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Fabrication of ultra-thin nanostructured bimetallic foils by Accumulative Roll Bonding and Asymmetric Rolling

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This paper reports a new technique that combines the features of Accumulative Roll Bonding (ARB) and Asymmetric Rolling (AR). This technique has been developed to enable production of ultra-thin bimetallic foils. Initially, 1.5 mm thick AA1050 and AA6061 foils were roll-bonded using ARB at 200 °C, with 50% reduction. The resulting 1.5 mm bimetallic foil was subsequently thinned to 0.04 mm through four AR passes at room temperature. The speed ratio between the upper and lower AR rolls was 1 : 1.3. The tensile strength of the bimetallic foil was seen to increase with reduction in thickness. The ductility of the foil was seen to reduce upon decreasing the foil thickness from 1.5 mm to 0.14 mm, but increase upon further reduction in thickness from 0.14 mm to 0.04 mm. The grain size was about 140 nm for the AA6061 layer and 235 nm for the AA1050 layer, after the third AR pass.

In recent years, metal laminates have become increasingly popular for engineering applications since they usually exhibit special service performance characteristics¹. Amir et al.² applied the deep drawing technique to produce bimetallic material samples. They found that the laminated deep-drawn metal sheets can be used in manufacturing parts with different ‘inner’ and ‘outer’ properties such as corrosion resistance, wear resistance and thermal and electrical conductivities. Such products are being increasingly used in various fields such as the automotive, aerospace, shipbuilding and electrical industries, and also in medical instruments. Meanwhile, micro-forming is being recognised as an emerging manufacturing process that involves the fabrication of products from ultra-thin foil/sheet material. The thickness of the foil/sheet may range from 0.001 mm to 0.3 mm. There is a great demand for micro-formed products. As the sheet thickness decreases to the same order of magnitude as the material grain size, the mechanical properties of the individual grains will dominate the properties of the sheet³. The strong anisotropy of the mechanical properties of the grains, combined with the variation in orientation of the individual grains, will lead to a strong variation in the forming properties of the sheet. Development of adequate manufacturing facilities that can supply micro-formed parts in large numbers is therefore a key factor in the successful development of this process^{4,5}. Thus, it is very interesting to investigate the mechanical properties of ultra-thin nanostructured bimetallic foils and develop technologies for their potential application in micro-forming.

The special properties of nanostructured materials have been attracting a great deal of attention. Bulk fabrication of metallic materials using the Severe Plastic Deformation (SPD) technique has probably brought us closer to enabling use of nano-materials for structural and functional applications⁶. Variants of the SPD technique, such as High-Pressure Torsion (HPT)^{7,8}, Equal Channel Angular Pressing (ECAP)^{9,10}, Asymmetric Cryorolling (ACR)^{11,12}, Accumulative Roll Bonding (ARB)^{13,14}, Asymmetric Rolling (AR)^{15,16}, etc have been developed to enable bulk fabrication of nanostructure or ultrafine grain samples of different metals. Among these, the HPT and ECAP techniques are more suitable for small samples, so that the material quantities which can typically be produced are rather limited. ACR, ARB, and AR are techniques that have potential application in industries to produce continuous products. Yu et al have employed the ACR technique to produce nanostructured Al 1050¹¹ and Al 6061 alloys¹². The grain sizes were seen to be 211 nm for Al 1050 and 250 nm for Al 6061 alloy.

Compared with the above methods, the ARB and AR techniques are more suited to the production of multi-layer materials. It has been shown that the ARB technique can be successfully used to produce multi-component

materials with tailored properties by reinforcement or grading. This allows optimizing the properties of the product based on those of two or more alloys¹⁷. The ARB technique has been used to fabricate products with composite layers of materials, such as Al/Cu/Ni¹⁸, Al/Mg¹⁹, and different series of aluminium alloys^{20–23}. Roy et al.^{20,21} used the ARB technique to produce AA2219/AA5086 laminates. They reported that a strong bond between adjacent layers, accompanied by a substantial grain refinement, was achieved after 8 ARB passes. Saito et al.²² used the ARB technique to produce AA1100/AA5083 samples with ultrafine grains and 300 MPa tensile strength after 6 passes. Quadir et al.²³ used the ARB technique to generate sheet material consisting of alternating layers of Al and Al-Sc. They found that boundary pinning by nano-size Al₃Sc particles slows the rate of grain coarsening in the Al-Sc layers and that these particles also severely impede the growth of grains from the Al layers into Al-Sc layers. Su et al.²⁴ used the ARB technique to produce ultrafine grained composite AA1050/AA6061 sheets, and found severe shear bands throughout the cross-section of the 5-cycle ARB processed composite. However, to date, there have been no reports on the production of ultra-thin bimetallic foils using the ARB method coupled with the AR technique.

In the AR process, sheets are passed between rolls that either have different diameters, or rotate at different angular speeds. AR has significant potential for industrial applications because it requires a lower rolling pressure and torque, making it more suitable to produce thin sheets or foils. The AR technique involves rolling with an imposed shear deformation. This changes the strain path and increases the total effective strain level for a given rolling reduction, which is also a suitable technique to produce long products with a refined grain size^{25,26}. Fig. 1¹¹ shows the friction force distribution in the rolling deformation zone in AR and in conventional rolling. In conventional rolling, there are mainly two zones: forward slip zone and backward slip zone, as shown in Fig. 1 (a). In the asymmetric rolling, a shear zone appears between the forward and backward zones, as shown in Fig. 1(b). In contrast to conventionally rolled sheets, AR-processed sheets exhibit uniform microstructure across the thickness, and also strength comparable to or exceeding that of the commercial Al alloy sheets commonly in use. The material is subjected to enhanced shear deformation, while develops high-angle grain boundaries, and ultrafine grains are formed by continuous recrystallization. Thus the complete strain state imposed on the sheet during asymmetric rolling is a combination of plane strain deformation and an additional shear component, which can potentially refine the grains. Zou et al.²⁵ used AR to obtain a pure aluminium sheet with 500 nm grain size. Wronski et al.²⁶ studied the grain refinement of an Al 6061 alloy by asymmetric warm-rolling. In asymmetrically rolled sheets with a 91.8% reduction in thickness at 300°C, fine grains with

an average size of 1 μm have been developed²⁷. The AR technique has also been used to produce multilayer metal products^{27–32}. Zu et al.^{28,29} used AR to produce bimetallic (Al/Cu) clad sheets. They found that the shear friction had a direct impact on the stress state of the individual metals. They also found that lateral shear had the most influence on metal flow and bonding strength, and that the difference between hard and soft metals gradually diminished with increase in the reduction ratio. Pan et al.³⁰ analysed the bonding strength of bimetallic clad samples produced using AR. They found that the friction factor at the interface increased and the relative bonding length increased as the reduction ratio increased.

Angella et al.³¹ carried out a comparison between the AR and ARB techniques as viable methods to bring about refinement in the Al 6082 alloy grain size. They found that both AR and ARB can result in ultrafine grained structures of the alloy. However, there has not been much research into a technique that integrates features of both these processes. This paper describes the use of a combination of the AR and ARB techniques to produce ultra-thin nanostructured AA1050/AA6061 bimetallic foils. The mechanical properties of the foils after each pass are analysed. The results show that both the ductility and strength of the rolled bimetallic foils increase with thickness reduction from 0.14 mm to 0.04 mm.

Results

Mechanical properties. Fig. 2(a) shows the Engineering stress vs. Engineering strain curves of the samples after each pass, along with those for annealed AA1050 and AA6061 sheets. Compared with the original annealed AA1050 and AA6061 sheets, the bimetallic foils show a nearly three-fold increase in strength. The strength of the samples increases with the number of passes, but the increase is only slight after the second pass. After the annealing, the tensile strength of AA1050 sheet (84 MPa) is less than that of AA6061 (115 MPa). After the fourth AR pass, the tensile strength of the bimetallic foils reaches 266 MPa. The ductility of annealed AA1050 is better than that of AA6061 sheets, and the ductility of the AA1050/AA6061 foils shows a large reduction after rolling. In addition, as shown in Fig. 2 (a), the elastic modulus after ARB decreases slightly compared with that of the annealed sheets, and increases with further AR passes. During the cold rolling process, the elastic modulus may change slightly with the deformation. Benito et al.³² studied the change in elastic modulus of pure iron deformed by cold rolling. They also found the above trends. When the annealed sheets are subjected to ARB processing, the number of dislocations or non-pinned dislocations increases, which results in a diminution of the elastic modulus³³. With further AR passes, the number of dislocations storage either in the interior cells or in the peripheral cells will increase. This will result in smaller regions of low dislocation

Figure 1 | Friction force distribution in (a) conventional rolling and (b) asymmetric rolling.

Figure 2 | Tensile test for rolled bimetallic foils after each pass. (a) Engineering stress vs. Engineering strain curves; (b) tensile stress, (c) engineering failure strain.

density. There will be more interactions between dislocations, and dislocation segment length will be smaller although the dislocation density increases, which results in a higher elastic modulus³². Fig. 2(b) shows the tensile stress of the samples as a function of the number of rolling passes. The tensile stress of the samples after the ARB process is 200 MPa, and it increases to 250 MPa after the first AR pass. With further AR passes, the tensile stress of samples increases gradually, reaching ~266 MPa after the fourth AR pass. Simultaneously, the engineering failure strain of the samples reaches a minimum after the second AR pass (Fig. 2(c)). The ductility of the

Table 1 | Tensile strength and failure strain of the rolled bimetallic foils

Rolling process	ARB	AR1	AR2	AR3	AR4
Tensile strength, MPa	199.4	247.2	249.5	261.4	266.6
Failure strain	0.069	0.045	0.036	0.058	0.063

samples reduces with the number of passes until the second AR pass, after which it increases gradually with further rolling passes. The values of the strength and fracture strain of samples after each rolling pass are listed in Table 1.

Fracture micrographs. The fracture surface of the materials was studied using SEM in order to investigate the mechanism of the fracture. Fig. 3 shows the fracture surfaces after tensile tests of the samples after ARB and also after AR passes. Fig. 3(a) is the sample after the ARB process, and Figs. 3(b) to 3(e) for the subsequent first to fourth AR passes. As shown in Fig. 2(a), the ductility of an annealed AA1050 sheet is much better than that of an AA6061 sheet. In addition, the ductility of an AA1050 sheet should be better than that of an AA6061 sheet after rolling with the same deformation because there are more alloy elements in the AA6061 sheets. On the fracture surface, greater necking and ductile fracture implies higher ductility. On the contrary, brittle fracture implies reduced ductility. These features are used to identify the material alloys on either side of the bimetallic foil. Fig. 3 (a) shows that both the AA1050 layer and AA6061 layer experience significant necking, implying good ductility of the samples but relatively weak interfacial bonding. In a subsequent tensile test, the bonding between the AA1050 and AA6061 layers can be easily undone. In contrast, as seen in Fig. 3 (b), after the first AR pass, only a limited number of gaps in the bonding interface are seen, implying greater bonding strength between the AA1050 layer and the AA6061 layer. Most of the fracture surfaces of the AA1050 and AA6061 layers have a gray fibrous appearance with hemispheroidal dimples which occur as a result of micro-void formation and coalescence. This is a

Figure 3 | Fracture graphs of tensile test samples, (a) ARB processed sample, (b) after the first AR pass, (c) after the second AR pass, (d) after the third AR pass, (e) after the fourth AR pass.

Figure 4 | TEM image of ARB processed sample.

characteristic of ductile tensile fracture³⁴, and a small zone near the bonding interface of the AA6061 layer appears to show brittle fracture. After the second AR pass, the AA6061 layer appears to have undergone total brittle fracture. The contact zone of the AA1050 layer also shows small ductility, as shown in Fig. 3(c). With further AR passes, the fracture surface in the AA6061 layer is still dominated by brittle fracture, and the AA1050 layer by ductile fracture, as shown in Fig. 3 (d) and (e). Fig. 3 (e) shows that the necking in the AA1050 layer is much larger than that in Fig. 3(c). This reflects the increase in the ductility of the bimetallic foil after the third and fourth AR passes compared to that after the second AR pass.

Microstructure. Fig. 4 shows a Transmission Electron Microscope (TEM) image of the ARB- processed sample. The material composition on either side of the interface was determined by Energy Dispersive X-ray (EDAX) spectroscopy, as shown in Fig. 5. The AA6061 is seen to contain more Mg than AA1050. After the ARB process, the average grain size in the AA6061 layer is about 250 nm. However, the mean grain size in the AA1050 layer is about 580 nm in the interface region. In the SPD deformation process, the Mg content in Aluminium can dramatically refine the grain size³⁵.

Fig. 6 shows TEM images of the interface zone in the rolled samples: Fig. 6 (a) is for the ARB- processed sample, and (b), (c) and (d) for after the first, third, and fourth AR pass respectively. In Fig. 4, it is difficult to observe residual voids at the bonding interface between the AA1050 layer and the AA6061 layer. However, it is clearly seen in Fig. 6 (a) that the interface between AA1050 layer and AA6061 layer is not bonded well through the ARB process. After the first AR pass, there are still some residual voids in the interface zone. Meanwhile, with the severe plastic deformation in this pass (using a 75% reduction ratio), the grain boundary becomes fuzzy, as shown in Fig. 6 (b). After the third AR pass, the two layers are bonded quite well, and residual voids are virtually eliminated from the test samples. In addition, the grain size of the materials has been refined, as shown in Fig. 6 (c). With further AR passes, the grain sizes in the two layers increase slightly, and they become more uniform, as shown in Fig. 6 (d). This may be the reason for greater ductility in the samples in the tensile test. Fig. 7 shows the log-normal distribution of grain size in the AA1050 and AA6061 layers. The average grain size of the AA6061 layer is refined to about 140 nm, and the grain size of the AA1050 layer is refined to about 235 nm.

Figure 5 | EDAX analysis of samples, (a) locations analysed, (b) AA6061, (c) AA1050.

Discussion

A discussion on interfacial bonding in ARB and AR is presented in the following paragraphs, followed by some observations on the material properties resulting from ARB and AR.

In a bimetallic material, the bond strength is an important material characteristic. Roll bonding is a solid-state process used to join similar and dissimilar metals. Bonding is obtained when the contaminant-free, stretched surfaces, ('virgin' metal surfaces), are exposed and extruded into the virgin material through the cracks of the fractured covering layer, thus establishing contact and bonding between the virgin surfaces in contact. Petrov and Razuvaeva³⁶ studied the effect of uniform pressure on voids and cracks located at the grain boundaries in polycrystalline metals at room temperature. They found that void healing is realized in polycrystalline metals via the dislocation mechanism. Under pressure, shear stresses appear near the residual voids, with the result that the pinned dislocation segments with edge orientation at the void surface became the sites of new dislocations. Above a critical stress, a dislocation loop forms and the void boundary is displaced. Thus, the bond dislocation

Figure 6 | TEM images: (a) ARB processed sample, (b) after the first AR pass, (c) after the third AR pass, (d) after the fourth AR pass.

mechanism is effective at room temperature. Theory and experiment have shown that the stabilized void size R as a function of pressure P is given by the equation,

$$1 - (R/R_0)^3 = CP^{1/4} \frac{(P - P^*)}{G} - P^{5/4} \quad (1)$$

where R_0 is the initial void size, C is a constant, P^* is the threshold pressure, and G is the shear modulus. Due to residual voids around the AA1050/AA6061 interface after the first AR pass, imposition of the shear stress in the process will improve their bond. In Fig. 6 (c) and 6 (d), no gaps are seen at the AA1050/AA6061 interface.

In Fig. 6 (a), there are some residual voids in the interface region, implying a weak bond between the AA1050 layer and the AA6061 layer, and the two layers undergo separation in the tensile test. A

number of theoretical models have been developed for the roll bonding process. Li et al.³⁷ have reported that the aluminium material bonding starts at a threshold reduction ratio of 40%, and this reaches a base value of about 60%–70% deformation. With increasing reduction ratio, most locations on the interface were seen to bond well. Residual voids were seen at a few locations, as shown in Fig. 6 (b). These may be caused by adsorbed contaminants or oxide films. Bay³⁸ proposed a theoretical model showing the basic effects of the both surface conditions and normal pressures on the bond strength:

$$\sigma_B = (1 - \psi^2)Y(p - p_E) + \psi^2 \frac{Y - Y'}{1 - Y'} p \quad (2)$$

where σ_B is the bond strength, ψ is the fraction of the film layer area with respect to the total area, Y the surface exposure of the bond

Figure 7 | Log-normal distribution of grain size after the third AR pass, (a) AA6061 layer, (b) AA1050 layer.

Figure 8 | Deformation of a bimetallic foil in AR ($V_2 > V_1$).

interface surface, p the normal pressure on base metal surfaces, p_E the extrusion pressure, and Y' the threshold surface exposure for the contaminant film. With further rolling passes, a good bond is seen over the entire interface. In addition, for prediction of the bonding strength of materials resulting from rolling, Govindaraj et al.³⁹ also presented a bonding strength model considering the effect of equivalent strain at the interface:

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

where σ_b is the bond strength; σ_0 the tensile stress of sheets; K_1 and K_2 are constants depending on the plasticity of the sheet material and the preparation of the welded surfaces, and ε_e is the equivalent strain at the interface. In the AR process, the equivalent strain is larger than that in conventional rolling. Eq. (4) shows the mean equivalent strain in the sheet induced by the AR process⁴⁰:

$$\varepsilon_e = \frac{2}{\sqrt{3}} \left\{ 1 + \left[\frac{(1-r)^2}{r(2-r)} \tan^2 \theta \right]^{1/2} \ln \frac{1}{1-r} \right\} \quad (4)$$

where $r = 1 - (t_1/t_0)$, with t_0 and t_1 the thickness of the sheet before and after AR, and θ is the apparent shear angle at a given position of the element perpendicular to the surface of the sheet before rolling. The shear angle θ will change with the processing parameters such as diameter, speed, and friction difference between the upper and lower rolls, and as a result affects the bond strength between layers in laminate material production. Eq. (4) implies that the equivalent

Figure 10 | Equivalent strain distribution in section at the exit of the rolling deformation zone.

strain induced by AR is larger than that by conventional rolling. However, the deformation behaviour of ARB-processed sheets subjected to subsequent AR is complex. Pan et al.³⁰ divided the rolling deformation region into four distinct zones for bimetallic sheets during the rolling process, as shown in Fig. 8: Z1 for the unbonded region, Z2, Z3 and Z4 for the bonded regions.

Following this model, a numerical model using the Finite Element Method (FEM) was set up to calculate the equivalent strain distribution in rolling processes. Fig. 9 (a) shows the equivalent strain distribution in a bimetallic sample in the rolling deformation zone during conventional rolling, while Fig. 9(b) shows the corresponding situation for AR. Fig. 9 shows that in both processes, the strain distribution is similar in zones Z1 and Z2. However, in zones Z3 and Z4, the strain distribution is more uniform following AR compared to conventional rolling. Fig. 10 shows the equivalent strain distribution in the interior of the foil at the exit near the deformation zone. Compared with conventional rolling, the equivalent strain is generally higher and more uniform than when using the AR technique. Under these rolling conditions, the mean equivalent strain at the interface reaches 1.9 using the AR technique, compared to only about 1.6 for conventional rolling. Eq. (3) suggests that the bonding strength of interface of AA1050/AA6061 by AR will be greater than that due to the conventional rolling technique.

Strength and ductility are the key mechanical properties of any material, but typically they have contrary characteristics.

Figure 9 | Equivalent strain distribution in rolling deformation zone, (a) conventional rolling, (b) asymmetric rolling.

Table 2 | Chemical compositions of AA 6061 and AA1050

wt%	Si	Fe	Cu	Mn	Mg	Zn	Ti	Cr	Al
AA6061	0.4–0.8	0.7	0.15–0.4	0.15	0.8–1.2	0.25	0.15	0.04–0.35	Balance
AA1050	0.25	0–0.4	0.05	0.05	0.05	0.05	0.03	-	Balance

SPD-processed metals are generally stronger and less ductile compared to coarse-grained materials. AR can induce significant shear strains arising from the contact condition between the rolls and the sheet surfaces. The shear stresses cause the grain boundaries to move⁴¹. Thus, it is possible to obtain a quasi-constant shear strain distribution across the material thickness²⁶. Asymmetrically rolled sheets exhibit uniform a microstructure across the thickness direction, in contrast to the conventionally rolled sheet, and a strength comparable to or exceeding that of the commercial Al alloy sheet commonly in use⁶. According to the Hall–Retch formula,

$$\sigma_r = \sigma_i + k_y D^{-1/2} \quad (5)$$

where k_y is the Petch parameter and D is the grain size. It is seen in Fig. 6 that the grain size gradually decreases with each deformation pass. As shown in Fig. 2(b), with a higher number of AR passes, the tensile strength gradually increases. Simultaneously, the ductility decreases until the second AR pass. Subsequently, with further AR passes, both the ductility and the strength of the bimetallic foil increase. Some recent studies show that the nanostructure may lead to a unique combination of high strength and ductility. Wang et al.⁴² created nanostructured copper by cryorolling and heat treatment, and the product showed high ductility and high strength. The reason for this behaviour is that, while the nanocrystalline grains provide strength, the embedded larger grains stabilize the tensile deformation of the material. Höppel et al.⁴³ found that in pure Al subjected to ARB, both the ductility and the strength could increase compared to a conventional cold rolled state. This can be explained by the process of thermally activated annihilation of dislocations at the grain boundaries⁴⁴. The grain size in Fig. 6 (d) is slightly greater than that in Fig. 6 (c). In addition, Liu⁴⁵ presented a model to predict the ductility of ultrafine materials:

$$\varepsilon_u = \varepsilon_y + \frac{\hat{\sigma}_{s\infty}(1 - \Lambda_s/D)^{0.5}}{\theta_0} \ln \left(\frac{\hat{\sigma}_{s\infty}(1 - \Lambda_s/D)^{0.5} + \theta_0}{\hat{\sigma}_{s\infty}(1 - \Lambda_s/D)^{0.5} + \theta_y} \right) \quad (6)$$

with

$$\theta_0 = C_1 M^2 \alpha \mu / 2 \quad (7)$$

where ε_u is the uniform strain; ε_y the elastic strain; D the grain size; $\hat{\sigma}_{s\infty}$ overall dislocation stress at steady state as $D \rightarrow \infty$; Λ_s the dislocation cell size at steady state; C_1 a measure of the probability of a moving dislocation of unit length to be stopped and subsequently stored at an obstacle; M the Taylor orientation factor; α a constant; μ the shear modulus, and σ_y the yield stress. The ductility is a function of temperature and strain rate, but independent of strain and grain size. This model suggests that the stability of plasticity in ultrafine grained materials could increase to maintain the small-angle grain boundaries. The SPD process results in many large grain boundaries in the samples. In addition, there are still some small-grain boundaries which might result in a change in the ductility. There are also some reports showing that the sheet thickness will affect the strength and ductility when the thickness is lower than a threshold value. Suh et al.⁴⁶ found that the tensile strength and ductility of Al 6K21-T4 sheets decreased almost linearly with reduction in thickness, when the thickness was reduced below a critical value. With a reduction in the sheet thickness, the grains on the free surface are less constrained and can be more easily deformed at a substantially lower flow stress than is the case in the bulk state⁴⁶. In present study, the results do not show an obvious size effect that might be due to the number of nano-sized grains compared to the foil thickness. For that reason, the

mechanical properties of thicker samples produced using the proposed technique are likely to be better.

In summary, the ARB and AR techniques were coupled to produce ultra-thin nanostructured bimetallic foils. A 1.5 mm thick AA1050/AA6061 was produced using the ARB technique, which was subsequently subjected to multiple AR passes to finally fabricate a 0.04 mm thick bimetallic sample. After the third AR pass, the grain size of the AA6061 layer was seen to be 140 nm and that of the AA1050 layer 235 nm. With an increasing number of AR passes, the tensile stress of the bimetallic increases gradually. The ductility reduces until the second AR pass. With further AR passes, both the tensile stress and the ductility increase. Following only the ARB process, the interface between AA1050 layer and AA6061 layer was not seen to bond well. Some residual voids in the interface region were observed after the ARB process. With an increasing number of AR passes, the number and size of residual voids was seen to decrease gradually. After the third AR pass, an excellent bond was observed.

Methods

Sheets of commercial aluminium alloys (AA6061 and AA1050) of dimensions 60 mm (width) $\times$ 1.5 mm (thickness) were used to fabricate the bimetallic foils. The chemical compositions of the two alloys are listed in Table 2. The AA1050 sheet was annealed at 450°C for 1 hour and the AA6061 sheet was annealed at 500°C for 2 hours to achieve a fully homogeneous microstructure²⁴. First, the two sheets were stacked together and welded at one end (total thickness 3.0 mm). The composite sheet was pre-heated to 200°C for 3 min in a furnace and then subjected to one ARB pass (rolled with a nominal reduction of 50% under dry conditions)²⁴. The resulting 1.51 mm thick ARB sheets were cooled to room temperature, and subjected to AR to reduce the thickness progressively from 1.5 mm to 0.04 mm. AR was carried out on a multi-function rolling mill¹¹. The maximum rolling force was 50 kN, and the 120 mm diameter rolls were independently driven by two 5.5 kW motors. The rolling speed ratio between the upper and lower rolls was set as 1 : 1.3. During the rolling process, the AA6061 side of the bimetallic sample was in contact with the lower roll. The rolling schedule is presented in Table 3. During AR, the reduction ratio in each pass is larger than 50%, which means that the final equivalent strain in the foil after one ARB pass followed by four AR passes is larger than that after five circular ARB passes.

After each AR pass, the samples were machined into 25 mm gauge length ASTM specimens. Uniaxial tensile tests were conducted with an initial strain rate of $1.0 \times 10^{-3} \text{ s}^{-1}$ on an INSTRON machine operating at constant speed. All tests were repeated three times. Scanning Electron Microscopy (SEM) was used primarily to reveal details of the fracture mechanism in bonding of the bimetallic sample after each pass. The tensile-tested samples were cut into small square (5 mm $\times$ 5 mm) pieces, and the fractured tips were mounted and carefully aligned upwards on the standard SEM stub. The morphology of the fractured surface of the bimetallic sheets was studied with a Zeiss Auriga Field Emission Scanning Electron Microscope (FESEM) operating at 20 kV with a working distance of about 15 mm. The secondary electron images for the top down view were recorded at the same magnification for comparison. The RD–ND plane of the rolled samples was employed to carry out the TEM test. Based on the nanoindentation marks near the interface of AA6061/AA1050 (with the same force, the indentation depths for two sides are different), an FEI xT Nova Nanolab 200 Dualbeam workstation was used to prepare thin-foil specimens from the bimetallic foils for further TEM observation. The specimens were then 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, and the EDAX was employed to determine the material composition on either side of the bond plane.

Table 3 | Schedule of rolling process

	ARB		AR		
Rolling process, number	Pass 0	Pass 1	Pass 2	Pass 3	Pass 4
Strip thickness, mm	1.5	0.35	0.14	0.07	0.04

Finite Element (FE) simulations of the deformation process under both conventional rolling and AR were carried out on ANSYS/LS-DYNA, using the actual rolling condition of the first AR pass with the ARB-processed bimetallic sheet. The FEM has been widely used in analysis of rolling problems⁴⁷, including bimetallic material rolling process⁴⁸. In the simulation, the thickness for both AA1050 and AA6061 layers after the ARB process was assumed to be 0.75 mm. As in the experiments, the ratios of rolls speed were set as 1 : 1.0 and 1 : 1.3 respectively. The friction coefficient between the sheet and the roll was set as 0.15⁴⁹. During the rolling process, the rolls were regarded as perfectly rigid. Isotropic material models were used for the AA1050 and AA6061 layers. The yield stresses for AA1050 and AA6061 layers were 150 MPa and 195 MPa respectively, as calculated by the relationship between hardness and yield stress using the hardness value in Ref. [24]. In the 2D simulation, temperature change and the sheet width spread were neglected. In the rolling process, the roll rotates with a constant angular speed, and the bimetallic foil enters the roll with an initial velocity and exits under the action of the friction force.

- Zhang, L., Meng, L., Zhou, S. & Yang, F. Behaviors of the interface and matrix for the Ag/Cu bimetallic laminates prepared by roll bonding and diffusion annealing. *Mater. Sci. Eng. A* **371**, 65–71 (2004).
- Amir, A. & Faramarz, F. Deep drawing process of steel/bass laminated sheets. *Comp. Part B* **47**, 75–81 (2013).
- Raulea, L., Goijaerts, A., Govaert, L. & Baaijens, F. Size effects in the processing of thin metal sheets. *J. Mater. Process. Technol.* **115**, 44–48 (2001).
- Qin, Y. *et al.* Development of a new machine system for the forming of micro-sheet-products. *Int. J. Mater. Form.* **1**, 475–478 (2008).
- Hadi, S., Yu, H. L., Tieu, K. & Lu, C. Simulation of defects in micro-deep drawing of an aluminium alloy foil. *AIP Conf. Proc.* **1532**, 298–303 (2013).
- Valiev, R. Nanostructuring of metals by severe plastic deformation for advanced properties. *Nat. Mater.* **3**, 511–516 (2004).
- Zhang, J., Gao, N. & Starink, M. J. Al-Mg-Cu based alloys and pure Al processed by high pressure torsion. The influence of alloying additions on strengthening. *Mater. Sci. Eng. A* **527**, 3472–3479 (2010).
- Loucif, A., Figueiredo, R. B., Baudin, T., Brisset, F. & Langdon, T. G. Microstructural evolution in an Al-6061 alloy processed by high-pressure torsion. *Mater. Sci. Eng. A* **527**, 4864–4869 (2010).
- Jin, Y. G., Baek, H. M., Hwang, S. K., Im, Y. T. & Jeon, B. C. Continuous high strength aluminum bolt manufacturing by the spring-loaded ECAP system. *J. Mater. Process. Technol.* **212**, 848–855 (2012).
- Soliman, M. S., El-Danaf, E. A. & Almajid, A. A. Effect of equal-channel angular pressing process on properties of 1050 Al alloy. *Mater. Manuf. Process.* **27**, 746–750 (2012).
- Yu, H. L. *et al.* Asymmetric cryorolling for fabrication of nanostructural aluminum sheets. *Sci. Rep.* **2**, 772 (2012).
- Yu, H. L., Tieu, A. K., Lu, C., Liu, X. H., Godbole, A. & Kong, C. Mechanical properties of Al-Mg-Si alloy sheets produced using asymmetric cryorolling and ageing treatment. *Mater. Sci. Eng. A* **568**, 212–218 (2013).
- Pirgazi, H., Akbarzadeh, A., Petrov, R. & Kestens, L. Microstructure evolution and mechanical properties of AA1100 aluminum sheet processed by accumulative roll bonding. *Mater. Sci. Eng. A* **497**, 132–138 (2008).
- Rezaei, M. R., Toroghinejad, M. R. & Ashrafzadeh, F. Effects of ARB and ageing processes on mechanical properties and microstructure of 6061 aluminum alloy. *J. Mater. Process. Technol.* **211**, 1184–1190 (2011).
- Simões, F., De Sousa, R., Grácio, J., Barlat, F. & Yoon, J. Mechanical behavior of an asymmetrically rolled and annealed 050-O sheet. *Int. J. Mech.* **50**, 1372–1380 (2008).
- Cui, Q. & Ohori, K. Grain refinement of a 6061 aluminum alloy by asymmetric warm-rolling. *J. Jpn Inst. Light Met.* **52**, 185–189 (2002).
- Goken, M. & Höppel, H. W. Tailoring nanostructured, graded, and particle-reinforced Al laminates by accumulative roll bonding. *Adv. Mater.* **23**, 2663–2668 (2011).
- Shabani, A., Toroghinejad, M. & Shafyei, A. Fabrication of Al/Ni/Cu composite by accumulative roll bonding and electroplating processes and investigation of its microstructure and mechanical properties. *Mater. Sci. Eng. A* **558**, 386–393 (2012).
- Wu, K., Chang, H., Maawad, E., Gan, W., Brokmeier, H. & Zheng, M. Microstructure and mechanical properties of the Mg/Al laminated composite fabricated by accumulative roll bonding (ARB). *Mater. Sci. Eng. A* **527**, 3073–3078 (2010).
- Roy, S., Nataraj, B. R., Suwas, S., Kumar, S. & Chattopadhyay, K. Accumulative roll bonding of aluminium alloys 2219/5086 laminates: Microstructural evolution and tensile properties. *Mater. Des.* **36**, 529–539 (2012).
- Roy, S., Nataraj, B. R., Suwas, S., Kumar, S. & Chattopadhyay, K. Microstructure and texture evolution during accumulative roll bonding of aluminium alloys AA2219/AA5086 composite laminates. *J. Mater. Sci.* **47**, 6402–6419 (2012).
- Saito, Y., Tsuji, N., Utsunomiya, H., Sakai, T. & Hong, R. Ultra-fine grained bulk aluminium produced by accumulative roll-bonding (ARB) process. *Scr. Mater.* **39**, 1221–1227 (1998).
- Quadir, M. Z., Al-Buhamad, O., Bassman, L. & Ferry, M. Development of a recovered/recrystallised multilayered microstructure in Al alloys by accumulative roll bonding. *Acta Mater.* **55**, 5438–5448 (2007).
- Su, L., Lu, C., Tieu, K., Deng, G. & Sun, X. Ultrafine grained AA1050/AA6061 composite produced by accumulative roll bonding. *Mater. Sci. Eng. A* **559**, 345–351 (2013).
- Zou, F., Jiang, J., Shan, A., Fang, J. & Zhang, X. Shear deformation and grain refinement in pure Al by asymmetric rolling. *Trans. Nonferr. Metal Soc. China (Eng. Ed.)* **18**, 774–777 (2008).
- Wronski, S., Ghiliani, B., Chauveau, T. & Bacroix, B. Analysis of textures heterogeneity in cold and warm asymmetrically rolled aluminium. *Mater. Charact.* **62**, 22–34 (2011).
- Jamaati, R. & Toroghinejad, M. R. Cold roll bonding bond strengths: review. *Mater. Sci. Technol.* **27**, 1101–1108 (2011).
- Zu, G. Y., Li, X. B., Ding, M. M. & Yu, J. M. Investigating deformation behaviour of asymmetrically rolled Cu/Al bimetal clad sheets. *J. Northeastern Uni. (Nat. Sci.)* **32**, 675–678 (2011).
- Li, X., Zu, G., Ding, M., Mu, Y. & Wang, P. Interfacial microstructure and mechanical properties of Cu/Al clad sheet fabricated by asymmetrical roll bonding and annealing. *Mater. Sci. Eng. A* **529**, 485–491 (2011).
- Pan, S. C., Huang, M. N., Tzou, G. Y. & Syu, S. W. Analysis of asymmetrical cold and hot bond rolling of unbounded clad sheet under constant shear friction. *J. Mater. Process. Technol.* **177**, 114–120 (2006).
- Angella, G., Dellasega, D., Fare, S. & Vedani, M. A comparison between asymmetric rolling and accumulative roll bonding as means to refine the grain structure of an Al-Mg-Si alloy. *Metall. Sci. Technol.* **28**, 22–26 (2010).
- Benito, J. A., Jorda, J. & Roca, A. Change of elastic constants of pure iron deformed by cold rolling. *Mater. Sci. Forum*, **426–432**, 4435–4440 (2003).
- Wang, C., Li, F., Wei, L., Yang, Y. & Dong, J. Experimental microindentation of pure copper subjected to severe plastic deformation by combined tension-torsion. *Mater. Sci. Eng. A* **571**, 95–102 (2013).
- Rezaei, M. R., Toroghinejad, M. R. & Ashrafzadeh, F. Effects of ARB and ageing processes on mechanical properties and microstructure of 6061 aluminum alloy. *J. Mater. Process. Technol.* **211**, 1184–1190 (2011).
- Sauvage, X., Ganeev, A., Ivanisenko, Y., Enikeev, N., Murashkin, M. & Valiev, R. Grain boundary segregation in UFG alloys processed by severe plastic deformation. *Adv. Eng. Mater.* **14**, 968–974 (2012).
- Petrov, A. I. & Razuvaeva, M. V. Initial stage of void and crack healing in polycrystalline metals under uniform compression. *Phys. Solid State* **47**, 907–913 (2005).
- Li, H., Nagai, K. & Yin, F. Progress in cold roll bonding of metals. *Sci. Technol. Adv. Mater.* **9**, 023001 (2008).
- Bay, N. Mechanisms producing metallic bonds in cold welding. *Weld. Res. Suppl.* **5**, 137–142 (1983).
- Govindaraj, N. V., Lauvdal, S. & Holmedal, B. Tensile bond strength of cold roll bonded aluminium sheets. *J. Mater. Process. Technol.* **213**, 955–960 (2013).
- Cui, Q. & Ohori, K. Grain refinement of high purity aluminium by asymmetric rolling. *Mater. Sci. Technol.* **16**, 1095–1101 (2000).
- Rupert, T. J., Gianola, D. S., Gan, Y. & Hemker, K. J. Experimental observations of stress-driven grain boundary migration. *Science* **326**, 1686–1690 (2009).
- Wang, Y., Chen, M., Zhou, F. & Ma, E. High tensile ductility in a nanostructured metal. *Nature* **419**, 912–915 (2002).
- Höppel, H. W., May, J. & Göken, M. Enhanced strength and ductility in ultrafine-grained aluminium produced by accumulative roll bonding. *Adv. Eng. Mater.* **6**, 781–784 (2004).
- Böhner, A., Maier, V., Durst, K., Höppel, H. W. & Göken, M. Macro- and nanomechanical properties and strain rate sensitivity of accumulative roll bonded and equal channel angular pressed ultrafine-grained materials. *Adv. Eng. Mater.* **13**, 251–255 (2011).
- Liu, W. J. Modelling the dependence of uniform elongation on grain size. *Mater. Sci. Forum* **539–543**, 2576–2581 (2007).
- Suh, C. H., Jung, Y. C. & Kim, Y. S. Effects of thickness and surface roughness on mechanical properties of aluminum sheets. *J. Mech. Sci. Technol.* **24**, 2091–2098 (2010).
- Yu, H. L., Kiet, T., Lu, C., Deng, G. Y. & Liu, X. H. Occurrence of Surface Defects on Strips during Hot Rolling Process by FEM. *Int. J. Adv. Manuf. Technol.* DOI: dx.doi.org/10.1007/s00170-012-4556-7 (2012).
- Chen, D. C. & Cao, J. H. Finite element simulations of A1100/A3003(A6063) sandwich sheet rolling processing. *Key Eng. Mater.* **443**, 51–56 (2010).
- Wang, S. Z. & Ou, L. Establishment of friction coefficient model of cold rolling aluminum plate. *Metal Mater. Metall. Eng.* **35**, 34–36 (2007).

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Author contributions

Y.H. conceived the study. Y.H., L.C., T.K., G.A. wrote the main manuscript text. Y.H., S.Y., L.M., S.L., T.D. and K.C. conducted the experiments. Y.H. carried out the FE analysis. Y.H., L.C., T.K. analysed the data. All authors reviewed the manuscript.

Additional information

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