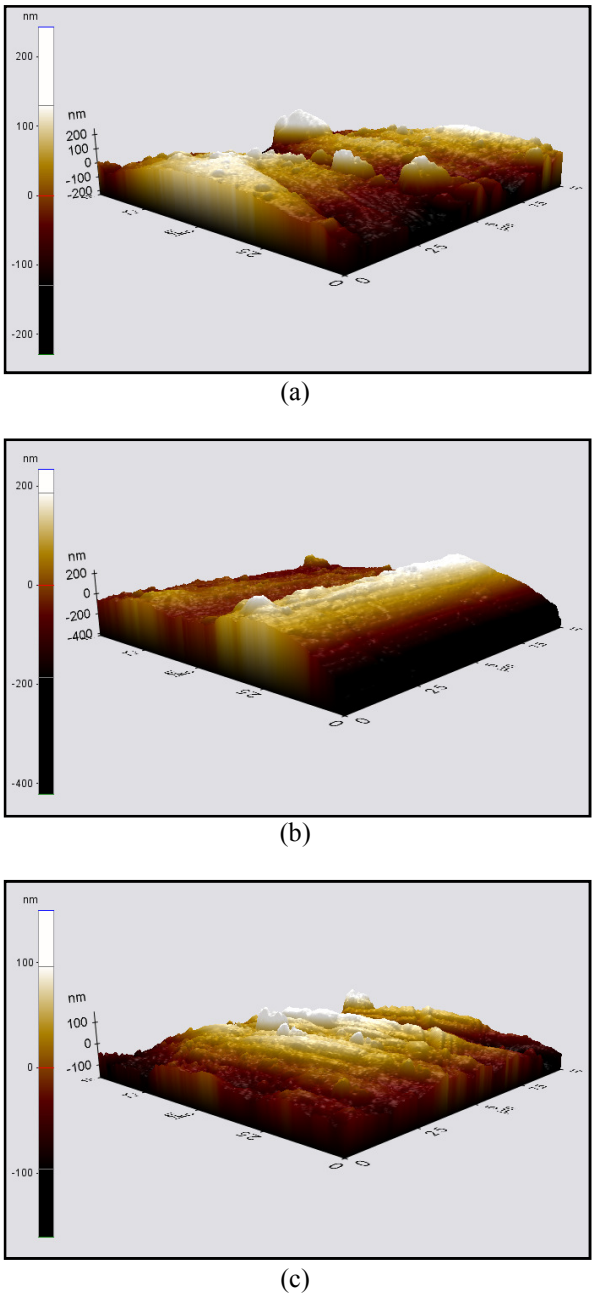


Figure 3. 3D observation on FeCrAl a) untreated, b) ultrasonic treatment with SiC, c) ultrasonic treatment with Al_2O_3.

Table 2.
Mean roughness of FeCrAl.

Materials	Mean Roughness, Ra (nm)
FeCrAl untreated	31.409
FeCrAl ultrasonic treatment with SiC	15.790
FeCrAl ultrasonic treatment with Al_2O_3	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 Al_2O_3 has the highest surface roughness compared the two others. It can be estimated that the roughness of FeCrAl resulted from treatment using ultrasonic approach, assisted with SiC and/or Al_2O_3 powders depend on the particle size and homogeneity of the powders.

B. The Influence of Ultrasonic Treatment on FeCrAl Surface Area

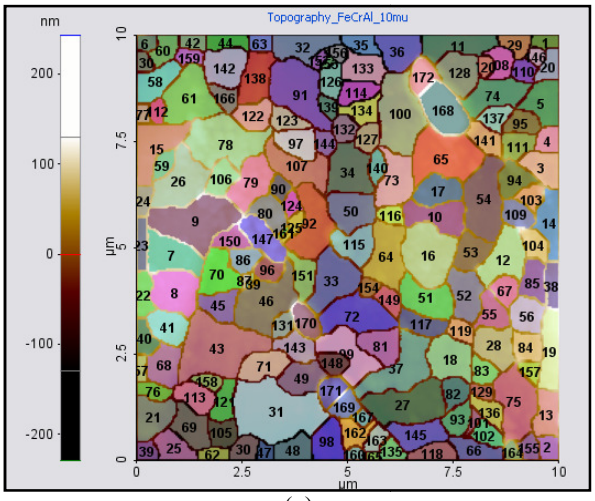
Figure 4 shows the grain area of FeCrAl which were (a) untreated, (b) ultrasonic treatment with SiC, (c) ultrasonic treatment Al_2O_3 were taken using AFM, respectively. These images resulted from 10 µm x 10 µm random scanning area of each specimen. The grain numbers on FeCrAl ultrasonic treatment with SiC was the highest, followed by FeCrAl untreated, and the lowest was FeCrAl ultrasonic treatment with Al_2O_3. Whereas the higher grain area was FeCrAl ultrasonic treatment with Al_2O_3, 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.

Table 3.
Mean of grain area of FeCrAl.

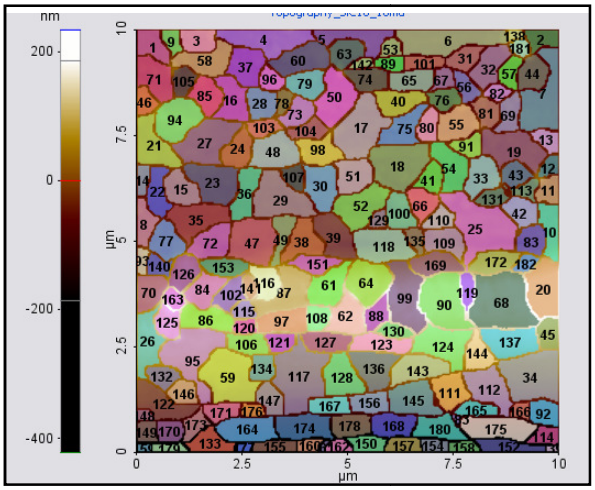
Materials	Grain numbers	Mean area, (μm^2)
FeCrAl untreated	172	5.412×10^{-1}
FeCrAl ultrasonic treatment with SiC	183	5.058×10^{-1}
FeCrAl ultrasonic treatment with Al_2O_3	167	5.599×10^{-1}

Table 4.
Surface area of FeCrAl.

Materials	Grain numbers	Mean of half nodules surface area, (μm^2)	Total surface area, (μm^2
FeCrAl untreated	172	10.82×10^{-1}	1861.73×10^{-1}
FeCrAl ultrasonic treatment with SiC	183	10.12×10^{-1}	1851.96×10^{-1}
FeCrAl ultrasonic treatment with Al_2O_3	167	11.20×10^{-1}	1870.40×10^{-1}



(a)



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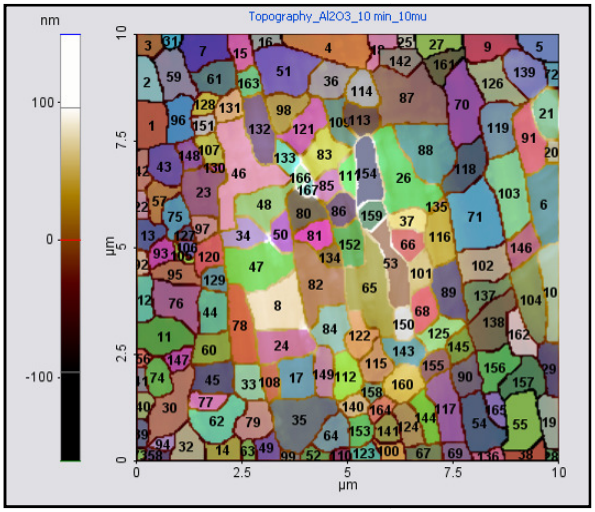

(c)

Figure 4. Grain area of FeCrAl a) untreated, b) ultrasonic treatment with SiC; c) ultrasonic treatment with Al_2O_3.

The total surface area of 10 µm x 10 µm scanning FeCrAl area obtained from a half area multiplied by grain numbers, which presented in Table 4. From the Table, it can be seen that the FeCrAl treatment with Al_2O_3 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 Al_2O_3 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 Al_2O_3 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 Al_2O_3 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 Al_2O_3 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 Al_2O_3 grain on the FeCrAl ultrasonic treatment with Al_2O_3 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 Al_2O_3 powders (Figure 5.c), the point with arrow-sign indicates the Al_2O_3 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 Al_2O_3 and SiC powders stick on FeCrAl after ultrasonic treatment process was a proof that

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 Al_2O_3 and SiO will be formed on FeCrAl substrate. It was known that Al_2O_3 and SiO are include in the type of higher surface area catalyst carrier [3].

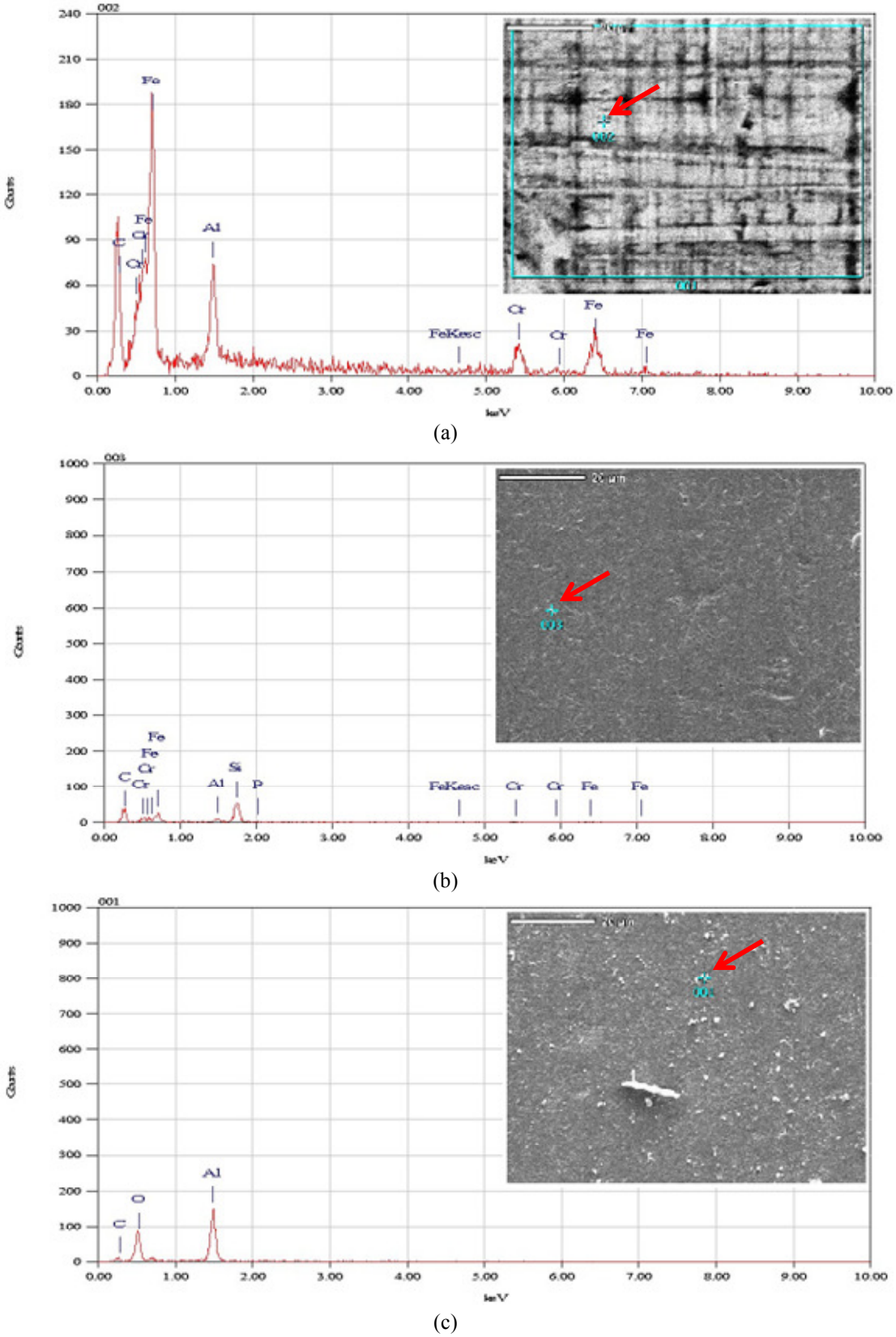

Figure 5. Morphology and EDS of FeCrAl a) untreated, b) ultrasonic treatment with SiC; c) ultrasonic treatment with Al_2O_3.

IV. CONCLUSION

The surface treatment using ultrasonic assisted with Al_2O_3 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 Al_2O_3 powders, compared to one assisted with SiC powders. The Al_2O_3 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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