ALUMINUM ALLOYS STRENGTHENING BY ACCUMULATIVE ROLL- BONDING (ARB) PROCESS

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ABSTRACT

In the present study, strengthening of AA6061 and AA1100 sheets by using the accumulative roll-bonding (ARB) process were studied. The produced sheets had been characterized in the deformed state. The accumulative roll-bonding (ARB) process is an intense plastic straining process. In the ARB process, a strip is neatly placed on top of another strip. The two layers of material are joined together by rolling (reduction degree of 50\%) like in roll-bonding process. Then, the length of rolled material is cut into two halves. These two halves are again stacked and roll-bonded. The whole process is repeated again and again. By the ARB process, the both AA6061 and AA1100 sheets were strengthened dramatically. After 8 cycles (\(\phi = 6.4\)), The ultimate tensile strength (UTS) yield strength (YS), and total tensile elongation of AA6061 reached to 270 MPa, 250 MPa, and 14 \% respectively whereas UTS, YS, and total tensile elongation of AA1100 reached to 155 MPa, 140 MPa, and 9 \% respectively. In present case, the increments in tensile strength for both AA6061 and AA1100 were about 150 and 70\% from the tensile strength of initial sheets respectively. For ARB AA6061 processed sheets, the effect of artificial aging at 175 \(^\circ\)C on the mechanical properties was investigated. It was found that ARB AA6061 processed sheets were strengthened by the ARB process and the artificial age hardening.

1. INTRODUCTION

In recent years aluminum alloys are widely used in automotive industries. This is particularly due to the real need to weight saving for more reduction of fuel consumption and exhaust emissions. Aluminum alloys of the 6xxx series, containing major elemental additives of Mg and Si, are now being used to replace steel skin or closure panels of various car models. For these reasons such alloys subjected to several studies in the past two decades for optimal properties in demanding applications\textsuperscript{[1,2]}. AA6061 and AA1100 alloys have been practically employed as structural material for water-cooled reactors, as cladding material for material testing reactor-type (MTR-type) fuel elements, because its have good nuclear properties, such as small cross section for neutron absorption, good corrosion resistance against coolant water, toughness even after long term exposure in a neutron field, and short life-times of the radioactive nuclei produced by nuclear reactions. Recently, high neutron flux reactors have been developed and the core materials are required to have better resistance against neutron flux higher than 10\(^{15}\) n/cm\(^2\) [3].

There is much current interest in producing metals with very small grain sizes. This interest arises because a reduction in grain size leads to an increase in both the strength and toughness of the material at ambient temperatures. If the small grain sizes are retained to elevated temperatures, where diffusion is reasonably rapid, there is also a potential for achieving good formability and superplastic ductilities\textsuperscript{[4,8]}. Traditionally, small grain sizes are obtained through the development of appropriate thermomechanical processing routes. This procedure is specific for each selected alloy. The processing routes must be adjusted when there are significant changes in the alloy composition and the smallest grain sizes attained through these methods are often in the range 1-10 \(\mu\)m. As a consequence, there is a need to develop simple processing procedures for fabricating materials with ultra-fine grain sizes in the submicrometer or nanometer range. Fine grain structure can be produced by various methods: rapid solidification, mechanical milling/alloying of powder metals, vapor deposition crystallization of amorphous and severe plastic deformation (SPD). Among these methods, SPD is the most promising process for producing fine grained materials. There are three principle methods for subjecting a material to severe plastic deformation: these are known as equal-channel angular pressing (ECAP), high pressure torsion (HPT)\textsuperscript{[6]}, and the accumulative roll-bonding (ARB) process.
The accumulative roll-bonding (ARB) process was developed recently by Saito et al., and many of researchers to achieve ultra-fine grains in metallic materials without changing the specimen dimensions. The accumulative roll-bonding (ARB) process is an intense plastic straining process. In the ARB process, a strip is neatly placed on top of another strip. The two layers of material are joined together by rolling (reduction degree of 50%) like in a roll-bonding process. Then, the length of rolled material is cut into two halves. The two strips are again stacked and roll-bonded again. The whole process is repeated again and again, see Fig (1).

This process can introduce ultra-high plastic strain without any geometrical change, if the reduction in thickness is maintained to 50% in every rolling pass, because the increase in width is negligible in sheet rolling. The achieved strain is unlimited since repetition times are endless in principle. Arbitrarily large deformation is possible by the ARB process. When the reduction is 50% per cycle, the thickness of the initial layer (T), the total reduction (r), and the total equivalent strain (ε) after n cycles can be calculated by using Eq.1, Eq. 2, and Eq. 3 respectively:

The thickness of the initial layer (T) after n cycles is;

\[ T = \frac{T_0}{2^n} \Rightarrow [1] \]

where \( T_0 \) is the initial thickness of strips.
The total reduction (r) after n cycles is

\[ r_n = 1 - \frac{T}{T_0} = 1 - \frac{1}{2^n} \Rightarrow [2] \]

The total equivalent strain (ε) after n cycles is

\[ \varepsilon = \frac{2}{\sqrt{3}} \ln \left( \frac{1}{2} \right) \times n = 0.8 \times n \Rightarrow [3] \]

In the present study, the accumulative roll-bonding ARB process was applied to AA6061 sheets, a typical Al–Mg–Si alloy at 500 °C and AA1100 sheets, (pure aluminum). Since the AA6061 is an age-hardenable alloy, it can be strengthened appreciably by heat treatment. The 6061 alloy has been also currently attracted interests of many researchers because its matrix shows high strength and a good formability. AA100 is a work hardening alloy. Its matrix shows low strength, good formability, and high resistance to corrosion where high strength is not necessary. The objective of the present study is to investigate the evolution of the microstructures and the mechanical properties of both AA6061 and AA1100 with number of ARB cycles.
2. MATERIAL AND EXPERIMENTAL PROCEDURES

AA6061 and AA1100 sheets with chemical composition as shown in Table 1 was used in the present study. The received material was extruded sheets of 6mm thickness at T₀ heat treatment condition, where T₀ is full annealed materials. These materials were subjected to hot rolling at 500 °C until the thickness reached 1 mm in three passes. Then these sheets are undergone to the full annealing process again before they were subjected to accumulative roll bonding process at 500 °C. Then, the ARB processed sheets will be recrystallized by static annealing at 500 °C for 0.5 h, followed by water quenching.

2.1 Microstructure examination

The grain size of the ARB process was too small to be determined by optical microscope (OM). Moreover, the contrast obtained by etching was insufficient for the observation in the scan electron microscope (SEM). Therefore, another method of visualization of grain boundaries had to be sought. The grain boundaries sliding (GBS) occurring during superplastic deformation leads to mutual shifts of grains and freshly exposed grain boundary facets may be observed at the prepolished surface. Supposing that all interfaces are concerned with the grain boundary sliding (GBS) process all grain boundaries should be visualized and the grain size might be determined.

2.2 Tensile Test

Standard sheet tensile specimens, with cross sectional area of 6x1 mm and gauge length 25 mm were used. The tensile specimens were machined from the produced sheets after the accumulative roll boning process by Charmi wire cutting machine according to ASME (E8). Tensile tests were carried out to fracture at room temperature using an Instron tensile machine. The machine has loading range from 0 to 100 KN. The cross-head speed (C.H.S) used in this investigation was 2.0 mm / min.

<table>
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<tr>
<th>Table 1 Chemical Composition of AA6061 and AA1100 Sheets</th>
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<td><strong>Element</strong></td>
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<td>AA6061</td>
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<td>AA1100</td>
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3 RESULTS AND DISCUSSIONS

AA6061 and AA1100 sheets had been subjected to ARB process as shown in Fig. (1). The rolling process was taken place at 500 °C. The ARB process up to eight cycles had been successfully performed without shape defects of the produced specimens.

Grain Refinement ARB process

The microstructure of AA6061 ARB processed sheets

The grain size of the initial AA6061 sheets was in range of 45 to 55 micron. A new method of visualization of grain boundaries, the grain boundaries sliding (GBS), was used by scanning electron (SEM) microscope. The microstructure of the specimen ARB processed by one cycle has relatively large grains, elongated in the rolling direction with clear grain boundaries. As the number of ARB cycles increases, the microstructure became finer. Figure 2 showed that the grain of produced sheet of AA6061 by ARB process was equiaxed grains and the size of these grains are less than three micron after eight passes of ARB process, Fig. 2 (d). At the early stages of the ARB, original grains were subdivided by deformation-induced boundaries such as dense dislocation walls and cell boundaries. With increasing ARB cycles, the lamellar boundaries parallel to rolling direction were introduced in the course of grain subdivision. For further cycles, the decrease in the spacing of the lamellar boundaries occurred with increasing ARB cycles and the lamellar boundaries were divided by interconnecting boundaries. Finally, the very fine grains with high angle grain boundaries, which have a pancake shape.
The microstructure of AA1100 ARB processed sheets

Unfortunately, the using of the mentioned method in experimental work, which were successfully applied to characterize AA6061 ARB processed sheet, for determination of the grain size of AA1100 ARB processed sheets at different ARB cycles was inaccurate, see Fig.3, because the need of transmission electron microscopy (TEM) for this purpose. Huang et al.[20] studied the microstructure in commercial purity aluminum deformed from medium to high strain ($\varepsilon = 1.6 - 6.4$) by accumulative roll bonding (ARB) at room temperature and at 473 K using transmission electron microscopy. They found that a sub-micrometer lamellar structure characterized the microstructure at high strains ($\varepsilon = 1.6$), and that the lamellar boundary spacing decreases and the misorientation across the lamellar boundaries increases with increasing rolling strain. Also, They observed that the structural changes during ARB and during cold-rolling had a great similarity. However, the boundary spacing and misorientation angle evolve with a faster rate during ARB than during rolling. This difference is related to a significant amount of shear strain which during ARB is superimposed on the strain caused by the nominal thickness reduction.

AA6061 Strengthening

The relationship between the number of ARB cycles and tensile properties of the produced sheets after rolling process are shown in Fig. 4. The ultimate tensile strength (UTS) yield strength (YS) level monotonously increased with increasing ARB cycles. It was observed that the difference between UTS and YS was decreased with increasing ARB cycles, (i.e. with decreasing the grain size). The difference between yield stress and tensile strength corresponds to the amount of work-hardening. In the ultra fine grained materials, the tensile strength becomes nearly equal to yield strength, in other words, work-hardening becomes hard to occur[24]. The total elongation decreased with increasing the ARB cycles until 2 cycles, thereafter the total elongation began to increase slightly with increasing the ARB cycles. For example the UTS increased due to the ARB process for one cycle ($\varepsilon = 0.8$) from 110 MPa (sheets in full annealed condition) to 180 MPa whereas the tensile elongation decreased from 29 % to 15 %. However, after 3 cycles ($\varepsilon = 2.4$), the UTS increased slowly when the number of the cycles increased. After 8 cycles ($\varepsilon = 6.4$), UTS, YS, and total tensile elongation reached to 270 MPa, 250 MPa, and 14 % respectively. These trends of strength correspond well with the reduction of the grain size, which suggests that the strength of the ARB processed AA6061 sheets is determined primarily by the very fine grained structures as described by Hall-Petch relationship[25]. At the same time, however, dislocation strengthening, solution strengthening and precipitation hardening also affect the strength of the ARB processed AA6061 sheets.

Artificial Aging at 175 °C

In the present study, the effect of artificial aging on the mechanical properties of AA6061 ARB processed sheets was investigated. This process was carried out at 175 °C for different times (0-20 hrs). The choice of 175 °C for study the artificial aging behavior of these sheets was related to the pervious studies on the precipitation hardening of AA6061 which indicate that 175 °C is optimum temperature to achieve the maximum yield strength and ultimate strength.

Fig. 5 and 6 showed the yield strength and ultimate tensile strength of AA6061 ARB processed (3, 6, and 8 cycles) and initial AA6061 sheets at various aging time. The peak aging yield strength (YS) and ultimate tensile strength (UTS) of the tested sheets reached at 175°C for in different time pried as follows: 1.5, 2.7, and 10 hrs for 8 ARB cycles, 5 ARB cycles, 3 ARB cycles, and initial AA6061 sheets respectively. The results indicated that the yield strength and tensile strength increase with increasing the ARB cycles. The artificial aging of the AA6061 ARB processed sheets was accelerated with increasing the ARB cycles. Fig. 7 illustrated the effect aging times on the tensile elongation of the tested sheets at 175 °C. The tensile elongation decrease with increasing aging time of artificial aging at 175 °C until the peak aging point depending on the AA6061 sheets conditions. After that it remained constant in the ARB processed sheets. But, the tensile elongation of initial AA6061 slowly increased after the peak aging point.
Figure 2 SEM micrographs of the AA6061 ARB specimens after; a) two, b) four, c) six, d) eight cycles in the rolling direction.

Figure 3 SEM micrographs of the AA1100 ARB specimens after eight cycles in the rolling direction.
From the above results and previous studies\cite{6, 25, 26, 27}, the significantly high strength of the FG 6061 Al alloy may be attributed to: (i) solid solution strengthening, (ii) grain refinement strengthening, (iii) dislocation strengthening, oxide particles strengthening, and (iv) precipitation strengthening. The precipitation strengthening results from the precipitate’s ability to impede dislocation motion by forcing dislocations to either cut through or circumvent the fine precipitates. In either case, higher density of oxide particles and precipitates leads to higher strength. For aged AA6061, the cutting of fine oxide particles and precipitates by dislocation is the main strengthening mechanism. The enhanced aging kinetics of a given ARB condition directly correlates with the marked increase in the number density of matrix. The increase in the number density of nucleation sites decreases the average distance between precipitates. This inter particle spacing is the limiting factor in the soft impingement of neighboring precipitates. The greater the number of nucleation sites, the finer the distribution of precipitates, the smaller the average diffusion Field, and therefore the shorter the time required age hardening process. The precipitation strengthening results from the precipitate’s ability to impede dislocation motion by forcing dislocations to either cut through or circumvent the fine precipitates. In either case, higher density of precipitates leads to higher strength\cite{26, 27}.

AA1100 Strengthening

The relationship between the tensile properties and number ARB cycles for AA1100 sheets after hot rolling process are shown in Fig. 8. The ultimate tensile strength (UTS) and yield strength (YS) level monotonously increased with increasing ARB cycles. It was observed that the difference between UTS and YS was decreased immediately after one cycle of ARB. Whereas, the difference between yield stress and tensile strength at an identical grain size corresponds with the amount of work-hardening. In the ultra fine grained materials, the tensile strength becomes nearly equal to yield strength, in other words, at which work-hardening becomes hard to occur\cite{25}. The total elongation decreased immediately from 48 %, (tensile elongation of initial AA1100 sheets), to 9 % after one cycles of ARB. Then, the tensile elongation decreases very slowly with increasing the ARB cycles until 5 cycles, thereafter the total elongation began to increase slightly with increasing the ARB cycles. For example the UTS increased due to the ARB process for one cycle ($\varepsilon = 0.8$) from 92 MPa (sheets in full annealed condition) to 144.5 MPa and fracture within 9 per cent of strain. After that cycle, the tensile strength increases very slowly with increasing ARB cycles till cycle six ($\varepsilon = 4.8$). After that, the tensile strength remains at constant value with increasing the ARB cycles. After 8 cycles ($\varepsilon = 6.4$), UTS, YS, and total tensile elongation reached to 155 MPa, 140 MPa, and 9 % respectively. This values are much better than those of high strained AA1100, H16\cite{28}, specially the total tensile elongation i.e. the tensile elongation of AA1100 at temper H16 is about 4%. This mean that the tensile elongation increased due applied ARB process about 120 % than that of high strained AA1100 at the same strengthening condition.

It is made clear that the proposed accumulative roll-bonding (ARB) process causes very fine grains and surprising strength. These effects are confirmed experimentally by two materials; Al-Si-Mg-alloy (AA6061) and pure aluminum (AA1100). The results indicted that the strengthening effect of AA6061 sheets due to ARB process was more higher than that of AA1100 sheets. This is due to the strengthening of AA6061 is summation of reduction of the grain size, oxide dispersion, dislocation strengthening, solution strengthening and precipitation hardening while the increase in strength of AA1100 was due to reduction of the grain size, oxide dispersion, and dislocation strengthening only. Moreover these strengthening parameters are affected by the type of the alloy, Fig 9. These SEM micrographs showed that the aluminum oxide particles which dispersed in ARB specimens had different size, shape, and concentration in both used alloys.

There are two possible additional mechanisms in the ARB process which differ from other high straining processes. The first possible mechanism is the effect of severe shear deformation just below the surface. It has been reported that severe shear deformation is introduced by friction between the work piece and the roll under dry conditions\cite{13, 29}. This shear deformation significantly increases the equivalent strain from the value calculated by equation (3) and promotes grain refinement. Moreover, the ARB process can introduce this severely deformed region into the interior of the material by repetition. The whole thickness of materials may be severely strained after several cycles. The other mechanism is the introduction of new interfaces. A large number of interfaces are introduced by several ARB cycles. These interfaces show a well-developed fiber structure. The oxide films on the surfaces, as well as inclusions, are dispersed.
**Fig. 4** Tensile properties of the AA6061 ARB processed sheets at 500 °C

**Fig. 5** The effect of artificial aging on yield strength of AA6061 ARBed sheets

**Fig. 6** The effect of artificial aging on ultimate strength of AA6061 ARBed sheets

**Fig. 7** The effect of artificial aging on tensile elongation of AA6061 ARBed sheets
uniformly by repetition\textsuperscript{[16]}. These things contribute to the strength and may act as obstacles for grain growth. However, the general mechanism of the grain refinement during ARB is still unclear at this stage and requires further study.

The advantage of this process against other high straining processes is its high productivity and the feasibility of large-sized material production. Although the experiments have been carried out with narrow 100 mm wide materials in this study, it is supposed that application to bulk materials such as wide strips in a coil is not difficult. The process does not require any special machines because the roll-bonding is widely adopted in clad metal production\textsuperscript{[30]}. The process can be readily industrialized.

4 CONCLUSION

The present results clearly showed that fine grained AA6061 and AA1100 with surprising strength can be easily obtained by ARB process. It is practically very important because rolling is the most appropriate process to produce the bulk materials. If this process were applied to practical use, we could obtain high-strength materials without additive special alloying elements by a simple process and without complicated thermomechanical treatment. This satisfies the recent social demands of recycling and energy saving.

In addition to the grain refinement, the dispersion hardening by oxides which form on the surfaces and are taken inside during the repetition of stacking and roll-bonding would contribute to the strengthening to some extent in the present case.

By ARB process, the aluminum alloys strengthening was achieved. In present case the increments in tensile strength for both AA6061 and AA1100 were about 150 and 70 \% from initial condition respectively.

It was found that ARB AA6061 processed sheets were strengthened by the ARB process and the artificial age hardening. These two strengthening effects were made additive. So, it was possible to significantly improve the strength of AA6061, making them much more attractive in high strength structural applications.

![Graph showing tensile properties of AA1100 ARB processed sheets at 500 °C](image)

*Fig. 8 Tensile properties of the AA1100 ARB processed sheets at 500 °C*
5 REFERENCES


Fig. 9  SEM micrographs of eight ARB cycles processed specimens in the rolling direction.