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# Hot and cold regions during accumulative roll bonding of Al/$Al_2O_3$ nanofibre composites

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**Abstract.** Accumulative roll bonding (ARB) is a severe plastic deformation process invented in order to fabricate ultrafine-grained (UFG) materials. Having sub-micrometre sized or nano grains UFG materials generally have considerably higher strength than conventional materials with grains at micrometre length scale. Further increase in mechanical properties could be achieved by reinforcing UFG materials with nanoparticles or nanofibres. In this work two metal sheets were roll bonded to each other by inducing plastic deformation via the ARB process. The plates were then cut into two, stacked, and rolled again. Rolling was repeated multiple times. Observations and measurements were made in the hot and cold regions of the plate. In this study the aluminium matrix was reinforced with aligned and non-aligned $Al_2O_3$ nanofibres. The morphology of the reinforcement–matrix interphase and mechanical properties of the composites were studied. The microstructure of the material was investigated using scanning electron microscopy accompanied with quantitative microstructure analysis. It was demonstrated that the plastic deformation in the first cycle due to broken $Al_2O_3$ nanofibres continued in the second cycle. The $Al_2O_3$ nanofibres were embedded in the aluminium matrix surface area to improve the mechanical properties during the multiple cycle ARB process. In the cold regions micro-defects such as porosity and cracks were observed while in the hot regions no such defects were detected.

**Key words:** Al-based composite, accumulative roll bonding, severe plastic deformation, ultra-fine grain.

## 1. INTRODUCTION

Accumulative roll bonding (ARB) has a potential for becoming an industrial process for producing composite and ultrafine-grained (UFG) metal sheets. Generally UFG materials have higher strength compared to conventional materials, which have grain sizes larger than a few tens of micrometres. A UFG polycrystalline material is defined as the material having grains with an average size of less than 1 μm. For bulk UFG materials, the presence of a high fraction of high-angle grain boundaries with low dislocation density has been shown to be important for higher strength with retained ductility [1,2].

During the ARB process the material is cut into two, the parts are stacked on top of each other, and then rolled again to achieve a thickness reduction of 50%. It is well known that large plastic deformation can have considerable effects on the microstructure and properties of metals and alloys. For example, intense rolling or drawing is accompanied by microstructure refinement and the formation of cells, sub-grains, and fragments, which can increase strength and enhance other properties. Even in the laboratory conditions it is possible to produce UFG sheets large enough for mechanical testing using ARB [2–5]. Strengthening of aluminium during ARB takes place in the first cycle of rolling due to strain hardening. From the second until tenth cycles, there is no significant increase in the mechanical properties [6]. In order to further improve the mechanical properties using the ARB process 9 to 13 passes of rolling are needed. As has been shown by several researchers [7,8], repetitive cycles of rolling can yield flawless bonding, being also followed by a significant improvement in mechanical properties.

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The ARB process was introduced by Saito et al. with aluminium and steel in 1999 [9]. The ARB process has several advantages over other severe plastic deformation (SPD) processes: (1) high load forming facilities and expensive dies are not needed; (2) the productivity rate is high, and (3) the amount of material to be produced is not limited. Because the process is continuous, ARB is appropriate for manufacturing nanostructured and UFG strips.

In the present study the effect of ARB on hot and cold regions where aluminium AA1070 matrix was reinforced with aligned and non-aligned $Al_2O_3$ nanofibres was investigated. The morphology of the reinforcement–matrix interphase and mechanical properties of the composites were studied.

## 2. EXPERIMENTAL METHODS AND MATERIALS

An annealed aluminium sheet measuring 50 mm × 400 mm with a thickness of 3 mm was used for the experiments. The surface of the aluminium plate was cleaned with a steel brush, followed by de-greasing the contacting surfaces. A scheme of the ARB process for manufacturing composites is presented in Fig. 1.

The starting gamma aluminium oxide nanofibres are aligned bundles, their diameter is 7 nm and length approximately 50 mm [10]. Strips of about 1 mm in thickness were cut from the block and placed parallel to the rolling direction on the brushed and cleaned surface. The reinforcement content in each sample was 0.4 wt%. The composite samples with aligned nanofibres are designated as NFA.

To study the role of alignment, the $Al_2O_3$ nanofibres were mixed with aluminium powders by ball milling. A tumbling ball mill was used to mix 6 wt% of nanofibres in aluminium powder (AMG Aluminum Powder Company). Ball milling destroys the bundles, shortens the nanofibres, and introduces their random alignment. The application of reinforcements was performed similarly to that of NFA samples. The composite samples with randomly aligned and shortened nanofibres are designated as NFRA. As a reference, ARB of AA1070 was performed without reinforcement. These materials are designated as A0. The roll bonding process was performed without lubrication, using a laboratory rolling mill with a roll diameter of 150 mm and the length of the rolls 180 mm. The samples for ARB were cut from AA1070 with a length of 380 mm and width of 50 mm.

The roll bonding process was performed with a 50% reduction in each pass. The Al roll bonded strips were cut in half and preheated to recrystallization (500 °C with dell time of 1 h). The sample was transported to the rolling mill in less than 3 s. The side that entered first was named the hot region and the other half is referred to as the cold region. The ARB process was done in two cycles in which the sample was observed in the HR and CR. The samples after the ARB process are shown in Fig. 2.

The effect on the interphase layer formed on the microstructure and the hardness and density values of the HR and CR in the ARB process of two cycles were studied. A tabletop Scanning Electron Microscope Hitachi TM-100 was used to determine the microstructure and morphology of the grains. Tensile properties were determined using an Instron 8516 type 100 kN servo-hydraulic test machine. The Vickers hardness method indentation with a load of 100 N was used.

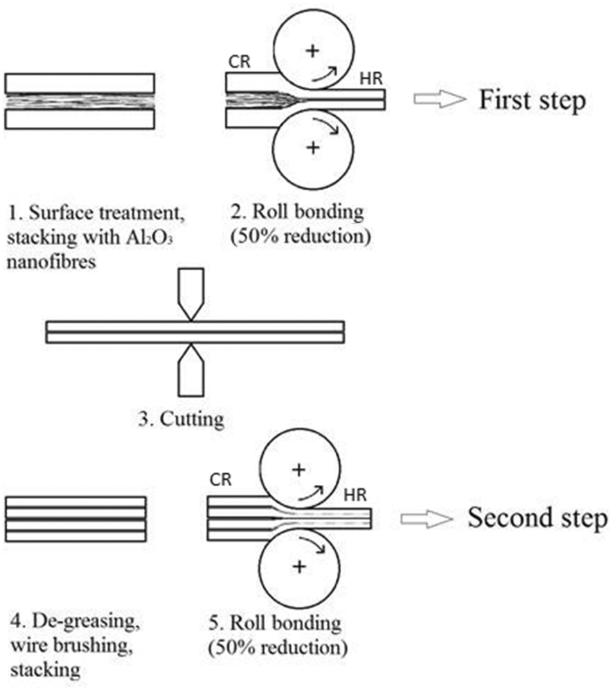



**Fig. 1.** Process stages of accumulative roll bonding (ARB). HR – hot region, CR – cold region of bonding.

**Fig. 2.** Samples after ARB processing.

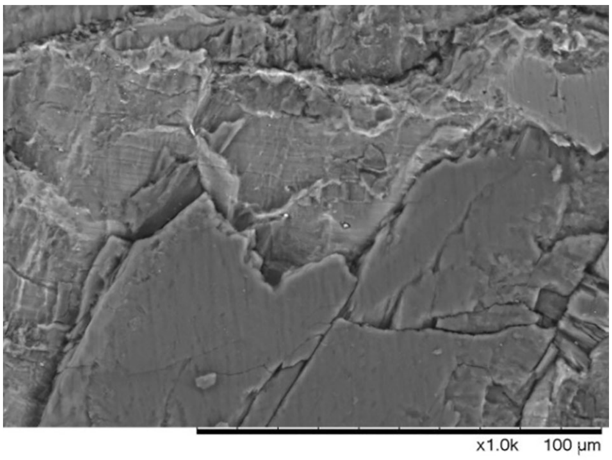
## 3. RESULTS AND DISCUSSION

### 3.1. Development of the microstructure

The first cycle of ARB introduces a significant plastic deformation of the mating surfaces, which leads to the fragmentation of the nanofibre bundles. An example of a nanobundle at the interphase after one pass is presented in Fig. 3a. Further plastic deformation after the second cycle leads to a partial embedding of $Al_2O_3$ nanofibres (Fig. 3b).

The following cycles lead to a further fragmentation, embedding, and uniformity of $Al_2O_3$ nanofibres across the aluminium matrix. The refinement of nanofibres is accompanied by the reduction of the porosity of composites. With each cycle the distance between nanofibre bundles increases. This results in a higher fraction of the metal–metal contact area on the interphase, and therefore squeezes the metal matrix between nanofibre bundles. The amount of 0.4 wt% of $Al_2O_3$ nanofibre reinforcement allows for the elongation of the aluminium matrix, the length of the sample increasing up to 100% only after just two rolling cycles for $Al/Al_2O_3$ compaction. According Ahmadi et al. [11], different mechanical properties of the matrix and reinforcement result in the fracture of the alumina layer, followed by the separation of fragments of alumina during plastic deformation. Fragmented alumina brakes down into particles uniformly distributed throughout the aluminium matrix during the cycle of the ARB process.

(a)




(b)

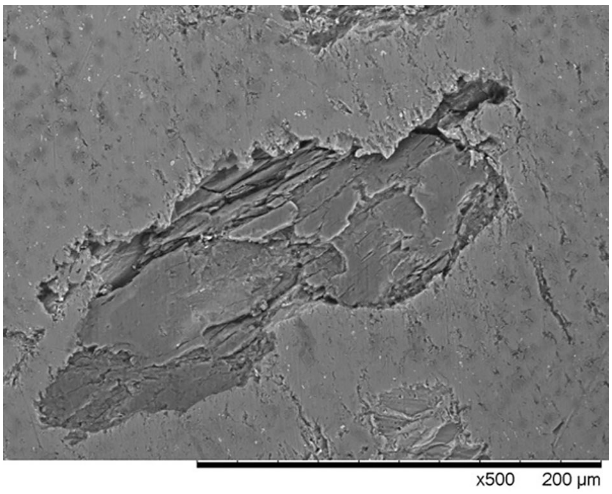



**Fig. 3.** Aluminium oxide nanofibre bundles at the interphase of bonded strips after (a) one pass and (b) two passes of accumulative roll bonding.

The microstructure of the materials processed by ARB is presented in Fig. 4. Chipping and micro-fracturing can be observed in the microstructure of sample AA1070 without reinforcement in the cold region (Fig. 4a). Defects like those in the cold region are not observed in the hot regions (Fig. 4b, d, f); however, plastic deformation and compacting temperature above recrystallization results in the grain growth. The delamination and development of defects in the cold region occur due to the excessive shear strains in the material during ARB.

As demonstrated in Fig. 4c, sample AA1070 with aligned $Al_2O_3$ nanofibres obtains a microstructure with areas of increased porosity in the ARB process. This is caused by the exfoliation of nanofibre bundles at the interface. In the hot regions shown in Fig. 4d, the higher temperatures of the $Al_2O_3$ nanofibres are relatively evenly distributed and well embedded in the aluminium matrix. The porosity remains; however, it is evenly spread as small pores in the interfacial region.

Also the microstructure of the composites with non-aligned $Al_2O_3$ nanofibres was characterized. A mismatch of the stiffness between the matrix and reinforcement in the rolling direction was expected for these composites. This would reduce the anisotropy of the composites' mechanical properties in the rolling direction as well as in the normal direction. The severest delamination was observed in the cold region (Fig. 4e), associated with large elongated pores at the interphase. In the hot region of sample NFRA (Fig. 4f) the bond between the interlayer and the matrix is pore free. Nevertheless, the nanofibre–metal particles inside the interlayer are not fully embedded and a significant amount of porosity can be observed. The metallurgical bond is realized at the interlayer–metal strip interface through the solid state sintering of the compacted surfaces. The high pressure resulting in the deformation of the metal causes shearing, resulting in a bond at the atomic level. Deformation causes the breaking of the creation of clean metal surfaces through the cracking of the native oxide layer on the aluminium surface [9–13].

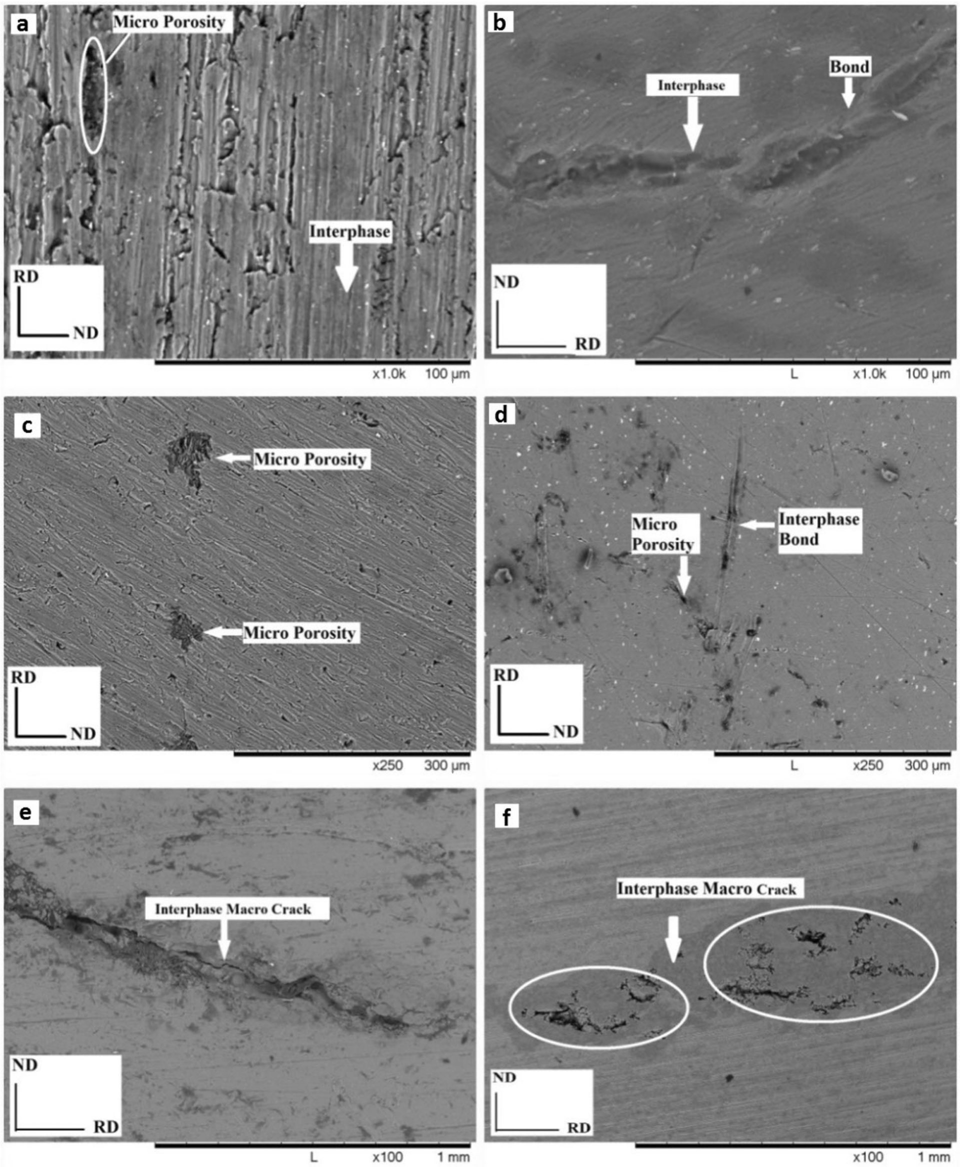



**Fig. 4.** Microstructure of Al-based composites by the ARB process: (a) sample AA1070 in the cold region (CR); (b) sample A0 in the hot region (HR); (c) sample NFA in the CR; (d) sample NFA in the HR; (e) sample NFRA in the CR; (f) sample NFRA in the HR. RD – rolling direction, ND – normal direction. A0 – aluminium AA1070; NFA – AA1070 + aligned nanofibres; NFRA – AA1070 + randomly aligned nanofibres.

## 3.2. Mechanical properties of the composites

Table 1 presents the hardness of the composites after the second cycle for cold and hot regions. The hardness of AA1070 in the hot areas (44 HV10) was slightly higher than in the cold zone (40 HV10). This can be explained by an increase in density in the hot region. Increased hardness in the hot region is accompanied with an increase in tensile strength, ranging up to 148 MPa in the hot region compared to 130 MPa in the cold region. Although this increase is no significant, it does show a change in the trend of the mechanical properties. According Argentero [6], the increase in hardness and tensile strength during the ARB process is minimal after a single cycle. The increase in mechanical properties occurs gradually, corresponding to the increase in the number of cycles. With each cycle the microstructure becomes more uniform and an increase in mechanical properties is reported up to the tenth cycle [6]. For AA1070 with the addition of aligned $Al_2O_3$ nanofibres a higher hardness is observed after two cycles in the cold region. The hardness number is 48 HV10, which

**Table 1.** Mechanical properties of AA1070 based composites

| Material* | Vickers hardness, HV10 | Tensile strength, MPa | Elongation, % | Density, g/cm$^3$ |
|---|---|---|---|---|
| A0–CR | 40.7±0.9 | 130 | 4.0 | 2.74±0.02 |
| A0–HR | 44.0±1.6 | 148 | 3.2 | 2.79±0.31 |
| NFA–CR | 48.5±1.1 | 113 | 2.5 | 2.72±0.01 |
| NFA–HR | 44.7±0.9 | 114 | 2.7 | 2.73±0.02 |
| NFRA–CR | 33.8±3.3 | 57 | 0.9 | 2.67±0.02 |
| NFRA–HR | 32.3±2.6 | 96 | 0.5 | 2.65±0.03 |

* A0 – aluminium AA1070; NFA – AA1070 + aligned nanofibres; NFRA – AA1070 + randomly aligned nanofibres. CR – cold region, HR – hot region.

is the highest among the tested materials. This refers to an effective embedding of the nano reinforcements. The ultimate tensile strength and elongation are not affected by the temperature of the ARB process. An ultimate tensile strength of 114 MPa with 2.7% elongation in the cold region and respectively 113 MPa and 2.5% elongation for the cold region were recorded (see Fig. 5).

Mechanical properties of both unreinforced AA1070 and aligned nanofibre reinforced NFA demonstrated higher values than randomly aligned nanofibre reinforced NFRA materials. The hardness of NFRA was 33 HV10 for the cold region and 32 HV10 for the hot region. The same two samples also showed a decrease in density (2.67 g/cm$^3$ in the cold regions and 2.65 g/cm$^3$ in the hot regions). In samples with NFRA, the tensile strength in the cold area is at 57 MPa, which is lower than in the hot area (96 MPa).

Mechanical properties (hardness and tensile strength) are affected by the bond interphase. For the sample A0, the tensile strength in the hot region was higher than in the cold region; however, the elongation in the hot region was lower than in the cold region. The study of the microstructures revealed some differences in the size and morphology of the defects at the interphase of hot and cold regions. This is the main influencing parameter for differences in mechanical properties. In the case of NFRA the lower tensile strength and hardness values are explained with the high impact of the interface layer. As the nanofibres are premixed with aluminium powder, the volume of the interphase is significant compared to the original volume of the strip metal (by up to 10 vol%). In addition, also the random orientation of the nanofibres may have an influence, especially on tensile strength in a unidirectional tensile test. The influence of the orientation of the nanofibres could be clarified by additional mechanical tests in the transverse direction.

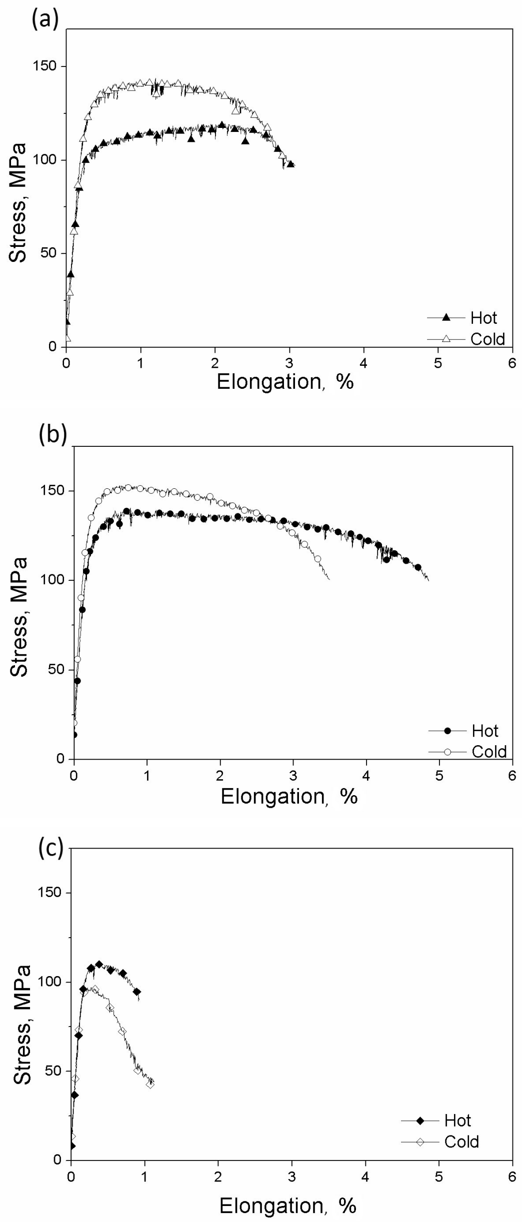



**Fig. 5.** Tensile strength of aluminium-based composites: (a) A0 (aluminium AA1070); (b) NFA (AA1070 + aligned nanofibres); (c) NFRA (AA1070 + randomly aligned nanofibres).

## 4. CONCLUSIONS

- The present research showed the applicability of accumulative roll bonding for producing aluminium-based composites with aligned nanofibre reinforcement.
- ARB has a potential for manufacturing aluminium-based metal matrix composites. The temperature of the ARB process has to be controlled in order to achieve constant mechanical properties throughout the sample. To avoid undesired effects such as

interphase de-bonding and pore formation, critical process temperatures have to be determined. This could be achieved by using of heated rolls during the ARB process.

- The use of pre-mixed nanofibres in the aluminium matrix in powder form was found to be ineffective with the given powder–fibre ratio. The volume of the reinforcement should be decreased for effective bonding.
- In future research combined pressing and rolling, where the goal is to reduce the number of process cycles will be explored.

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# Akumulatiivselt valtsliidetud $Al/Al_2O_3$ nanofiiberkomposiitide kuumad ja külmad piirkonnad

Agus Pramono, Lauri Kollo ja Renno Veinthal

Akumuleeriv valtsliitmine (ARB) on süvaplastse deformatsiooni protsess, millega valmistatakse ülipeene kristall-struktuuriga materjale. Tavamaterjalidega võrreldes tagavad antud meetodiga saavutatavad submikromeetriliste või nanosuuruses teradega materjalid reeglina tunduvalt paremad mehaanilised omadused. Nende edasine täiustumine on saavutatav, lisades nanoosakesi või nanofiibreid. Käesolevas töös liideti kaks metallplaati plastse deformeerimise teel, kasutades ARB protsessi. Vahekihina kasutati suunatud ja mittesuunatud alumiiniumoksiidnanofiibreid. Komposiitide struktuuriuuringud viidi läbi ja mehaanilised omadused mõõdeti valtskeevituse külmas ning kuumas alas. Näidati, et plastse deformatsiooni tõttu esimesel tsüklil nanofiibrid purunesid. Edasine purunemine, peenenemine ja segunemine jätkus ka teisel valtsimisel. Defektivaba materjal ja tugevdatud mehaanilised omadused saavutati, kui materjale deformeeriti kuumalt. Tugevdavate nanofiibrite suunatusel on nanokomposiitide plastsusele märkimisväärne tähtsus.