

Aluminium Alloys Nanostructures Produced by Accumulative Roll Bonding (ARB)

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Abstract

The accumulative roll-bonding process (ARB) is one of the severe plastic deformation methods. It aims at producing nano/ultra-fine-grained materials along with enhancements in the mechanical properties. In this work, ARB was performed on commercially cheap and available aluminium alloys in Jordan's local market; AL-2024-O and AL-1100-O alloys. Four bonding cycles were applied to promote grain refinement at room temperature with no pre/post heat treatment. In ARB processes, the thickness is reduced by 50% in each pass. A new stacking technique has been performed at the alternate layers depending on the friction of its scratched edges. After the production of samples and the investigation of mechanical properties through micro hardness test, tensile test was accomplished at room temperature after each cycle with the aim of determining whether ARB increases the mechanical properties of both aluminium alloys besides identifying the instance where material experiences high ultimate tensile strength. Information about the texture, microstructure and average crystalline size of the samples was obtained using SEM and XRD. The hardness test shows improvements for AL-2024-O/1100-O for each cycle and reported 125 HV and 80 HV respectively after four rolling cycles. The highest UTS was recorded for AL-2024-O and 1100-O on the 4th pass and reported 370 MPa and 170MPa respectively. It was also found out that the percentage elongation decreased due to a decrease in ductility after undergoing the ARB process. Moreover, after four rolling cycles, the average grain size for AL-2024/1100 decreased to 39.6 nm and 59.9 nm respectively.

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1. Introduction

In this era, the world is eager to find new metals with superior features and properties, especially in the metallurgical industry. In fact, the experts are endeavouring for new hard, strong and refractory materials, but it's really difficult to find materials that possess all these specific properties. Very recently, the experts and engineers have been able to find a new way to fabricate and develop the existing materials by using a severe plastic deformation (SPD). Over the past few decades, numerous studies have been conducted with a primary emphasis is on the development and production of alternative materials, with high strength-weight ratio, unique and distinguished mechanical properties. Aluminium is extensively utilised in products and markets. In fact, it is the most common material used in numerous applications. Aluminium alloys can offer high strength alongside simple fabrication, manufacturing and formability at a relatively low cost as compared to other metals[1].The studies have focused primarily on the production of UFG materials and improving the mechanical properties. Many SPD processes have been performed in order to produce nano/ultra-fine

grain materials such as accumulative roll bonding (ARB) where two metal sheets are scratched, chemically polished, stacked and then subjected to high strain using a roller mill for several cycles to achieve thickness reduction. Since the thickness of the sheets remain unchanged, ARB has been performed at the metal sheets for many cycles to obtain ultra-fine grain size and enhanced mechanical properties [2], [3].Using ARB is effective to enhance mechanical properties, DEGHAN and QODS cold rolled AA1050 sheets. They enhanced the U.T.S and Y.S from 69.25 MPa and 40 MPa to 201.7 MPa and 185.05 MPa respectively after 10th rolling cycle with 191% and 167% rise in tensile strength [4]. Yang wang and his colleagues investigated the microstructure and mechanical properties. of ultra-lightweight Mg-Li-Al/Al-Li composite produced by ARB at room temperature. They documented an increase in tensile strength (UTS) of 308 MPa after 4-cycles of ARB process [5]. Another investigation of the microstructure and mechanical properties of Ni/Ti/Al/Cu composite produced by (ARB) at ambient temperature showed enhancement in tensile strength after eight passes, reaching 298.2 MPa [6].Mechanical properties of nano-structured Cu/Ni multilayer fabricated by ARB revealed high ultimate tensile strength of 950 MPa which is five time higher than pure Cu

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metal [7]. It was found that applying low strain at room temperature is slightly effective to produce nano-grained metals less than 100 nm[8].

Others used SPD methods include groove pressing (CGP)[9, 10], equal channel angular pressing (ECAP) [11], friction stir processing and high pressing torsion (HPT)[12],[13],[14]. These methods have two major drawbacks. To begin with, the forming machines have large load capacities and are outfitted with expensive dies. Furthermore, productivity is extremely low, therefore the quantity of the created material is extremely constrained as compared to ARB[15],[16]. Accumulative roll bonding is a new SPD method created and developed by satio, in which a very large plastic strain is imposed in bulk process to produce ultra-fine grain metallic materials[17]. During this method, two metal sheets of comparable thickness are treated, stacked and roll bonded by a 50% thickness reduction or more during a single cycle. The process is often administered using a standard steel mill. Within the next cycles of ARB processing, the deformed bonded sheet is halved into two equal pieces, cleaned again, grinded and stacked together for the second ARB cycle and so forth. At low temperatures, ultra-fine-grained metals and alloys exhibit outstanding mechanical properties such as high strength, high toughness, and super plasticity.[18].

In this work, the accumulative roll bonding process was used in the production of bulk nanostructure alloy with a high strength-to-weight ratio of commercial aluminium for different applications in the automobile and aerospace industries. This bulk nanostructure has very high strength with a noticed decline in ductility and exhibits high values of fatigue resistance and fracture toughness [19]. Two types of aluminium alloys were used: (AA2024-O, AA1100-O). These aluminium alloys were rolled at room temperature (cold roll bonding) using a rolling mill machine. Surface polishing and cleaning process were applied on aluminium alloys before ARB. It can be noted from previous experiences that copper wires and riveting tools were used to stack aluminium sheets together, allowing a slight slipping and deviation leading to mismatch and cracking of the stacked sheets while applying a huge strain. The reason is that the copper wires or the riveting material have different mechanical properties. In this work, we were able to use a new method to stack the aluminium sheets utilising the roughness of the surface edges in order to enhance the cohesion of the sheets together, preventing the slipping during the rolling process. This new technique provided

proportional cohesion strength to the applied strain. In addition, the pre-conditioned roughened surface is an additional advantage in enhancing the bonding between the aluminium sheets. The homogeneity and the particle size for the new aluminium alloys were tested by using X-ray diffraction (XRD) and scanning electron microscope (SEM), while the strength was tested by the tensile test machine.

2. Materials and Experimental procedure

Two types of aluminium alloys were used, AA2024-O and AA 1100-O, with a geometry of $15 \times 10 \text{ cm} \pm 0.05 \text{ mm}$ and a thickness of $1 \text{ mm} \pm 0.03 \text{ mm}$ and $1.5 \text{ mm} \pm 0.05 \text{ mm}$ respectively. The chemical compositions areas shown in table 1 and table 2 were analysed by using XRF spectrometer. The main reason behind selecting AA2024-O and AA1100-O is their availability and low cost. These aluminium alloys were rolled at room temperature (cold roll bonding) up to four rolling cycles using a roller mill with 40 cm diameter and a speed of 13 rev/min fig (1/a). These specimens were abraded using a (CORYN) $115 \times 22 \text{ mm}$ in order to roughen its surface, for improving friction and bonding between the sheets as well as to remove the oxidized layer. In addition to abrading, the surface was polished and cleaned as an important part of the surface preparation using acetone due to its natural propensity to melt most oils and greases [20]. In addition, this volatile chemical solvent removes contaminants and dust and prevents the emergence of oxidation layer on the materials surface alloys before accumulative roll bonding. The roughened edges provided the ability of stacking the sheets instead of using a riveting tool and a copper wire. These roughened edges provided excellent cohesion stacking strength once huge strain is applied. Afterwards, the sheets are introduced to ARB for many cycles. Fig (1/b) shows the sheets after cut into two similar pieces. Figure (1/c) shows AL-2024 sheet thickness after four rolling cycles.

Table 1. chemical composition of Al 2024-O

material	Cu	Mg	Si	Mn	Fe	Ti	Al
Wt.%	4.4	1.5	0.5	0.6	0.5	0.20	balance

Table 2: chemical composition of Al 1100-O

Material	Fe	Si	Cu	Zn	Mn	K	Ca	Ni	AL
Wt.%	0.43	0.21	0.041	0.036	0.033	0.03	0.015	0.008	99.2

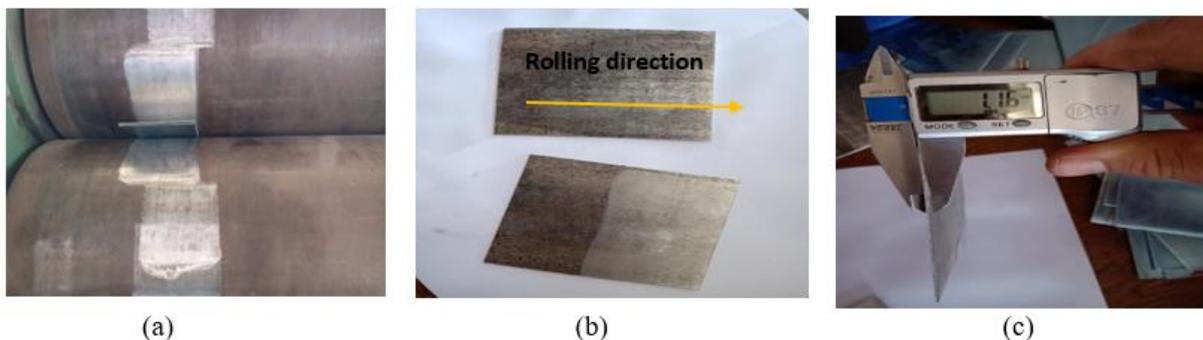


Figure 1. (a) ARB process, (b) two similar sheets after cut and (c) AL-sheet alloy after four rolling cycles.

3. Results and Discussion

Hardness test: The AA2024-O and AA 1100-O sheets have been tested for hardness by VICKERS hardness test. A load of 100 kgf (AA2024-O) and 30 kgf (AA1100-O) were used. To ensure an accurate test result, three measurements were conducted for each sample and the average hardness values are as shown in fig (4). Figure (4/a) shows the improvement in hardness for AA2024-O. After four cycles, we noticed that there is obviously increase in hardness. After one rolling cycle, the hardness improved by (70 %) and after the next cycles, the hardness improved by 8 % and 4 % respectively. The original hardness for unhardened aluminium alloy AA2024 was 60 HV which means that the hardness was doubled after four rolling cycles.

The hardness test for AA 1100-O was set on 30 kgf. The results show a slight increase in hardness through each cycle to reach 80 HV. The unhardened Al-AA1100-O was measured 48 HV as shown in figure (2). The improvement in hardness after one rolling cycle was 20 %. After the next cycles, the hardness improved by 11 %, 12.3 % and 13 % respectively.

Tensile test: Tensile test has been performed using the Shimadzu 1000 Kn Digital tensile testing machine. The original aluminium alloy AA2024 is of ultimate tensile strength (U.T.S) and yield strengths (Y.S) are 175 MPa and 70 MPa respectively. As shown in fig (3), the strength (U.T.S) changes after one cycle of cold rolling to 183 ± 2 MPa. After two rolling cycles, the U.T.S is improved by 13 %. After performing the 3rd pass, the U.T.S is dramatically increased by 9 % to record $230 \text{ MPa} \pm 2$ MPa. After four cycles of cold rolling, the U.T.S improved incredibly by 38

% to reach 370 ± 2 MPa. As seen, the cold-rolling leads to a notable change in mechanical properties of the AA2024 alloy as shown in figure (3/a). The impact of the strength of the composed layer on the bond strength of sheets has not been fully understood yet and still being investigated. [21], because the weak bonding provides an ideal environment for germination and cracks growth, fractures frequently arise in the distorted layers [22]. By increasing cold-rolling reduction, the U.T.S increases gradually, while the elongation reduces. Since the metal lost its ductility, the mechanical behaviour depends primarily on the number and distribution of dislocations introduced due to cold deformation.

ARB has proved to be an outstanding method to enhance material strength. However, low ductility is usually noticed in this method [23] often because the appearance of premature necking minimizes the required general energy to failure [1]. Figure (3) shows the improvements of tensile strength for AA1100-O. The U.T.S and Y.S before hardening were $85 \text{ MPa} \pm 2$ MPa and $35 \text{ MPa} \pm 2$ MPa respectively. After one rolling cycle, the U.T.S was improved to $120 \text{ MPa} \pm 2$ MPa, showing an improvement of 29.2 %. For second, third and four cycles, the U.T.S improved tremendously to 150 MPa, 163 MPa and 173 MPa respectively, showing an improvement of 20 %, 8 % and 6.3 % respectively. The improvement in the mechanical behaviour of the aluminium alloy can be attributed to the following reasons. First, as the rolling cycles increase, thickness decreases and grains were extremely elongated along the rolling direction, which raises the tensile strength. This can result in a higher density of dislocations and the formation of new substructures, resulting in increased in tensile strength and lower elongation.

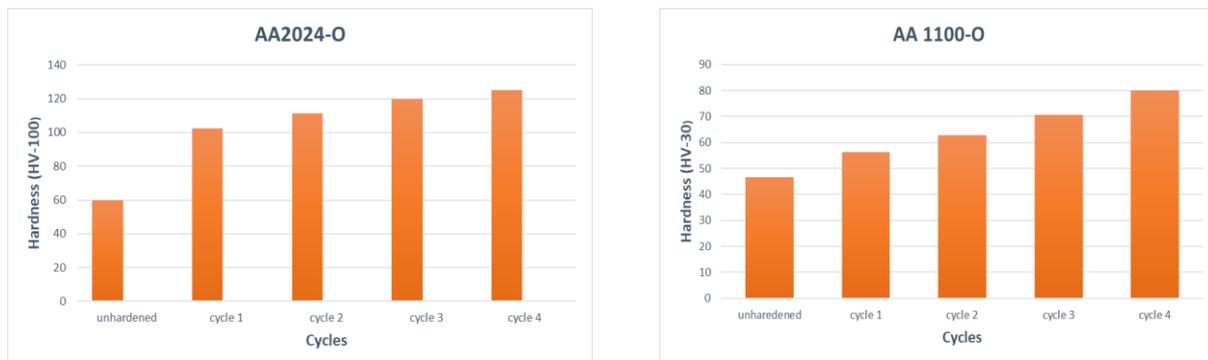


Figure 2. hardness value for AL- alloy for each cycle. (a) Hardness value for AA2024-O, (b) hardness value for AA1100-O.

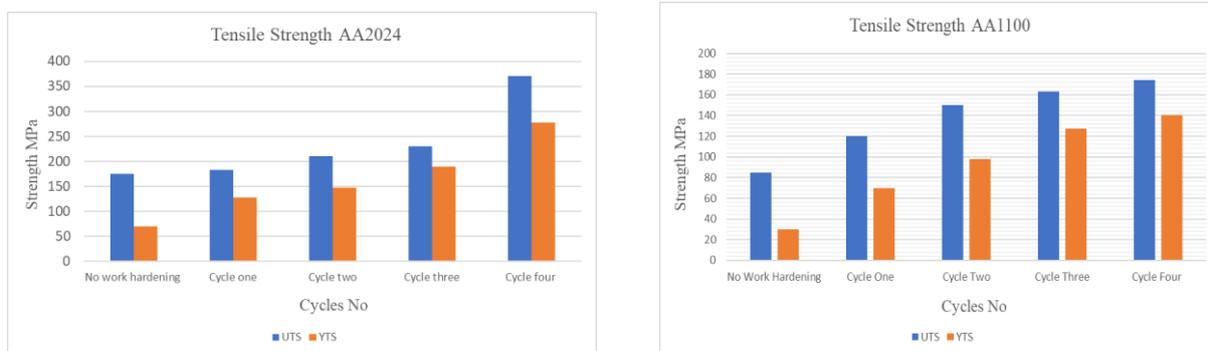


Figure 3: Tensile test for AL-alloys for each cycle. (a) tensile strength for AA2024-O, (b) tensile strength for AA1100-O

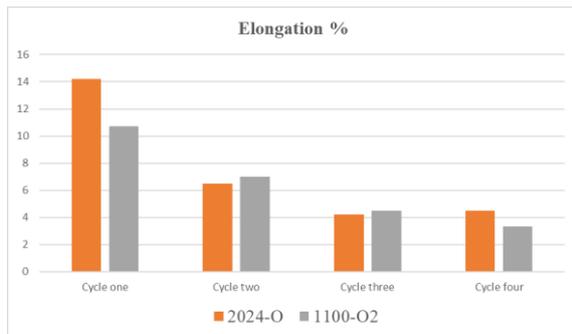


Figure 4. Elongation

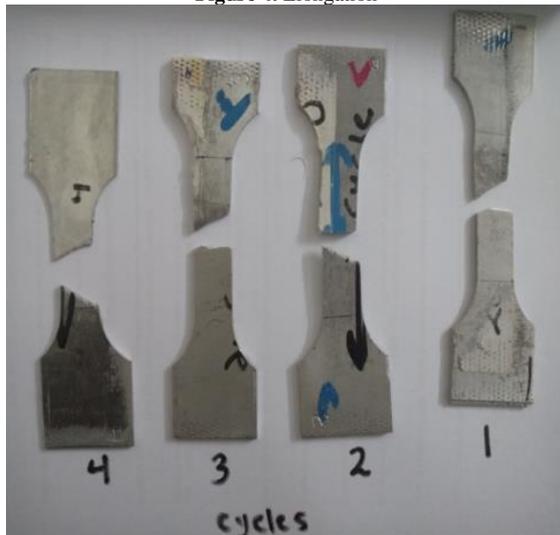


Figure 5. Dog-bone shape

Figure (4) shows a significant decrease in the elongation of both metals due to the decrease in ductility. Some results showed different values due to the possibility of the presence of cracking or slipping of the sample during testing which effect dislocations introduced due to cold deformation. The shapes of tested samples after fracture are shown in figure (5) with failure angle.

3.1. XRD & SEM

The XRD patterns of AA 2024-O and AA1100-O aluminium alloys in different cycles are shown in Fig (6) and Fig (7). Each peak represents a diffraction event from two successive crystallographic planes with a certain orientation. Peaks could be analysed with respect to the value of 2θ (X-axis) in order to calculate the value of d-spacing (the distance between two successive crystallographic planes) from which the diffraction took place. The width of the peak is an indication parameter that reflects the crystallite size, represented by a factor known as a full width at half maxim (FWHM), which is a function of any size. It is expressed by Scherrer's equation $(0.9 \times \lambda) / (d \cos\theta)$ [24]. Density of the plane means that the plane is crowded with atoms, leaving no significant spaces. The width of peaks is an indication of crystalline size. The effect of crystallite size on the diffraction pattern appears in the width of the peak. Table 3 shows the average crystalline size for AA2024/1100 for each cycle. The XRD patterns of AA 2024-O and AA1100-O samples in annealed and ARB processed conditions at different cycles are shown in Figure 6. They include typical corresponding alloy diffraction peaks. The increase in XRD peaks is an indicator of a large deformation of polycrystalline materials in plastic reign due to an increase in lattice strain. In turn, this process helps create UFG and NG structures strength. Peak broadening becomes more pronounced as the number of ARB cycles increases. In addition, the ARB rolling process produces texture in the microstructure, resulting in variation in ARB samples of the relative intensities of different planes.

The (111) plane, therefore, becomes the sharpest peak in the samples on 4 passes for AA 2024-O and 3 passes for AA1100-O samples, while the (200) plane has the highest intensity after the first ARB pass for each material.

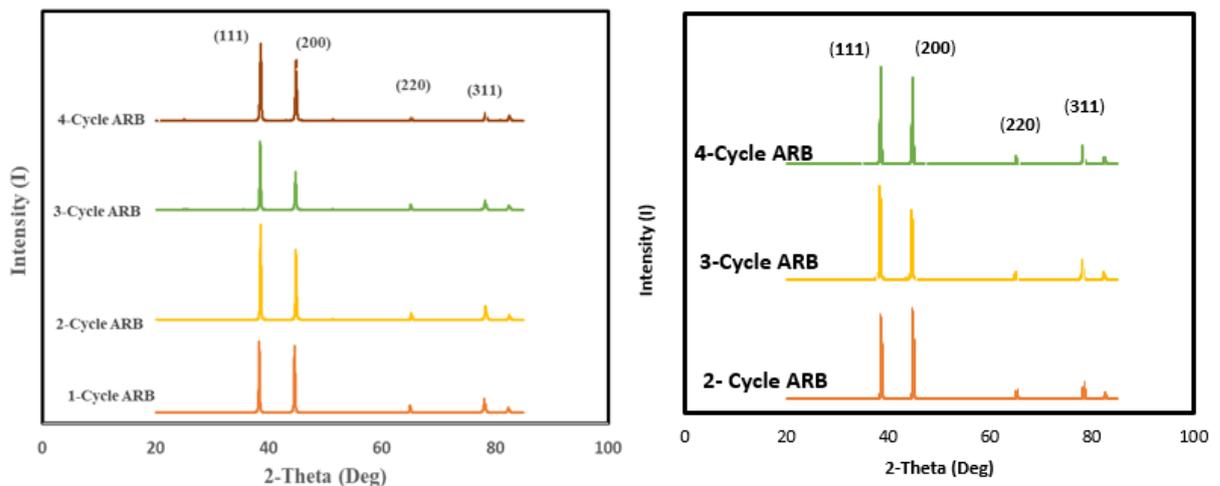


Figure 6. XRD patterns of rolled AA 2024-O alloy and AA 1100-O with 50% reduction.

Table 3. average crystalline size for AA2024 for each cycle.

#NO of rolling.	Peak position 2θ (°)	FWHM Size (°)	Dp (nm)	Dp Average (nm)
1 st pass	38.2999	0.1631	53.89	58.28
	44.5478	0.1633	54.95	
	64.9358	0.1613	61.02	
	78.0702	0.1885	56.71	
	82.2825	0.1701	64.82	
2 nd pass	38.5354	0.1533	57.38	57.34
	44.7845	0.1591	56.45	
	78.2741	0.1984	53.96	
	65.156	0.1604	61.44	
	82.4846	0.1922	57.46	
3 rd pass	38.4652	0.1797	48.94	47.73
	44.7087	0.186	48.27	
	78.1988	0.2384	44.88	
	65.0867	0.191	51.57	
	82.4097	0.2454	44.98	
4 th pass	38.5439	0.2309	38.10	39.60
	44.7918	0.2354	38.15	
	78.2703	0.2713	39.46	
	82.4667	0.2671	41.34	
	65.1659	0.2407	40.94	

Table 4. average crystalline size for AA1100 for each cycle.

#NO of rolling.	Peak position 2θ (°)	FWHM B_{size} (°)	Dp (nm)	Dp Average (nm)
1 st pass	44.8337	0.1521	59.06	60.57
	38.5873	0.1522	57.80	
	78.3141	0.1733	61.79	
	65.1965	0.1609	61.26	
	82.517	0.1755	62.94	
2 nd pass	38.3079	0.1873	46.93	53.55
	44.5615	0.1796	49.97	
	78.0878	0.1882	56.81	
	64.9463	0.1752	56.18	
	82.2971	0.1905	57.89	
3 rd pass	38.4599	0.1824	48.21	51.69
	44.7098	0.1838	48.85	
	65.0884	0.1852	53.19	
	78.2114	0.1985	53.91	
	82.4185	0.2034	54.27	

Electron Scanning Microscope (SEM): The deformed aluminium alloys AA2024-O and AA1100-O for all cycles have been tested and scanned to different scale ranges in order to study the surface and the textured topography. The figures below show SEM images for both aluminium (AA2024-O, AA1100-O) for each cycle. Figure (7) shows 10 μ m and 5 μ m of the aluminium surface. The scratch lines appear clearly and the small grain are seen blurry. After

many rolling cycles, these grains are still blurry but the X-ray diffraction equation (Scherrer's equation) $(0.9 \times \lambda) / (d \cos\theta)$ proves that the new deformed metal contains a nano-grain size.

For AA1100-O, figure (8) below shows 20 μ m and 10 μ m, where the small grain size parts can be seen in nanoscale, providing us a proof of nano grains of the material.

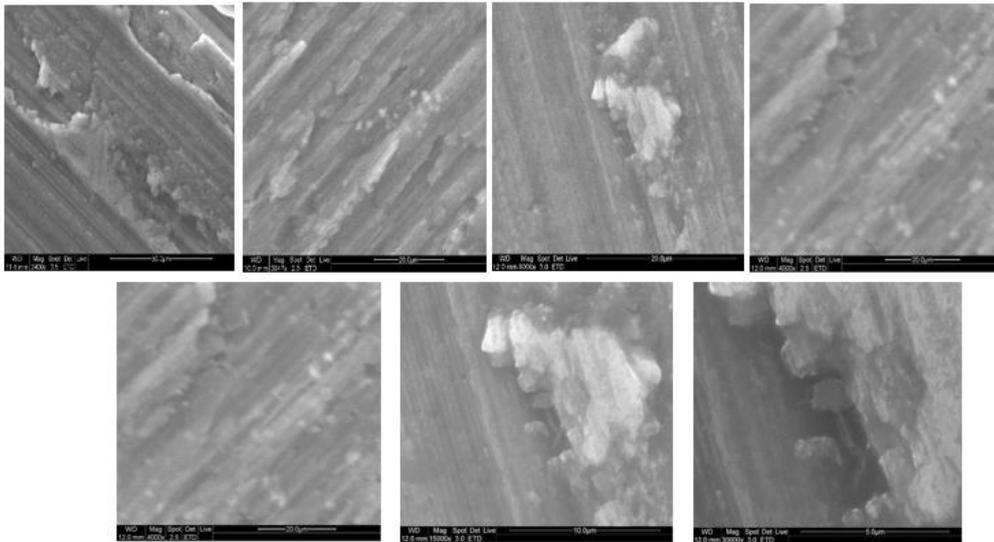


Figure 7. SEM image for Al-2024 surface.

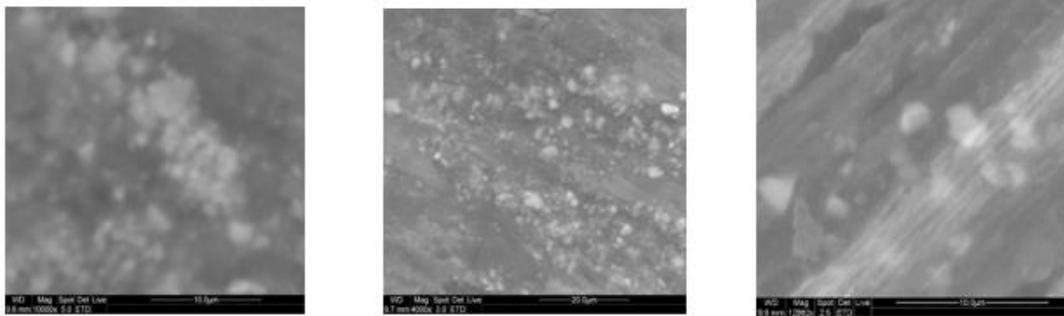


Figure 8. SEM image for AA1100-O after three and four rolling cycles.

4. Conclusion

The presented outcomes expound the high potency of the process that induces high plastic strain like ARB for AA2024-O and AA1100-O at ambient temperature in order to produce bulk nanostructure with high strength-weight ratio, durability, toughness and low ductility as compared to the conventional coarse-grained techniques. The results are visible after only one rolling cycle, making the technique appealing for practical applications. The novel stacking approach has been shown to be salutary in reducing slippage and increasing bonding time. Furthermore, the mechanical characteristics of this novel bulk nanostructure make it superior to other valuable metals such as 7070-O and 6063-O. This new distinguished material could be used widely in many applications that include, ballistic protection, the aviation and automobile industry.

By increasing the number of ARB cycles, the ultimate tensile strength for both aluminium alloys is increased. The highest ultimate tensile strength for AA2024 and AA100 is 370 MPa and 173 MPa respectively.

It is obvious that the hardness and tensile tests show extreme improvement in mechanical properties. It is also noticed that the elongation for both new nanostructure metals is decreased due to the reduction in ductility.

According to X-ray diffraction equation and (SEM) images, the average crystallite size is decreased for each single rolling cycle. For AA2024-O, the average crystallite size after four rolling cycle is 39.9 nm, while the average

crystalline size for AA2024-O after four rolling cycle is 51.69nm. The grain size has a significant influence on the mechanical properties of alloys.

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