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RESEARCH PAPER

Influence of Zirconia Percent on Physical Properties of Zirconia - Aluminum Chip Matrix (Al6061) Nanocomposites

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ABSTRACT

This research intends to propose a new approach to improve the performance of aluminium composites made of chips with the addition of ZrO_2 nanoparticles. Moreover, the chip-based composite-reinforced ZrO_2 contents offer alternative sources to manufacturing automotive industries to recycle, reuse the machined materials as a secondary source of metal, and protect our earth from greenhouse gas for a sustainable life. This study focused on examining the effects of preheating time (t), preheating temperature (T), and volume fraction (VF) on the mechanical and physical properties of a ZrO_2 aluminium chips nanocomposite. This nanocomposite was produced through the hot extrusion method followed by ECAP to compare the result with heat treatment. The influence of each factor was analysed using the factorial design, followed by RSM. The microstructure and the average grain sizes of the extrudates were also investigated. Direct solid-states, such as hot extrusion and equal channel angular pressing (ECAP), are alternative and efficient solid-state processes for use in recycling aluminium scrap. These processes utilise less energy and are eco-friendly. Ceramic nanoparticles such as ZrO_2 are suggested as alternatives in the production of metal composites. This study investigated and optimised the effects of various parameters of reinforced ZrO_2 nanoparticles on the mechanical and physical properties via response surface methodology (RSM). In this study, the nanocomposites made of aluminum AA6061 chips reinforced with 5%, 10%, and 15% volume fraction of ZrO_2 powder were produced under different processing temperature of 450°C, 500°C and 550°C. The experimental results were analyzed using the design and analysis of experiments (DOE) principle, and assisted by the Minitab 18 software. It was reported that the maximum yield strength and hardness increased to 119.26 MPa and 65.25 VH compared to 100.26 MPa and 50 VH (as-received AA6061), respectively.

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INTRODUCTION

Aluminum alloy is extensively used in the engineering automotive industry because of its properties, for instance, high strength, good toughness and superior workability. High amount of aluminum consideration is focused on when the product is manufactured, and chips are discarded. The need of recycling process where

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the chips could be recycled without remelting process to avoid the disadvantages occurs through a conventional method. So, a solid-state recycling is introduced due to its low cost and ability to recycle 95 % of the materials [1,2]. In recent years, ceramic particles-reinforced recycled metal matrix composites (MMCs) have been wide investigated due to their superior mechanical and physical properties [3]. Also, processing methods have been developed for the manufacturing of aluminum metal matrix composites (AMMC). ECAP method is invented by Segal (1972), thus, the main purpose of ECAP technique is to induce high mechanical properties and enhance high strain into billets [4]. However, ECAP is an excellent SPD method for the enhancement of ultra-fine grained (UFG) microstructure based composites by imposing a large strain with the conservation of billet across dimensions [5]. ECAP is a useful and economical tool from the micro to nano-sized for the production of particle reinforced aluminum matrix composites [6]. Sanusi at el. [7] reported that AA6061 chips are reinforced ceramics based on composites to develop the ultra-fine grained material using ECAP technique. The proposed study intensively reviews the light metal recycled articles on MMCs composites, and the trends in the design of the ECAP die, and processes its parameters. Abdizadeh at el. [8] has investigated the effect combining aluminum with zircon (ZrSiO4) particles on mechanical properties of material. Compressive strength increases by increasing the zircon contents up to 248 MPa at 5% of $ZrSiO_4$ and 650°C as the optimum values before starting to decrease, due to the internal stress and density of material dislocations and results for better composite properties. Naglieri et al. [9] studied different volume fractions of zirconia particles ranging from 5 to 20 vol. % by wet chemical method at 600 °C for 1 h. the report shows a characterized homogeneity of ZrO_2 distribution in the matrix. The composites of 10 vol % ZrO_2 exhibited better hardness and the highest value of fracture threshold. The physical and mechanical properties and microstructure of products extruded using the solid-state recycling of aluminium alloy chips are dependent on the number of hot extrusion parameters [10-13]. Temperature-related parameters, the extrusion ratio, die geometry, chip morphology, and ram speed are relevant factors that need to be well regulated to obtain qualitative products from the recycling process [14,17].

In short, several investigations on ECAP processing routes concentrate on room temperature and pure metallic alloys, while limited studies reported on ECAP to AMMCs composites [18,19]. The used reinforcement particles in this study are primarily 5% vol. ZrO_2, 10% vol. ZrO_2, and 15% vol. ZrO_2 of contents, and no reports are found on Al based chips reinforced with ZrO_2 ceramics. The process objective subject to a hot ECAP at 300 °C is to avoid the material failure billet segmentation. Finally, design of experiment (DOE) was used to determine the optimum combinations of yield strength composites of mechanical properties.

MATERIALS AND METHODS

The presented matrix in this study was basically turned into machining scraps by dry milling. Dry milling was conducted to minimize complications due to the usage of coolant. The ECAP technique [20] and commercial Al6061 chips with ZrO_2 particles were mixed by a 3D mixer machine, and compacted into billet with length of 80 mm and 12 mm diameter. The weighted Al/5% vol. ZrO_2, Al/10% vol. ZrO_2, and Al/15% vol. ZrO_2 samples were treated at three types of temperature, 450 °C, 500 °C and 550 °C for 3 hours. Once the experiment was prepared, the heat supply was controlled up to 300 °C and recorded by K-type thermocouples which were connected to a data logger to avoid material failure. In contrast, the designed table was statically performed to analyse and obtain the results, the Minitab 18 software designed the

Table 1. Values of the design of experiment.

Parameters	Level used		
	Low (-)	Middle (0)	High (+)
ZrO_2 contents, %	5	10	15
Temperature,°C	450	500	550

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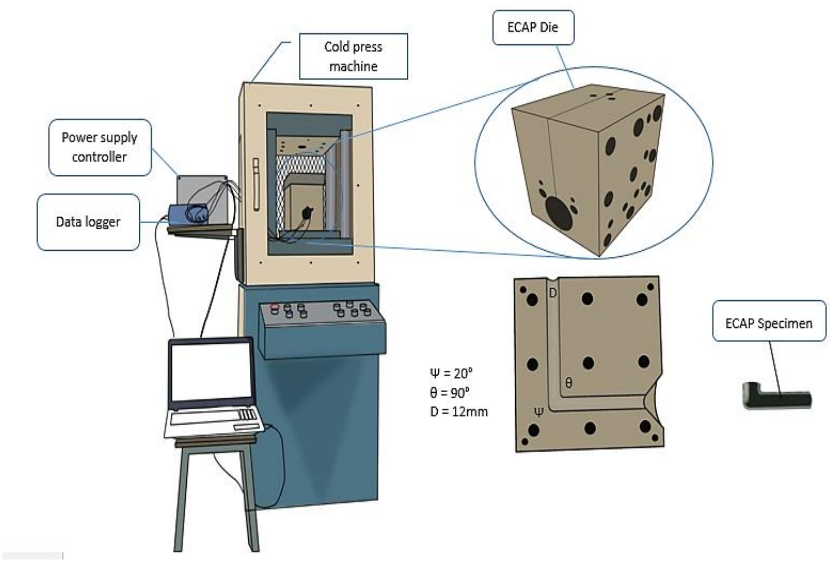

Fig. 1. Schematic Diagram for ECAP die.

experiment and investigated the yield strength of mechanical testing response. Also, the inputted values were named as temperature and volume friction of ZrO_2. Table 1 explains the designed values of the experiment.

To obtain the $Al_2O_{3/5}$ vol% ZrO_2 nanocomposite, the commercial Al_2O_3 powder AKP-53 (99.99% purity and mean particle size of 0.2 µm) produced by Sumitomo Chemical Co., Japan and ZrO_2 powder partially stabilised with 3% Y_2O_3 (99.90% purity and mean particle size of 50.0 nm) and produced by Nanostructured Materials Inc. were used. The procedure used by Santos et al. (2017) was adopted to prepare the nanocomposite [21].

The design of the experiment was conducted in this work to develop the MMC composite optimization and proposed the best overall optimum parameters such as material strength properties [22]. Fig. 1 shows the hot ECAP die schematic diagram.

RESULTS AND DISCUSSIONS

Characterization

Microstructural analysis showed that the nanometric inclusions of ZrO_2 were uniformly distributed and localized in the grain boundaries,

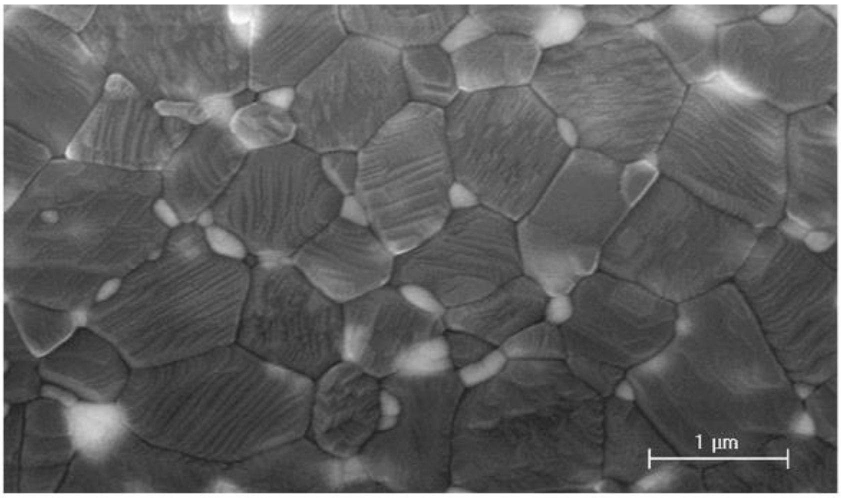

Fig. 2. SEM images of the surface of the Al_2O_3/ZrO_2 nanocomposite.

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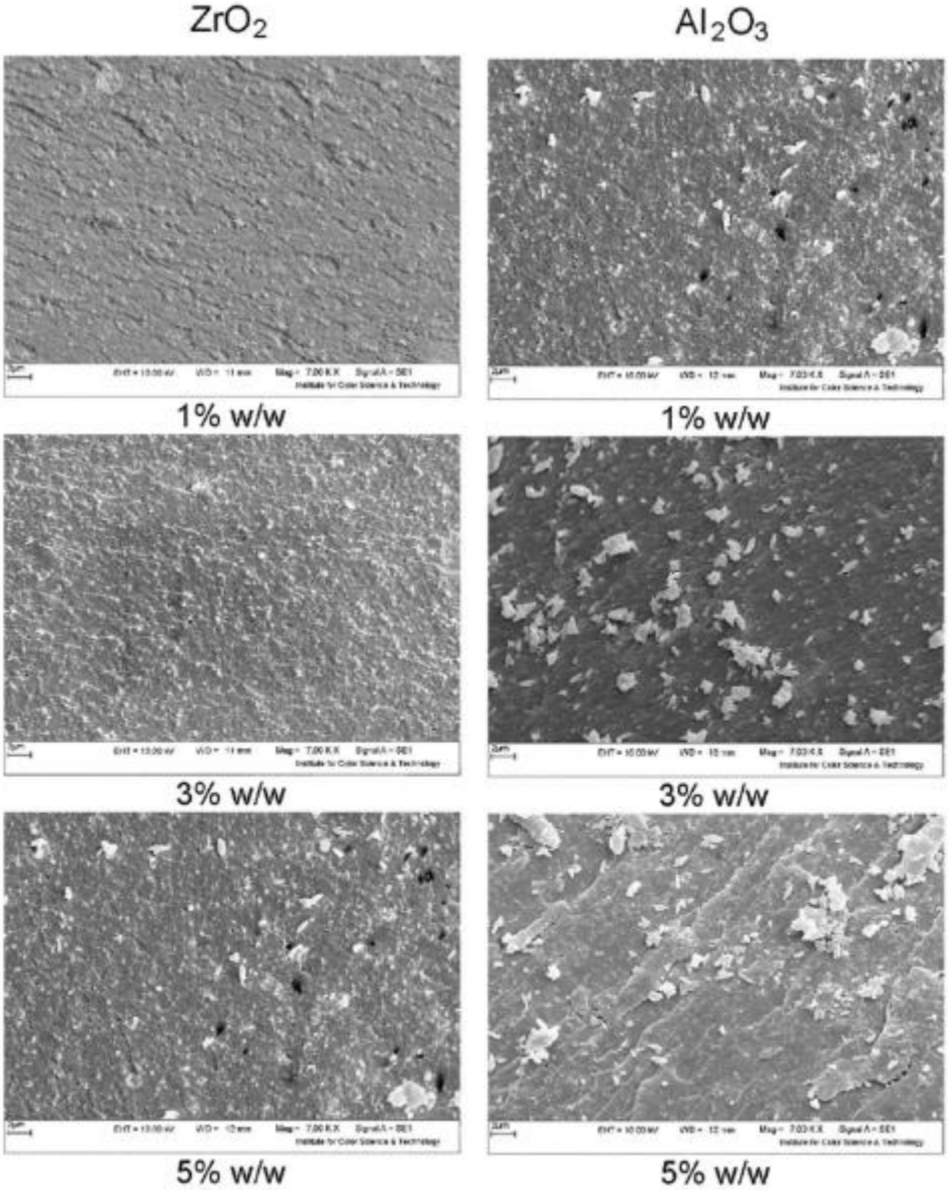

Fig. 3. SEM images of polyurethane nanocomposite coatings with different amounts of nano ZrO_2 and Al_2O_3.

even at the triple points of the Al_2O_3 matrix, as shown in Fig. 2. The inclusions of ZrO_2 at these positions favour the pinning effect in the grain boundaries of the matrix, thus inhibiting its growth.

The scanning electron microscope (SEM) images of polyurethane nanocomposite with different amounts of modified nano ZrO_2 and Al_2O_3 particles are shown in Fig. 3. In general, a good dispersion of nanoparticles in the urethane matrix could be viewed in samples with modified nano ZrO_2, which is the effect of modification on the dispersion of nanoparticles. The best dispersion with the suitable percentage of nanoparticles was observed in a sample with 3% w/w nano ZrO_2 Increasing the nanoparticles to 5% w/w results in the adhesion and aggregation of the particles. It's well known that property and response of composites depends to the amount and disperse state of nano reinforcement materials [23-26]. The addition of nanomaterial in a certain amount could improve the properties, while the excessive addition could result in the agglomeration that causes a drop in the properties. The SEM images indicate that the reason for better absorption and emissivity coefficients for 3% w/w nanoparticles is the result of a proper dispersion of nanoparticles in a polyurethane matrix. The same results were observed in the SEM images of the modified nano Al_2O_3. The dispersion in the coating is approximately the same for both particles, and the modified nanoparticles are evenly dispersed. This proper distribution is also evident in the results. However, in modified aluminum oxide nanoparticles, the aggregates of particles are higher than in zirconium oxide and larger particles are seen, which is related to the intrinsic properties of aluminum oxide and its intermolecular interactions. These gatherings are also well-distributed. As can be seen in Fig. 3, in the 5% sample of both nanoparticles, these aggregations have increased and the properties have changed. The results of absorption and emissivity coefficients shown that coating with modified nanoZrO_2 was acted better than modified nano Al_2O_3. Regardless of the differences in the nature of the particles, the microscopic images for samples with 3% w/w show that particle dispersion of modified nano ZrO_2 is better than modified nano Al_2O_3, as the agglomeration of modified nano Al_2O_3 can be seen clearly.

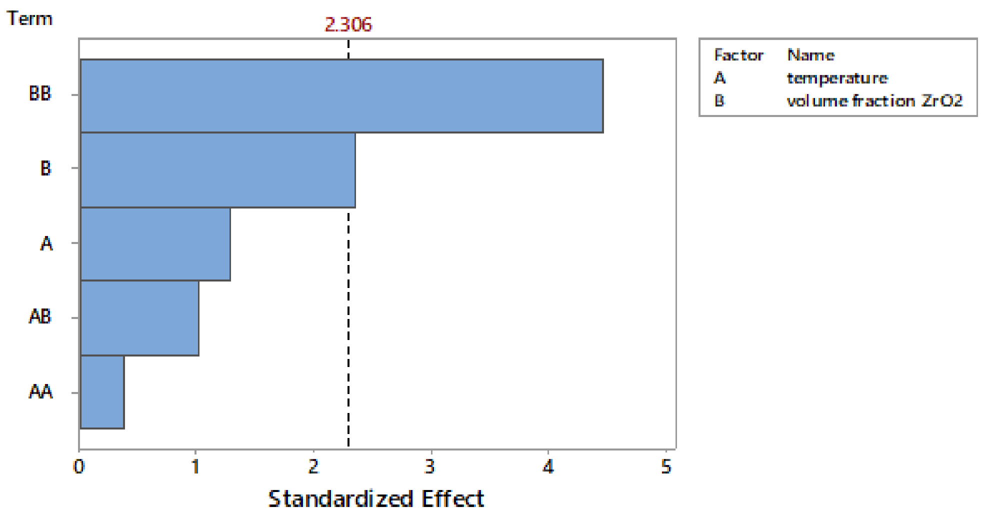

Fig. 4. Pareto Chart of Standardization Effect.

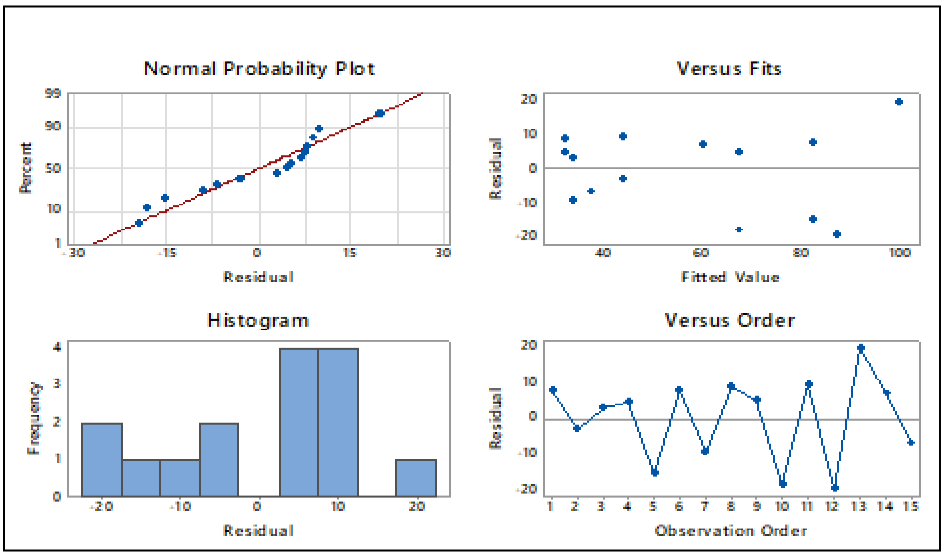

Fig. 5. Design of Experiment Al-ZrO_2.

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Table 2. Model Summary of Al-ZrO_2 Composite.

S	R-sq	R-sq(adj)	R-sq (pred)
15.3083	80.73%	66.28%	13.76%

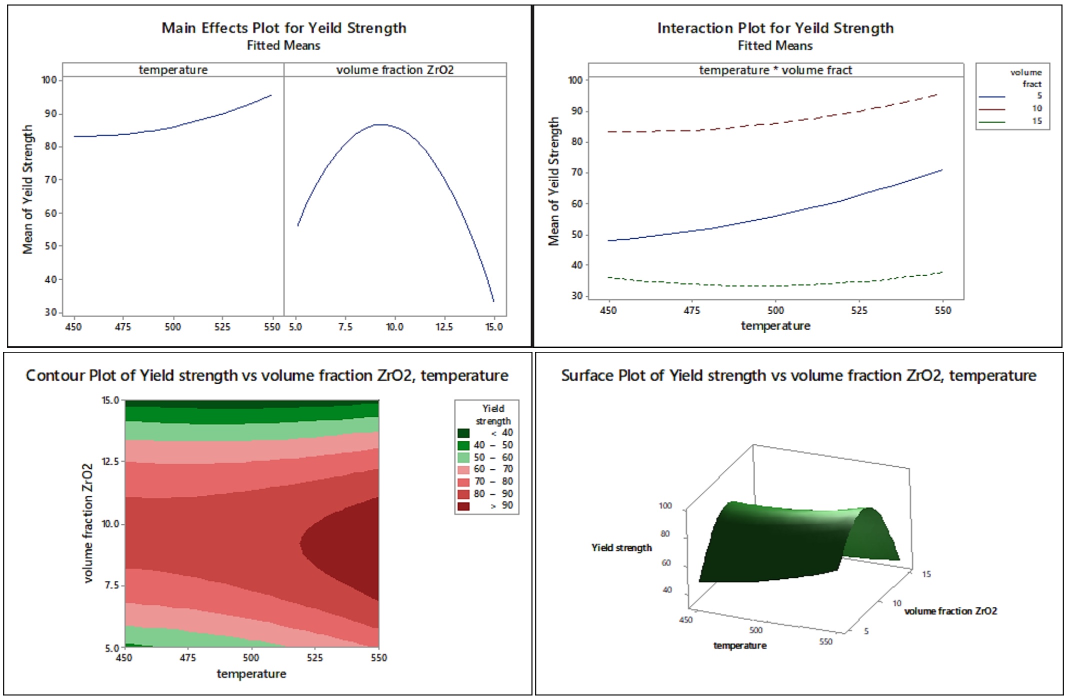

Fig. 6. RSM Data analysis Al-ZrO_2 composites.

Design of Experiment

The statistical software Minitab 18 was chosen to form the design table and analyses the experimental data. The input parameters were temperature and B4C volume fraction. The design of experiment (DOE) investigated the interactions between all parameters in order to determine the optimum values of responses. DOE factorial design of all combination of levels resulted in increasing number of experiments as the parameters increased [27]. This model was fit to be applied for linear models but could not determine the curvature points with the same number of experiments. However, two parameters at three levels were used for full factorial performance. With this type of design, 11 experiments were achieved and another extended 4 experiments were run to obtain experimental data. The effect of zircon ZrO_2 to the composite's yield strength was analysed by ANOVA, a statistical process that could summarize a set of logical conclusions based on the analysed experimental data. The ANOVA table shows the F-value measure of variation of the presented data. Also, P-value presents the lowest value of significance to the hypothesis rejection. It shows the findings of temperature and volume fraction of ZrO_2 coefficient interacted with tow process parameters, where P-value was significant to the composite yield strength. Table 2 indicated that the yield strength was 80.73% which means the model is acceptable, and the R^2 in this research achieved good fitting of the findings. According to Figs. 4-6, the data confirm the reliability and high desirability of the present model.

Yield Strength

The compressive test was done at room temperature according to the ASTM E9 – 09 standard at speed of 0.005 (m/m·min) with 5%, 10% and 15% of Al-ZrO_2 content [28]. Fig. 7 shows the results of tested composite samples with different zircon volume fraction

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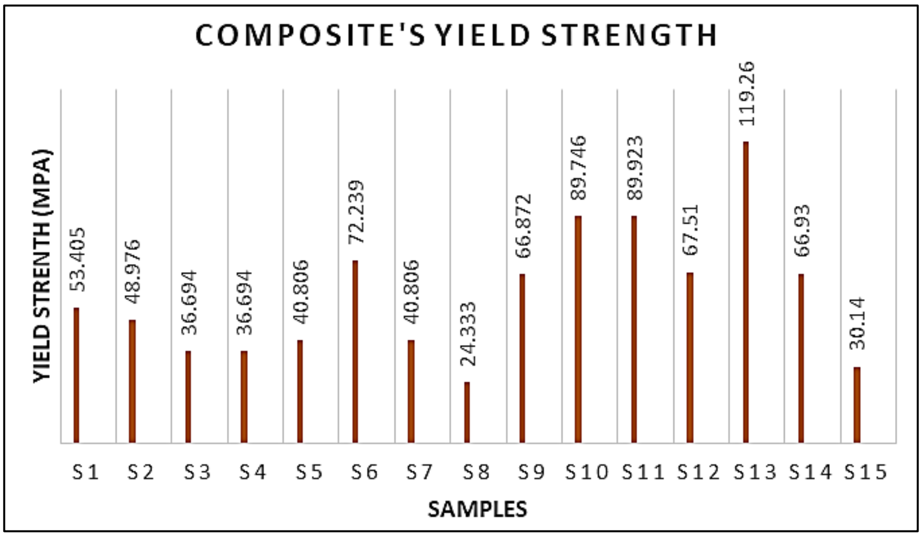

Fig. 7. Yield strength analysis.

Table 3. Design of Experiment data.

Sample notation	Std Order	Run order	V. F (B_4C) (%)	Temperature (°C)	Yield Strength (MPa)	Experimental design
S_1	1	11	5	450	53.405	
S_2	2	10	5	550	48.976	
S_3	3	4	15	450	36.694	
S_4	4	3	15	550	36.694	
S_5	5	2	5	450	40.806	Factorial design
S_6	6	9	5	550	72.239	
S_7	7	8	15	450	40.806	
S_8	8	7	15	550	24.333	
S_9	9	5	10	500	66.872	
S_{10}	10	1	10	500	89.746	Central design
S_{11}	11	6	10	500	89.923	
S_{12}	12	12	10	450	67.51	
S_{13}	13	13	10	550	119.26	Axial design
S_{14}	14	14	5	500	66.93	
S_{15}	15	15	15	500	30.14	

and processing temperature. The yield strength increased gradually by increasing the zircon contents. However, the optimum values of yield strength were 119.26 MPa at 8.94% of ZrO_2 and 550 °C, while the lowest record was 24.333 MPa at 4594.29 °C, due to the agglomeration of the particles. Increasing process temperature resulted in easier diffusion of composite's atoms and

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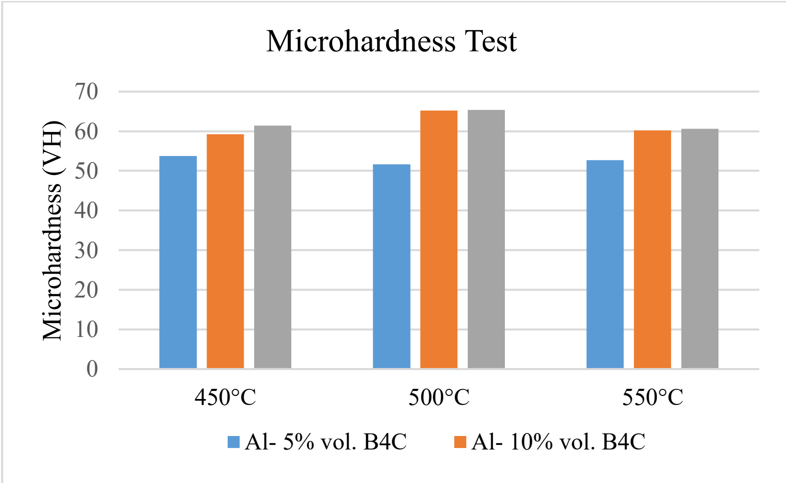

Fig. 8. Hardness of Al-ZrO_2 samples.

improved material bonding of chip boundaries. Finally, the yield strength of the MMCs reached higher value.

Hardness

The purpose of conducting hardness measurement was to test the material resistance to plastic deformation under a certain applied force [8]. The matrix was reinforced with 5% vol. of ZrO_2, 10% vol. of ZrO_2 and 15% vol. of ZrO_2 particles for all designed samples according to ASTM (ASTM E92 - 82) standard. However, Fig. 8 shows the findings of Vickers hardness measurement that confirmed the volume fraction and temperature increase to the MMCs composites. The report presented that 65.39 (VH) was the highest of all hardness values at 550°C and 10% vol. of ZrO_2, while 51.69 (VH) was the minimum reported value at 450°C and 10% vol. of ZrO_2. The calculated values were an average of five readings [3]. Also, after 10% of volume fraction, the hardness readings decreased due to material pores and particles' agglomeration.

Density

The density of Al-ZrO_2 was obtained by Archimedes' Principle. Distilled water was used as immersion liquid in this measurement. The theoretical density was calculated using the rule of mixing composites, and compared with the measured density. Table 4 indicates the findings of both theories and measurement, and it can be seen that higher temperature and volume of zircon caused increases on MMCs' overall densities due to agglomeration of ZrO_2 contents, and raised the temperature which also resulted in easier diffusion during ECAP process. From another point of view, as more zircon content particles were added, they agglomerated in one side of material region. It was also confirmed that more than 15% of ZrO_2 was not flexible to the composites, and caused weak binding between boundaries. The

Table 4. Densities and pores of Al-ZrO_2 samples.

Materials	Theoretical Density (g/cm^3)	Experimental Density (g/cm^3)
ZrO_2	5.68	-
Al6061	2.7	-
Al-5 vol.% ZrO_2	2.84	2.65
Al-10 vol.% ZrO_2	2.99	2.72
Al-15 vol.% ZrO_2	3.17	2.74

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calculated densities have been obtained by using the following formula:

$$1/\rho\_C = w\_f/\rho\_f + w\_m/\rho\_m \quad (1)$$

Where ρ is density, w volume fraction, while m, f, c is related to the composites and reinforcement [29].

CONCLUSION

Al-5 vol.% ZrO_2, Al-10 vol.% ZrO_2 and Al-15 vol.% ZrO_2 composites are successfully produced by ECAP technique under temperature of 450 °C, 500 °C and 550 °C, respectively. With increasing zirconium contents and temperature, the yield strength, hardness and density increased. The optimum reported value of yield strength is 119.26 (MPa) at 8.94% of zircon content and 550 °C, while the lowest value of yield strength is at 15% of ZrO_2 content and 494.29 °C after RSM analysis. Response Surface Methodology (RSM) is applied to validate the response validity. Hot ECAP is set up for Al-ZrO_2 composites with die angle of 90° to overcome material failure and weak bonding boundaries due to ECAP high strain hardening, and the ability to prevent plastic deformation.

No improvement is reported after 10% of ZrO_2 particles due to composite particle's agglomeration. Overall, using hot ECAP technique for solid-state recycling is highly recommended to create an environmentally friendly solution, and propose the MMCs products for engineering applications such as automotive industries.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this manuscript.

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