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Innovative catalytic converter from FeCrAl material coated by γ-Al_2O_3 and NiCr for increasing thermal stability at high temperature operation

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Abstract

The transportation sector plays a significant role in air pollution and the greenhouse gas effect. Therefore, the innovation of exhaust system component needs to be conducted to reduce those issues. The most effective technology is by using a catalytic converter have main function is to convert the exhaust emission. The main problemfaced by the previous study is the high degradation of FeCrAl material up to 19.58 mg at 1000ºC. Therefore, the objective of this study is to develop a catalytic converter that has high thermal stability. This research was conducted by various coating processes which is Ultrasonic Bath (UB) and ultrasonic bath combined with electroplating (UB+EL) with parameters of UB consisting of frequency of 35 kHz, various holding times of 1, 1.5, 2, 2.5 and 3 hours while parameters in electroplating process are current density of 8 A/dm^2 and holding time of 15, 30, 45, 60 and 75 minutes. That process was followed by a drying process at 60ºC for 12 hours. The materials used in this research are FeCrAl as substrate, Ni as catalyst, γ-Al_2O_3 and Cr as wash-coat material. The results show that coating process coating process of γ-Al_2O_3 and NiCr on FeCrAl substrate succeeded in analysis proved by microstructure and composition analysis. The appropriate coating is shown by UB+EL 30 minute with the highest thermal stability of 2.85mg and reaction point is 0.07 mg/minute. Several compounds developed during the coating process such as in UB process develop FeCrAl, FeO, γ-Al_2O_3 and $FeCr_2O_3$ compounds and in the UB+EL process FeCrAl, FeO, γ-Al_2O_3, $FeCr_2O_3$, NiO, NaO_2, $NiAl_2O_4$ and $NiCr_2O_4$ compounds. Therefore, UB+EL 30 minutes is the most parameters that are recommended to be applied due to the lowest mass degradation and more compounds.

Keywords:
Catalytic converter, thermal stability, protection layer.

1 Introduction

According to Central Bureau of Statistics, there were 133,617,012 million motor vehicles in 2019 with an increasing rate of 7,108,236 in 2018 [1]. Therefore, it effects air quality reduction due to exhaust emission gases issued by motor vehicles and it can affect human, animal and plant health [2]. Air pollution can affect to human health such as 44% toxic emission, 50% cancer risk and 74% non-cancer risk problems [3].

The composition of exhaust emission in normal operation condition consists of 0.5% CO, 350ppm of HC, 900ppm of NO_x, 0.17% of H_2, 10% of H_2O, 10% of CO_2 and 0.5% of O_2 [4]. The most effective technology is by using a catalytic converter which has the main function is to converting exhaust emissions to green gases such as H_2O and CO_2 [5]. The catalytic converter has three components such as substrate, catalyst and cover. It is located at between the engine manifold and the muffler (Fig. 1) where the process is exhaust gas flow through the catalytic converter and during that flow, the converting process happens between the catalytic converter material and exhaust gas [6].

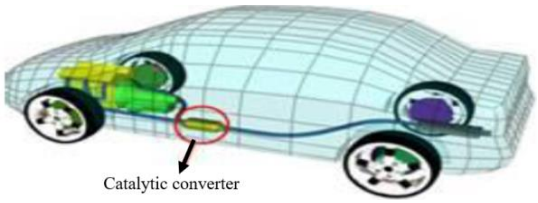

Fig. 1. Catalytic converter location.

There are 2 types of catalytic converter materials which are metallic and ceramic material where ceramic has more thickness of 0.178mm and metallic of 0.050 mm [7]. According to Masakuni and Kenichi [8], an appropriate material is FeCrAl alloy which has a chemical composition of 73% Fe, 20% Cr, 5% Al and a little Ni and Si which can operate up to 1500ºC. Metallic substrates such as FeCrAl are very interesting compared to ceramic materials [9-11] due to their higher thermal conductivity, lower heat capacity, better thermal and mechanical shock resistance and smaller pressure drops [12], higher coefficient thermal and surface area as shown in Fig. 2 [13, 14].

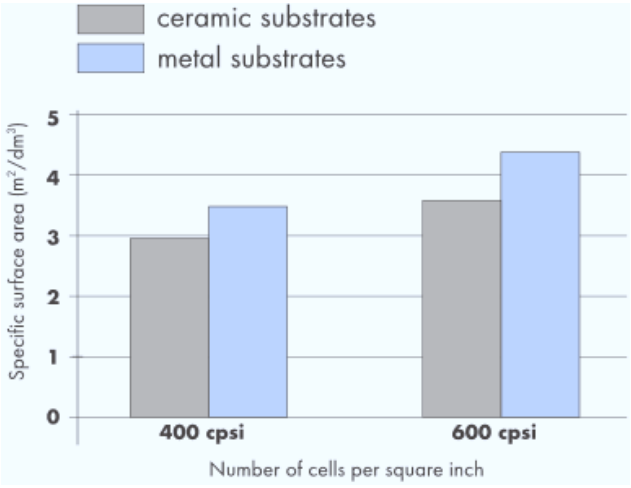

Fig. 2. Specific surface area of ceramic and metallic material [13].

Common catalyst materials are precious metals such as platinum (Pt), palladium (Pd) and rhodium (Rd), but these materials are very expensive and limited and easily break at 500-900ºC [5]. Therefore, need alternative catalysts that are more affordable such as NiO and Cr_2O_3 [5]. Apart from that, the working temperature of the catalytic converter on the intake manifold is at fairly high 300ºC-850ºC. Therefore, materials for the catalytic converter required can withstand these temperatures and above.

Various coating techniques have been used to improve the properties of catalytic converter materials such as dip coating by adding alumina to the metal [15, 16]. In addition, there are co-precipitation, sol-gel and spray-pyrolysis methods which are also applied for coating FeCrAl alloys [17, 18]. Other methods include electrophoretic deposition [19, 20], Solution Combustion Synthesis (SCS) [21, 22], aluminization techniques [23], and hydrothermal [24, 25]. However, still there are problems with the above methods when applied to coating material in powder form. Therefore, the most potential technique is the electroplating method combined with ultrasonics for coating FeCrAl substrate with powder material. Besides that, the problem that arises in previous methods is it have limitations in Ni do not diffuse directly into the substrate.

According to Putrasari et al., [13] and Feriyanto et al., [14], conduct the coating process using Al_2O_3 material on FeCrAl by ultrasonic method. They found problems with the alumina layer peeling off due to adhesion and unstable oxidation growth which led to transformation from Al_2O_3 to α-Al_2O_3. Moreover, handling pollutants from vehicle exhaust emissions is still not effective enough, this was stated by Alahmer and Aladayleh [3], Masakuni and Kenichi [8] and Shoffan et al., [26].

The novelty of this research lies in the combination of substrate, catalyst and coating materials which has not been carried out in previous research, where FeCrAl as the substrate, Ni as the catalyst, γ-Al_2O_3 and Cr as the coating. Moreover, the parameters of this research are also different from previous research in terms of holding time, current density and frequency.

The main objective of this research is to analyze the effects of ultrasonic and electroplating of γ-Al_2O_3 and NiCr on FeCrAl catalytic converters on their thermal resistance, microstructure and composition.

2 Research Method

The materials used in this research areFeCrAl with dimensions of 40 mm ×20 mm made in the United Kingdom with a chemical composition (% mass) of 53.11% Fe, 14.83% Cr, 5.29% Al and 26.77% O, γ-Al_2O_3 with the MERCK brand from Germany with 101.96 g/mol, Ni and Cr with the EMORY brand made in USA with purity of 99.96%. There are several coating methods used in this research such as ultrasonic called by UB and a combination of electroplating and ultrasonic called by UB+EL.

The electroplating process is carried out by several components such as electrolyte, anode and cathode. The electrolyte solution is called by sulphamate-type solution which consists of nickel (ii) sulphate 6-hydrate ($NiSO_4.6H_2O$), nickel (ii) chloride ($NiCl_2{\cdot}6H_2O$), boric acid (H_3BO_3), and sodium dodecyl sulfate ($C_{12}H_{25}OSO_3.Na$) under the HmbG chemicals brand. The electrolyte solution was prepared using distilled water at a constant temperature of 40±5ºC, and a pH of 5 which adjusted using HCl and NaOH and stirred using a magnetic stirrer. Nickel (Ni) and Chrom (Cr) as the anode material are cut to 50 mm ×10 mm, while FeCrAl as a cathode is cut to 40 mm × 20 mm. The distance between the anode and cathode is 25 mm.

The electroplating process was carried out with various holding times of 15, 30, 45, 60 and 75 minutes, and a current density of 8 A/dm^2, 3g γ-Al_2O_3 was inserted into the container. Then the drying process was carried out at a temperature of 60ºC for 12 hours. The electroplating process is combined with an ultrasonic method where the sample (FeCrAl, 3g γ-Al_2O_3 powder) is soaked in ethanol (20g/l) for 5 minutes, then the ultrasonic process is carried out by frequency of 35 kHz and various holding times of 1, 1.5, 2, 2.5 and 3 hours. That process was followed by a drying process that carried out at a temperature of 60ºC for 12 hours. A schematic diagram of the ultrasonic process is shown in Fig. 3.

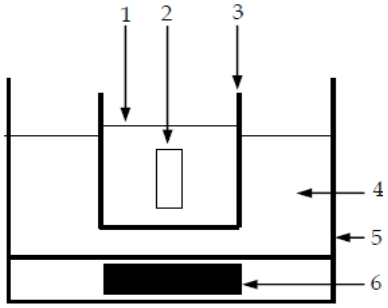

Fig. 3. Schematic diagram of ultrasonic. (1) Ethanol, (2) FeCrAl, (3) beaker, (4) water, (5) ultrasonic machine, (6) transducer.

The characterization process is conducted by several tools such as Scanning Electron Microscopy (SEM) with the SEM HITACHI brand to analyze the microstructure and chemical composition with the magnification of 1000 times on the raw material and 300 times on coated material, X-ray diffraction with the Bruker D8-Advance brand for analyzing alloys and phases that operated at diffraction angle of 20º-90º, while Thermo-gravimetric Analysis (TGA) with the LINSEIS brand, Model: L81/1550 from Germany to analyze thermal stability at 1000ºC.

3 Results and Discussion

3.1 Microstructure Analysis of UB Samples

Microstructure analysis is carried out on all samples, both raw material/substrate and coated samples using ultrasonics and a combination of ultrasonics and electroplating. Fig. 4(a) – Fig. 4(f) shows the results of the microstructure analysis of FeCrAl and coated FeCrAl by γ-Al_2O_3 powder using ultrasonics.

RawFeCrAl can be seen in Fig. 4(a) which shows a fairly even microstructure with no layers and this material has a composition of Al, Cr and Fe where the dominant composition is Fe of 74.13 wt%. The composition of the raw material shows that the material has quite high ductility with the presence of Al and Fe in the material [14]. This result is also supported by Yesuand Adhidesh [27] that when the material has a lower Cr content, it has the potential to have lower hardness and higher ductility. Samples that are coated by Ultrasonic methods is called UB sample and it is shown in Fig. 4(b) – Fig. 4(f). The figure shows that the UB samples have a more uneven surface compared to the raw material due to the γ-Al_2O_3 layer that is embedded during the coating process. This is because ultrasonics through water media produce speed bubbles that pass through the γ-Al_2O_3 and FeCrAl powder [13]. This ultrasonic process led to the changes in chemical composition where the elements C and O are present in each sample. The C and O elements have quite large values reaching 13.45 and 4.6 wt% which are shown in the UB 1 hour sample. This element has the potential effect on the protective layer forming on the base material to become FeO and $FeCr_2O_3$ [20]. The protective layer has the potential to improve the characteristics of the catalytic converter material by increasing the thermal stability up to 1000ºC and accelerating the conversion reaction from exhaust emissions in the form of CO_x, NO_x, HC to H_2O and CO_2 [26, 27].

3.2 Microstructure Analysis of UB+EL Samples

The results of the microstructure analysis of UB+EL samples can be seen in Fig. 5(a) – Fig. 5(e) with different holding times of 15, 30, 45, 60 and 75 minutes. This combination coating process is carried out simultaneously by electroplating NiCr first and followed by ultrasonic coating.

Fig. 5(a) – Fig. 5(e) shows a more uneven surface structure compared to the UB sample, accompanied by large lumps formed during the electroplating or ultrasonic coating process. These agglomerations occur due to the bonding of the γ-Al_2O_3 material that is initiated by voltage, current and frequency [14, 28]. The agglomeration that occurs in each sample is very clearly visible but there are small and large agglomerations, where the most agglomeration is shown in UB+EL 45-minute sample (Fig. 5(c)) and the most even surface structure is shown by UB+EL 30-minute sample (Fig. 5(b)).

The composition of the UB+EL sample has transformed when compared to the raw material and UB sample where there are additional elements of Na and Ni, although at low values. However, this element has the potential to produce protective layers such as NiO, NaO_2, $NiAl_2O_4$ and $NiCr_2O_4$ which significantly contribute to improving its initial characteristics. This result is supported by previous research [22] where the reaction between Ni, Cr and γ-Al_2O_3 with O and C creates a protective layer that functions as an addition to improve high thermal stability.

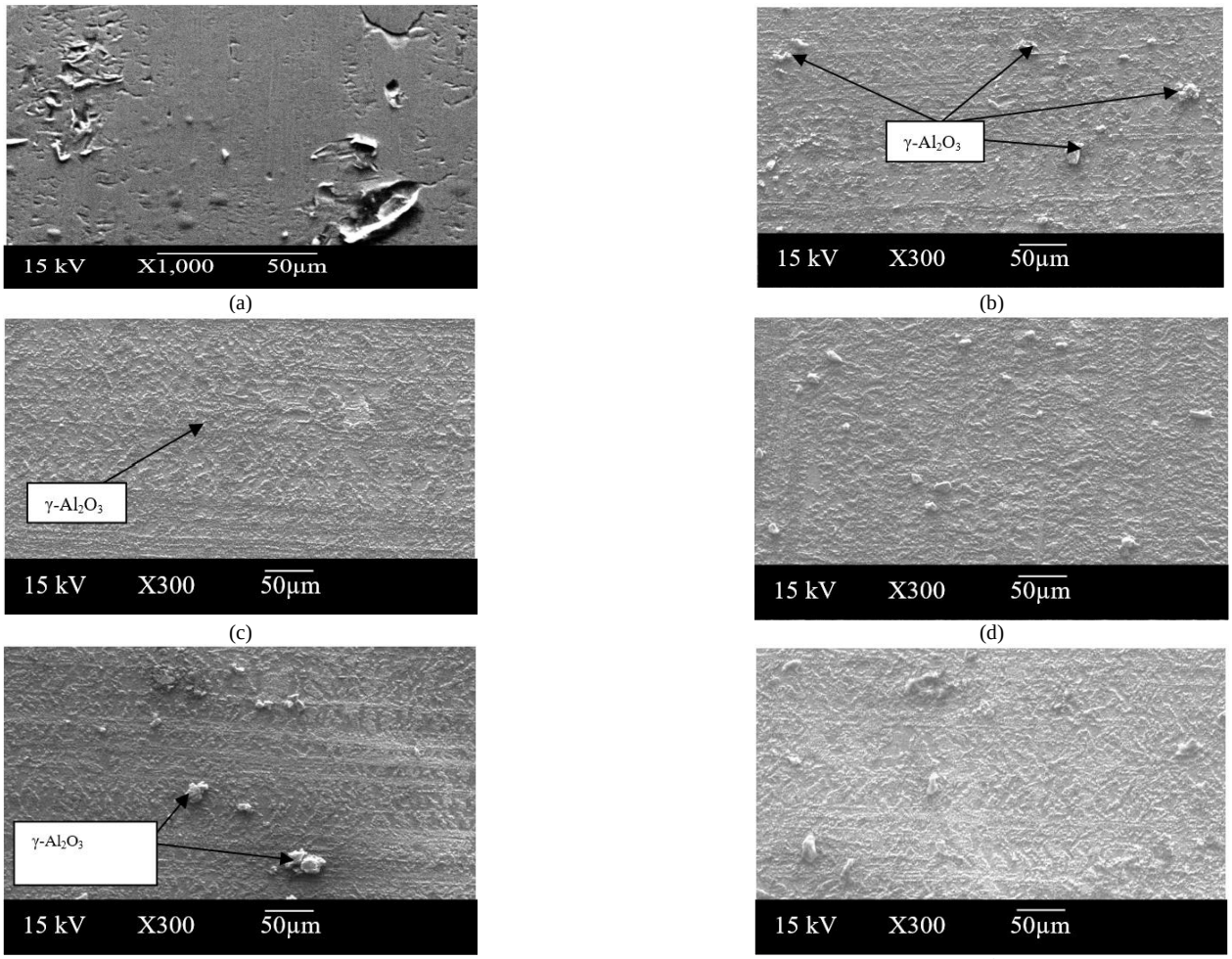

Fig. 4.Microstructure. (a) FeCrAl, (b)UB 1 hour, (c) UB 1.5 hour, (d) UB 2 hour, (e) UB 2.5 hourand(f) UB 3 hour.

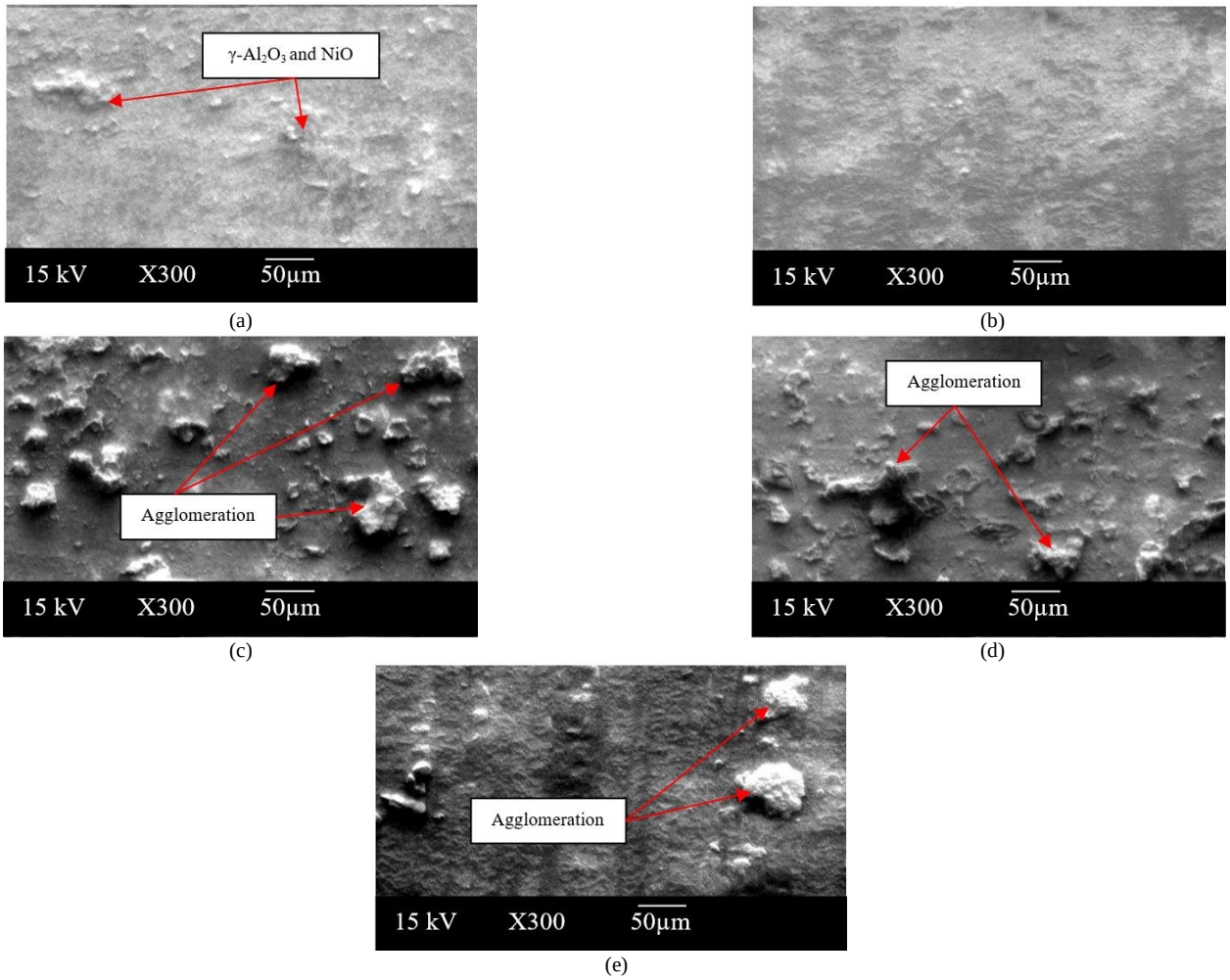

Fig. 5. Microstructure. (a) UB+EL 15min, (b) UB+EL 30 min, (c) UB+EL 45 min, (d) UB+EL 60 minand (e) UB+EL 75 min.

3.3 Thermal Stability Analysis

Thermal stability analysis has been carried out on all samples, both raw material and coated material. There are several parameters obtained such as mass changes and point of reaction as listed in Table 1. Large changes in mass indicate that the material has low thermal stability as shown by UB samples where there is a significant mass change of 20.33 mg even though the largest mass change was shown by the raw material up to 23.39 mg from the initial weight with a point of reaction reach 0.5 mg/min. The smallest mass change was shown by the UB+EL 30minute sample of 2.85mg.

Table 1. Thermal stability analysis

Sampledesignation	Temperature (°C)	Mass change (mg)	Point of reaction (mg/min)
Raw	1000	23.39	0.50
UB 1 hour	1000	20.33	0.37
UB 1.5 hour	1000	17.46	0.21
UB 2 hour	1000	17.96	0.44
UB 2.5 hour	1000	18.43	0.28
UB 3 hour	1000	18.75	0.24
UB+EL 15 min	1000	4.38	0.08
UB+EL 30 min	1000	2.85	0.07
UB+EL 45 min	1000	3.44	0.10
UB+EL 60 min	1000	3.67	0.09
UB+EL 75 min	1000	5.39	0.10

The smallest mass change of UB+EL samples during the thermal stability analysis process using TGA was caused by the protective layer that was formed. Therefore, the heat was retained by the protective layer before it hit the base material. This is very promising when applied to materials that operate at high temperatures [26, 29]. This protective layer has an alloy composition which can be seen in Fig. 6 and Fig. 7.

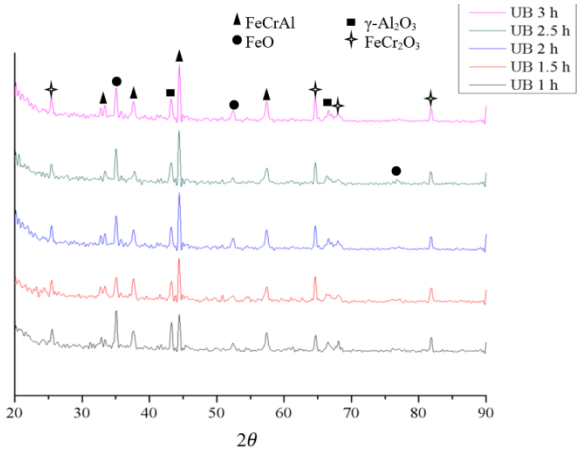

Fig. 6. Compound spectrum of FeCrAl material (UB samples).

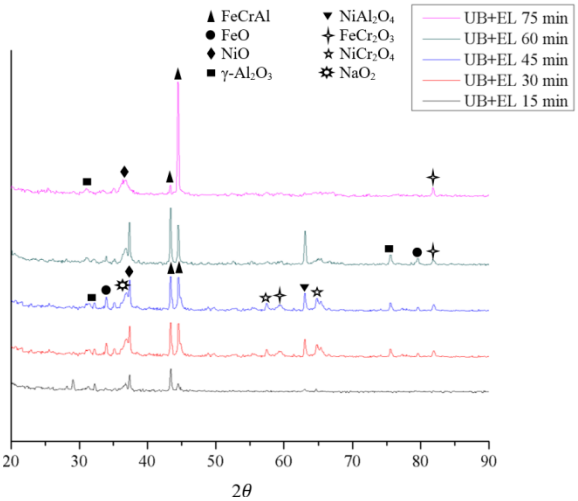

Fig. 7. Compound spectrum of FeCrAl material (UB+EL samples).

3.4 Compound Analysis

Compound analysis was carried out on the coated sample where the result of UB samples are shown in Fig. 6 while UB+EL samples is shown in Fig. 7. The XRD analysis process was carried out at a diffraction angle of 20°-90°.

The compound analysis of UB samples shows that there are several compounds detected in each peak such as FeCrAl, FeO, γ-Al_2O_3 and $FeCr_2O_3$. There are differences in the peak width of each sample, where the UB 1.5hour sample shows a very sharp peak when compared to other sample peaks. It indicated that the crystallite size is smaller compared to other UB samples [22].

The compounds detected in UB samples wereobtained from the influence of frequencies that produce high-speed bubbles that hit and coat the FeCrAl. When it correlated to the microstructure results, the UB sample has an uneven surface, it supports that the γ-Al_2O_3 material succeed embedded to FeCrAl using the ultrasonic method and this is also supported by the thermal stability analysis that the protective oxide layer was able to reduce the mass change of the FeCrAl material that applied to catalytic converters.

XRD results of UB+EL samples are shown in Fig. 7 and the results show that many compounds have the potential to improve the characteristics of the raw material such as FeCrAl, FeO, γ-Al_2O_3, $FeCr_2O_3$, NiO, NaO_2, $NiAl_2O_4$ and $NiCr_2O_4$. The spectrum shows that there is a sharp and wide spectrum, this indicates that the sharp spectrum has a smaller crystal size than the wide spectrum.

There are more compounds present in the the UB+EL sample compared to UB sample. This is possible because there are more materials involved in the process such as γ-Al_2O_3, Ni and Cr. The presence of Ni and Cr is intended to increase the mechanical properties of the material [16, 22] which led to the increment of thermal stability supported by data in TGA analysis where the 30minutes UB+EL sample has the smallest mass change.

4 Conclusion

The coating process of FeCrAl for catalytic converters using ultrasonic and electroplating processes has been successfully carried out and analyzed. In the microstructure, it was found that there was a change in the surface structure from initially flat to slightly less dense in the UB samples and very uneven as well as with many agglomerations in the UB+EL sample. The transformation of composition also occurred when the raw material only consists of 3 elements of Fe, Cr and Al, after the ultrasonic coating process the elements O and C are added. Apart from that, changes in elements also occur in the UB+EL samples where there are additional elements of Na and Ni in each sample. After carrying out a thermal stability analysis, it was found that the UB samples had a lower mash change of 4.64 mg compared to raw material and the smallest mass change of 2.85 mg or largest thermal stability was shown by the UB+EL 30minutes sample. Higher thermal stability is contributed by the protective oxide layer that formed during the coating process, where the protective layer on the UB sample is FeCrAl, FeO, γ-Al_2O_3 and $FeCr_2O_3$ while the UB+EL sample forms a higher alloy, namely FeCrAl, FeO, γ- Al_2O_3, $FeCr_2O_3$, NiO, NaO_2, $NiAl_2O_4$ and $NiCr_2O_4$. Therefore, this coating process was very recommended to be applied in order to increase the performance of catalytic converters regarding their thermal stability which has implications for increasing the lifetime of catalytic converter products.

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References

[1] Badan Pusat Statistik (BPS). JumlahKendaraan Bermotor di Indonesia [Internet], 2019 [cited 4 April 2023]. Available from:https://www.bps.go.id/indicator/17/57/1/jumlah-kendaraan-bermotor.html

[2] Badan Pengelolaan Lingkungan Hidup (BPLH), Jakarta. Pemantauan Kualitas Udara DKI Jakarta. 2020:12–24

[3] Alahmer A. Aladayleh W. Effect two grades of octane numbers on the performance, exhaust and acoustic emissions of spark ignition engine. Fuel. 2016;180:80–9

[4] TadrousM. Achieving Low Emissions from a Biogas Fuelled SI Engine Using a Catalytic Converter. Department of Mechanical and Industrial Engineering University of Toronto: Degree of Master Thesis. 2012.

[5] Bagus IRM, Purwanto P. Hadiyanto H. Optimum Design of Manganese-Coated Copper Catalytic Converter to Reduce Carbon Monoxide Emissions on Gasoline Motor. Procedia Environmental Sciences. 2015;23:86–92

[6] Kalam MA, Masjuki HH, Redzuan M, Mahlia TMI, Fuad MA, Mohibah, et al. Development and Test of A New Catalytic Converter for Natural Gas Fuelled Engine. Sadhana. 2009;34(3):467-481

[7] Kaspar J, Fornasiero P, Hickey N. Automotive Catalytic Converters: Current Status and Some Perspectives. Catalysis Today. 2003;77:419–449

[8] Masakuni O, Kenichi A. Effect of La Modification on The Stability of Coating Alumina Layer on FeCrAl Alloy Substrate. Surface & Coatings Technology. 2015;271:80–6

[9] Zeng S, Haiquan SU, Yuan L, Yan W,Dongfang W. CuO-CeO_2/Al_2O_3/FeCrAl Monolithic Catalysts Prepared by In Situ Combustion Synthesis Method for Preferential Oxidation of Carbon Monoxide. Journal of Rare Earths, 2011;29(1):69

[10] Domínguez MI, PérezA, CentenoMA, OdriozolaJA. Metallic Structured Catalysts: Influence of The Substrate on The Catalytic Activity. Applied Catalysis A: General. 2014;478:45–57

[11] Chandni R. ArthurG. Growth of Hierarchically Structured High-Surface Area Alumina on FeCr alloy ®Rods. Chinese Journal of Chemical Engineering. 2014;22:861–8

[12] Leman AM, Feriyanto D,Sebayang D. Catalytic Converter Material Treated By Ultrasonic Approach. ARPN Journal of Engineering and Applied Sciences.2016;11(11)

[13] Putrasari Y, Untoro P, Hasan S, Huda,Sebayang D. Modification of Surface Roughness and Area of FeCrAl Substrate for Catalytic Converter using Ultrasonic Treatment. Journal of Mechatronics, Electrical Power, and Vehicular Technology. 2010;01

[14] FeriyantoD, MalikSSA, FitriM, HidayatI, PranotoH. Effect of Material Composition on Thermal Stability Analysis of Coated and Uncoated FeCrAl CATCO by Î³-Al_2O_3 Ultrasonic-Electroplating Technique. Journal of Sustainable Materials Processing and Management. 2021;1(1):1-7

[15] Jia L, Shen M, Wang J. Preparation and Characterization of Dip-Coated γ-Alumina Based Ceramic Materials on FeCrAl foils. Surface & Coatings Technology. 2007;201(16-17):7159–7165

[16] Pranoto H, Feriyanto D, Zakaria S. Resistivity and Conductivity Analysis of Coated Metallic Catalytic Converter with Wavelenght Shape of Monolithresmilitaris. 2022;12(4):1759-1766

[17] Yanqing Z, Jieming X, Cuiqing L, Xin X, Guohua L. (). Influence of Preparation Method on Performance of a Metal Supported Perovskite Catalyst for Combustion of Methane. Journal of Rare Earths 2010;28(1):54-8

[18] Leman AM, Rahman F, Jajuli A, Zakaria S, Feriyanto D. Emission Treatment towards Cold Start and Back Pressure in Internal Combustion Engine against Performance of Catalytic Converter: A Review. MATEC Web of Conferences, 2017;87:02021

[19] Corni I, Ryan MP, Boccaccini AR. (). Electrophoretic Deposition: From Traditional Ceramics to Nanotechnology. Journal of the European Ceramic Society. 2008;28(7):1353–1367

[20] Feriyanto D, Zakaria S, Pranoto H, Pratiwi SE, Fitri M. Coating Thickness Analysis Of Electroplated Fecral Material For Catalytic Converter Application. Resmilitaris. 2022;12(4):1620-8

[21] Specchia A, Civera A, Saracco G. (). In-SituCombustion Synthesis of Perovskite Catalysts for Efficient and Clean Methane Premixed Metal Burner. Chemical Engineering Science. 2004;59(22-23):5091-8

[22] Feriyanto D, Abdulmalik SS, Pranoto H, Zakaria S. The Development of Fine Surface Roughness of FeCrAl Substrate by Gamma Alumina Coating Material Through Nickel Oxide Catalyst. International Journal of Advanced Technology in Mechanical, Mechatronics and Materials. 2021;2(2):70-5

[23] Wu X, Weng D, Zhao, S. & Chen W. Influence of An Aluminized Intermediate Layer on The Adhesion of a Gamma-AL_2O_3Washcoaton FeCrAl. Surface & Coatings Technology. 2005;190(2-3):434– 9

[24] Sivaiah MV, PetitS, Beaufort MF, Eyidi D, Barrault J, Batiot-Dupeyrat C, et al. Nickel Based Catalysts Derived from Hydrothermally Synthesized 1:1 and 2:1 Phyllosilicate as Precursors for Carbon Dioxide Reforming of Methane. Microporous and Mesoporous Materials. 2010;140(1-3):69-80

[25] Warju W, Sudirman RA, Ata SN, Muhammad YP. The Effectiveness of the Brass Based Catalytic Converter to Reduce Exhaust Gas Emissions from Four-stroke Motorcycle Engines. Advances in Engineering Research. 2021;209.417-22.

[26] Shoffan IAN, Sumarli S & Nauri IM. The effect of copper-based catalytic converter with circular tube shape on exhaust emission of Yamaha Vixion 1PA. IOP Conf. Ser.: Mater. Sci. Eng. 2019;669:012014

[27] YesuRamya K& AdhideshKumawat S. A mini review on the catalytic technology development for the mitigation of various air pollutants. Materials Today: Proceedings. 2023.

[28] Gedefaw AT& Nigus GH.Reactor configuration, operations and structural catalyst design in process intensification of catalytic reactors: A review. Chemical Engineering and Processing - Process Intensification. 2023;184;109290

[29] JoonHC, Jiseop H, Giyeong K, JongJC, CheolWA, JongWK, ByungDH, WoonHa Y&Yuho M. Catalyst adhesion enhancement by porous TiO_2 layer formed on anodized titanium honeycomb substrate. Ceramics International. 2021;47(5);7241-7247.