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Abstract

In this paper, an extended accumulative roll bonding (ARB) technique, called the 'Four-Layer Accumulative Roll Bonding (FL-ARB)' technique, is presented for the first time. This technique has been employed to produce ultrafine-grained commercial pure aluminum sheets with success. After three FL-ARB passes, the grain size of pure aluminum was seen to reduce to 380 nm. The bonding strength of the sheets after rolling has also been discussed. Theoretical calculations showed that the bonding strength of sheets processed by the FL-ARB technique can be 2-2.2 times greater than that by the traditional ARB technique. The main advantages of the FL-ARB technique are (a) improvement of the interface bonding, with increasing deformation in each pass, (b) applicability of the technique at room temperature to process most metals, and (c) generation of the largest equivalent strain in the workpiece with the same number of passes, compared with other severe plastic deformation techniques.

Keywords

roll, bonding, fabrication, nanostructured, aluminum, sheets, four, layer, accumulative

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Fabrication of Nanostructured Aluminum Sheets Using Four-Layer Accumulative Roll Bonding

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Abstract

In this paper, an extended Accumulative Roll Bonding (ARB) technique, called the 'Four-Layer Accumulative Roll Bonding (FL-ARB)' technique, is presented for the first time. This technique has been employed to produce ultrafine-grained commercial pure aluminum sheets with success. After three FL-ARB passes, the grain size of pure aluminum was seen to reduce to 380 nm. The bonding strength of the sheets after rolling has also been discussed. Theoretical calculations showed that the bonding strength of sheets processed by the FL-ARB technique can be 2 ~ 2.2 times greater than that by the traditional ARB technique. The main advantages of the FL-ARB technique are (a) improvement of the interface bonding, with increasing deformation in each pass, (b)

applicability of the technique at room temperature to process most metals (c) generation of the largest equivalent strain in the workpiece with the same number of passes, compared with other severe plastic deformation techniques.

KEYWORDS: Aluminum; Bonding; Deformation; Experimentation; Formation; Manufacturing; Metalforming; Processes; Rolling

INTRODUCTION

Ultrafine-grained materials (grain size ranging from 100 nm to 1000 nm) have a lot of desirable properties: (i) high strength at room temperatures (Hall-Petch relationship); (ii) high corrosion resistance (Relationship between hardness and strength); (iii) high-speed super-plastic deformation at elevated temperatures (reduction of stress concentration due to reduced grain size). Such materials have attracted increasing attention in the past twenty years [1].

A number of Severe Plastic Deformation (SPD) techniques have been employed to fabricate nanostructured / ultrafine-grained bulk materials. These techniques include: Equal-Channel Angular Press / Extrusion (ECAP/ECAE) [2, 3], High Pressure Torsion (HPT) [4, 5], Groove Pressing (GP) [6–8], Twist Extrusion (TE) [9], Asymmetric Rolling

(AR) [10, 11], Accumulative Roll Bonding (ARB) [1, 12–14], and Accumulative Back Extrusion (ABE) [15]. The ECAP technique was employed to produce ultrafine grained AA1070 and AA6060 samples by Hockauf and Meyer [2]. They found that the pinning effect of the sheared precipitates have a great influence on refinement of the grain size of the materials. Ito and Horita [4] studied the evolution of the microstructure in pure aluminum by HPT. The grain refinement was seen to occur in the following sequence: dislocation accumulation, subgrain boundaries formation, misorientation angle enhancement, and high-angle boundary formation, before finally attaining a steady state. Sajadi et al. [6] produced aluminum samples with uniform mechanical properties by the Constrained Groove Pressing (CGP) technique. The main drawbacks of the ECAP, HPT, TE and CGP techniques are: (i) the productivity is relatively very limited; (ii) the techniques are only suitable for small samples; (iii) expensive and large load capacity dies are required. Compared to the above techniques, the ARB technique has the ability to produce continuous ultrafine grained sheets in large quantities. Saito et al [12] used the ARB technique to produce AA1100 samples with ultrafine grains (grain size about 670 nm) after 6 passes. After ten ARB cycles on AA1100, Pirgazi et al [13] found the grain size to be about 500 nm. However, the ARB technique also has a weakness: as the reduction ratio in each pass is set to 50%, the bonding quality between the two layers is difficult to control and it is difficult to manufacture the product from most metallic materials at ambient temperature. A good quality of interface bonding during cold rolling of most metals requires a reduction ratio of more than 70%, as shown in Fig. 1 [16].

In this paper, we present for the first time a newly developed ‘Four-Layer Accumulative Roll Bonding’ (FL-ARB) technique. The FL-ARB technique has been successfully used to produce ultrafine-grained pure aluminum sheets. Compared with the traditional ARB technique, the FL-ARB technique has a greater ability to produce ultrafine-grained sheets at room temperature with a good interface bonding strength.

EXPERIMENTAL INVESTIGATION

Fig. 2 is a schematic illustration of the FL-ARB technique. In the FL-ARB process, a sheet is cut into four samples of equal size, and then the four samples are neatly stacked. The interfaces between the any two adjacent sheets are surface-treated in advance for improvement of the bond strength. With a 75% reduction ratio, the four layers are bonded together through a conventional roll bonding process. The rolled product is cut into four parts lengthwise. The pieces are repeatedly surface-treated, stacked and roll-bonded. The whole process is repeated a number of times. Compared with the traditional ARB technique, the main advantages of the FL-ARB technique are: (a) an improvement of the interface bonding with a higher rolling reduction ratio in each pass, and (b) potential application at room temperature.

The FL-ARB technique was used to produce ultrafine-grained pure aluminum sheets. Commercial Aluminum 1235 alloy sheets having 0.3 mm thickness were used. The chemical composition of the material is listed in Table 1. The sheets were annealed to achieve a fully homogeneous microstructure. The FL-ARB process was carried out using a multi-function rolling mill having a 120 mm diameter roll and a maximum rolling force of 50 kN. Initially, the four sheets were stacked together and welded at one end. The composite sheet was rolled with a nominal reduction of 75% under dry conditions for each FL-ARB pass. The resulting 0.3 mm thick sheets were then cut into four parts, and repeatedly subjected to the second and third rolling passes. The micro-hardness of the samples was tested after each pass. In addition, after each FL-ARB pass, an FEI xT Nova Nanolab 200 Dual-beam workstation was used to prepare thin-foil specimens from the samples for further TEM observation. The specimens were placed on a standard carbon film Cu grid using an ex-situ lift-out method. A Philips CM200 Field Emission Gun Transmission Electron Microscope (FEG/TEM) equipped with a Bruker Energy Dispersive X-ray (EDAX) Spectroscopy system operating at an accelerating voltage of 200 kV was used to investigate the details of the microstructure.

RESULTS AND DISCUSSION

Microstructure And Mechanical Properties

Fig. 3 shows the TEM images and Vickers micro-hardness of the FL-ARB processed samples. Figures 3(a) to 3(c) are the TEM images of the FL-ARB-processed samples after the first, second and third pass respectively. As the number of rolling passes increases, the grain size of the samples gradually decreases. After the first pass, the average grain size was about 700 nm; after the second pass, the average grain size reduced to 520 nm. After the third pass, the average grain size of the sample decreased to 380 nm. In the SPD process, the grain size gradually decreases with increasing equivalent strain. Kavarana et al. [17] produced multilayer brass/steel laminates with the bilayer thickness between 39.2nm and 78.4nm with reduction ratio up to 99.97% by cold rolling of stacked sheets. However, a number of studies have shown that in ARB processing, the grain size does not continue to decrease after the sixth pass [12, 14]. In the present study, the mean thickness of layers after the third pass was seen to be 4.6μm after three FL-ARB passes. Figure 3(d) shows the micro-hardness as a function of the number of FL-ARB passes. As seen in Figure 3(d), after the first FL-ARB pass, the micro-hardness increases from 43 HV to 55 HV. However, with further passes, the micro-hardness of the samples reduces again. This phenomenon has also been observed in many studies on pure aluminum subjected to SPD processes [18, 19]. Edalati and Horita [19] gave a detailed discussion about the 'stacking fault energy' which affects the dislocation mobility and the dynamic softening that occurs more quickly to reach a steady state in Al-rich materials. However, the eventual grain size (in the steady state) is independent of the stacking fault energy [19].

Fig. 4 shows the average grain size of pure aluminum samples attained with different SPD techniques. With the ECAP technique, the average grain size of the pure aluminum reaches 660 nm, with the equivalent strain reaching 9.2 [2]. Using the ARB technique, Saito [12] and Pirgazi et al. [13] developed ultrafine-grained bulk pure aluminum with an average grain size of 670 nm and 500 nm after six and ten passes respectively. When using the GP technique, the grain size was reduced to 1000 nm after four passes for commercially pure aluminum [8]. With a semi-constrained GP process, Morattab et al. [7] obtained an average grain size of 300 nm in pure aluminum after four passes. When using the TE technique, the average grain size was reduced to about 1600 nm at the outer edge after 4 passes [9]. When using the HPT technique, the grain size at the sample edge is about 100–200 nm, while at the sample center it is 600 nm for Al 1050 when the equivalent strain is in the range 4.2 ~ 5.8. Zuo et al. [11] observed extremely fine 500 nm grains in pure aluminum at room temperature using the AR technique. Yu et al. [10] obtained samples with grain size in the range 211 nm ~ 360 nm at cryogenic temperatures. When using the FL-ARB technique, the average grain size was seen to be 380 nm. With the exception of the TE technique, all the other techniques mentioned above can produce ultrafine-grained samples with a grain size less than 1000 nm.

Table 2 shows the equivalent strain per pass for various SPD techniques. The equivalent strain increases to 0.56 after one pass of GP processing, and to 0.8 for the ARB and HPT techniques. It increases to 1.0 and 1.2 for ECAP and TE, and 1.6 for FL-ARB. It is obvious that the FL-ARB can induce the largest strain in the samples. This implies that the process requires a smaller number of passes to obtain the same total equivalent strain in the workpiece. The FL-ARB is thus seen to be a technique that could be used to produce metallic nanostructured materials.

Bond Strength Of Interface

The rolling employed in the ARB is not only a deformation process, but also a bonding process. The bonding quality of the interface is not only affected by the surface treatment, but also by the processing temperature and the deformation ratio. As the reduction ratio is set to 50% in traditional ARB processing, the rolling temperature has to be raised to improve the bonding quality. Fig. 5 shows samples produced by the traditional ARB process (a) at room temperature and (b) at 200 ºC. When the rolling is carried out at room temperature, there is no bonding at the interface between the two layers. When the samples were rolled at 200ºC, there are still some residual voids around the interface [22]. In Fig. 3 (a) – Fig. 3 (c), it is seen that there are no residual voids in the interface zone, which implies that a good quality of interfacial bonding for the aluminum alloys could be obtained through the FL-ARB technique at room temperature. Yu et al. [23] studied the interface bonding quality under different heating temperatures, reduction ratios, etc. At higher temperature, the atoms on the interface surfaces can migrate more easily, corresponding to a higher diffusion coefficient. This improves the bond quality, as shown in Fig. 5 (b). With greater reduction ratio, the deformation of the protrusions on the interfaces increases, increasing the contact surface. For a given deformation temperature, the driving force for the diffusion is unchanged. With increasing deformation, the stress gradient increases, so that the driving force for atomic diffusion at the interfaces increases. This accelerates the formation of bonds across the interfaces, as shown in Fig. 3(a) - (c).

In the present study, the FL-ARB process is carried out at room temperature. As shown in Fig. 1, the bonding strength is related to the reduction ratio during cold roll bonding. For most metals, the rolling reduction ratio should be greater than 70%. Fig. 6 shows the relationship between the reduction ratio and the number of stacked sheets in each pass. When the number of stacked sheets is 2, the reduction ratio is 50%, the bonding quality will be very weak, as shown in Fig. 5 (a). When the number of stacked sheets is 3, the reduction ratio increases to 66.6% which is still less than 70%. When the number of stacked sheets is 4 and 5, the reduction ratios increase to 75% and 80% respectively. Under these rolling conditions, the rolled sheets will have high bonding quality according to the relationship between bonding quality and reduction ratio in Fig. 1. Thus, the number of stacked layers should be 4 or more, if a good bonding quality is the only consideration. However, with increasing reduction ratio, the rolling force, rolling torsion, roll wear, etc. will increase greatly, which will greatly increase the losses in the roll mill. Thus, here we suggest that the optimum number of stacked layers is 4.

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Eqs. (1) [24], and (2) [25] have been proposed to determine the bonding strength of the interface during rolling:

$$\sigma_b = (\frac{\sqrt{3}}{2})^n \frac{\sigma_0}{K} \ln(\frac{1}{1-R_f}) \quad (1)$$

$$\sigma_b = K_1 \sigma_0 \exp(-\frac{\sqrt{3}}{2} K_2 \varepsilon_e) \quad (2)$$

where σ_b is the bond strength, σ_0 the tensile stress of sheets, R_f the final reduction ratio; ε_e the equivalent strain in the strips; n, K, K_1 and K_2 are plasticity constants depending on the sheet material and the preparation process of the welded surfaces. Equations (1) and (2) suggest that the bonding strength at the interface of multilayer metallic materials achieved by rolling is related to the reduction ratio and the equivalent strain at the interfaces. It is seen in Table 2 that the reduction ratios in the strip are 50% for ARB and 75% for FL-ARB, and the equivalent strains in the strip are 0.8 for ARB and 1.6 for FL-ARB. Using Equations (1) and (2), it is possible to evaluate the bonding strength of interface by FL-ARB and traditional ARB techniques. Using Equation (1), the bonding strength of interface by FL-ARB is seen to be 2 times of that by ARB process with the same materials. In addition, Equation (2) gives the bonding strength of the interface by FL-ARB as 2.2 times of that achieved by traditional ARB. The FL-ARB can thus be proposed as an advanced manufacturing technique that can be used to produce long and continuous ultrafine-grained sheets at room temperature. In addition, because FL-ARB does not require the samples to be pre-heated, it is a more energy-efficient technique compared to the traditional ARB technique.

CONCLUSIONS

For the first time, a new method called the 'Four-Layer Accumulative Roll Bonding (FL-ARB)' technique, has been used successfully to fabricate ultrafine-grained pure aluminum with average grain size 380 nm after three passes. For each FL-ARB rolling pass, the material was subjected to a reduction ratio of 75%.

Compared with the traditional ARB technique, the main advantages of the FL-ARB technique are: (a) improvement of the interface bonding by more than 200%, with a high equivalent strain of 1.6 in each FL-ARB pass, and (b) possibility of carrying out the technique at room temperature, without any preheating necessary before rolling. Compared to other SPD techniques (ECAP, HPT, ARB, AR, TE, and GP), the FL-ARB technique can produce the largest equivalent strain in the workpiece with the same number of passes.

Further investigations are being carried out with different materials such as other Al grades, Ti, etc. to further assess the capability of the FL-ARB technique.

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Table 1.–Chemical composition of Al 1235

Component	Si+Fe	Cu	Mn	Mg	Zn	Ti	V	Al
wt %	≤0.65	≤0.05	≤0.05	≤0.05	≤0.10	≤0.06	≤0.05	Balance

Table 2.–Equivalent strain per pass after various SPD treatments.

SPD Techniques	Equivalent strain equation	Equivalent strain per pass
GP [6]	$\varepsilon_e = \frac{N\gamma_{xy}}{\sqrt{3}}$	0.56
ARB	$\varepsilon_e = \frac{2N}{\sqrt{3}} \ln(h_0 / h_1)$	0.8
HPT [20]	$\varepsilon_e = \frac{2N\pi}{\sqrt{3}} \frac{r}{L}$	0.8
ECAP [21]	$\varepsilon_e = \frac{N}{\sqrt{3}}[2\cot(\frac{\Phi}{2} + \frac{\Psi}{2}) + \Psi \mathrm{cosec}(\frac{\Phi}{2} + \frac{\Psi}{2})]$	1.0
TE [9]	$\varepsilon_{\max} = \frac{2}{\sqrt{3}} \tan\beta$ $\varepsilon_{\min} = 0.4 + 0.1\tan\beta$	1.2
FL-ARB	$\varepsilon_e = \frac{2N}{\sqrt{3}} \ln(h_0 / h_1)$	1.6
AR	$\varepsilon_e = \frac{2}{\sqrt{3}}\{1 + [\frac{(1-r)^2}{r(2-r)} \tan\theta]^2\}^{1/2} \ln\frac{1}{1-r}$	Uncertain

Figure 1.–Bond strength vs. reduction in deformation during cold rolling.

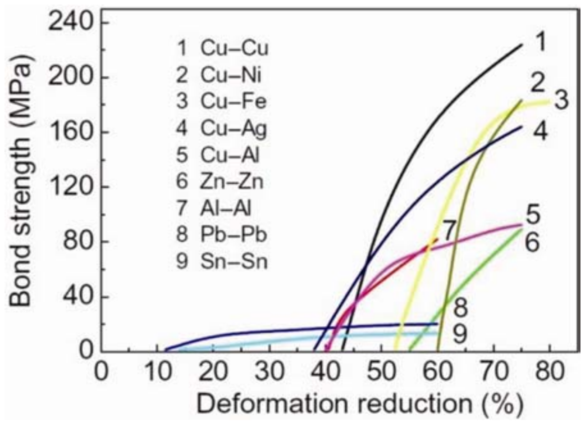

Figure 2.–Four-layer accumulative roll bonding technique.

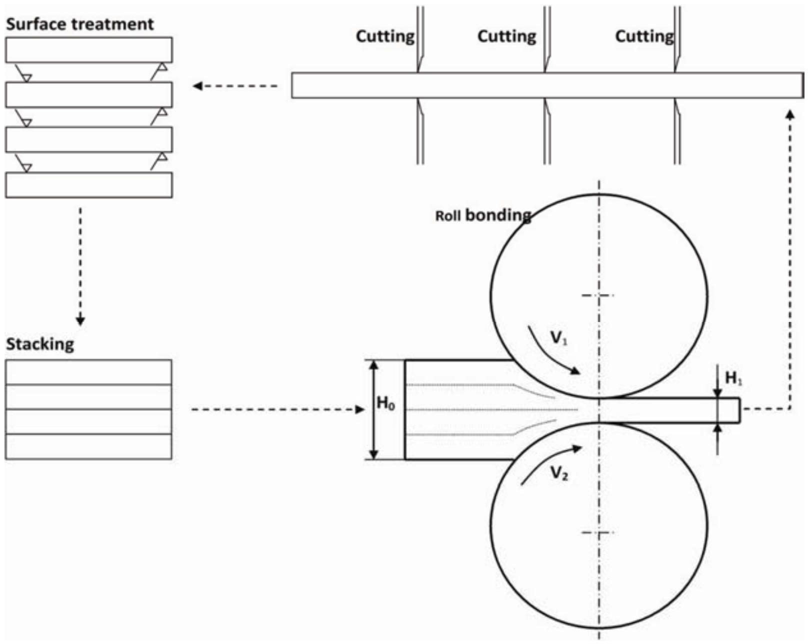

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Figure 3.–TEM images of samples after (a) first, (b) second and (c) third FL-ARB pass and (d) the Vickers micro-hardness after each pass.

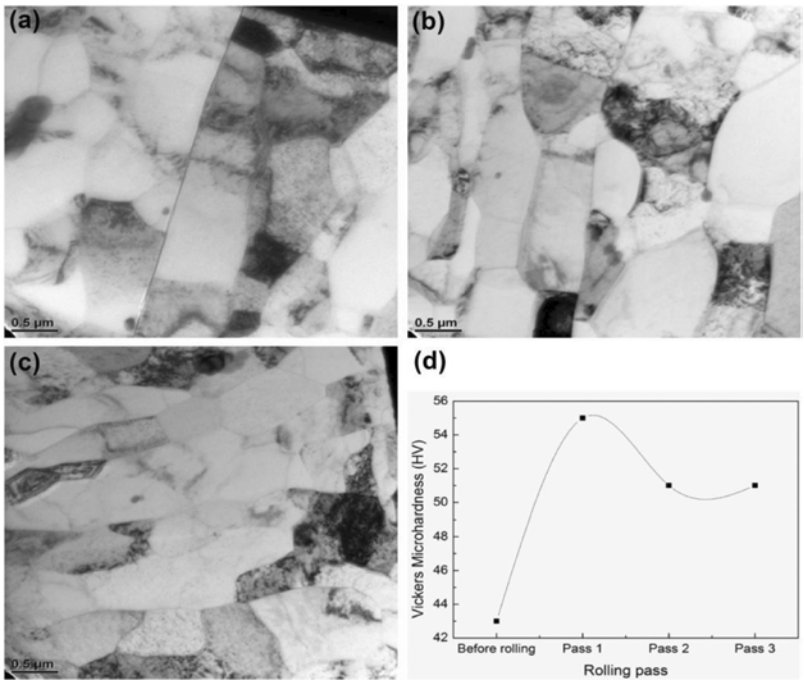

Figure 4.–Grain size of pure aluminum after various SPD techniques.

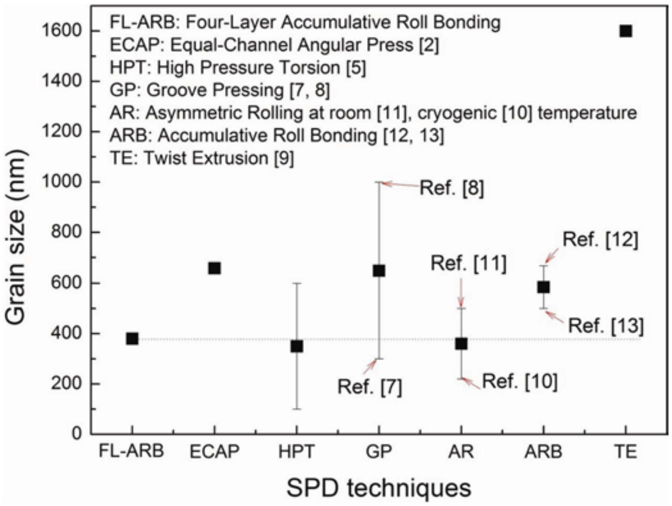

Figure 5.–Interface of samples by traditional ARB: (a) cold rolling; (b) warm rolling.

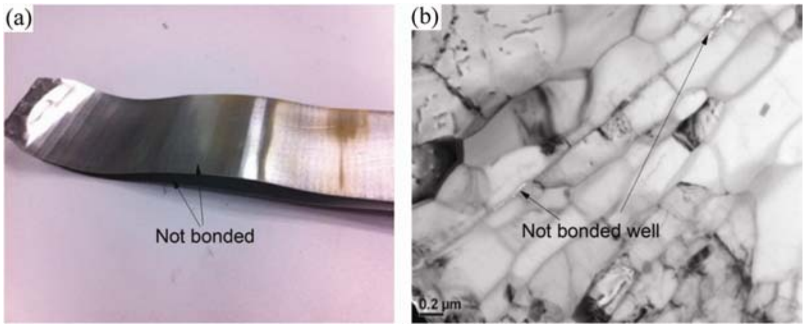

Figure 6. –Reduction ratio vs number of stacked sheets