PRODUCTION OF ULTRA-FINE GRAINED BULK ALUMINUM ALLOYS BY ACCUMULATIVE ROLL-BONDING (ARB) PROCESS

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ABSTRACT 1100 and 5083 aluminum alloys were highly strained by the Accumulative Roll-Bonding (ARB) process, which is a novel high-straining process for bulk materials. In ARB process, the rolled material is cut, stacked and rolled again. The stacked sheets are bonded during rolling simultaneously (roll-bonding). ARB of 1100 and 5083 alloys was successfully carried out on the condition that the reduction per cycle was 50% and the rolling temperature was 473K. After several cycles of ARB, ultra-fine grains whose grain sizes are less than 1μm formed in the both materials. The volume fraction of the ultra-fine grains increased with increasing the number of cycles, i.e., strain. Electron diffraction patterns showed that they are the polycrystals with large misorientations. The mean grain sizes of the 1100 after 8 cycles and the 5083 after 7 cycles were 670nm and 280nm, respectively. The ARBed materials with ultra-fine grains showed very high strength, i.e., 300MPa for 1100 after 6cycles and 550MPa for 5083 after 7cycles.

Keywords : *ultra-fine grain, intense straining, accumulative roll-bonding, high strength*

1. INTRODUCTION

It is expected that ultra-fine grained materials with grain sizes less than 1μm would perform high strength, high toughness, and other superior properties. Although some special processes, such as crystallization of amorphous or rapid solidification, would realize submicrometer grained materials, it has been clarified that ultra-fine grains can be obtained simply by intense plastic straining. Several intense straining techniques, such as torsion straining [1-4], equal-channel angular pressing (ECAP) [4,5], and mechanical milling of powder metals [6,7], have been proposed for grain refinement. However, those techniques are not appropriate to produce large bulk materials. On the other hand, we have recently developed a new intense plastic straining process for bulk materials, named Accumulative Roll-Bonding (ARB) [8-10]. **Figure 1** illustrates the principle of ARB. In ARB, the rolled material is cut, stacked to be the initial thickness and rolled again. In order to obtain the bulk material, the stacked sheets are bonded during rolling simultaneously (roll-bonding). Therefore, the achieved strain is unlimited in this process because the repetition times are endless in principle. The purpose of this paper is to clarify the course of the

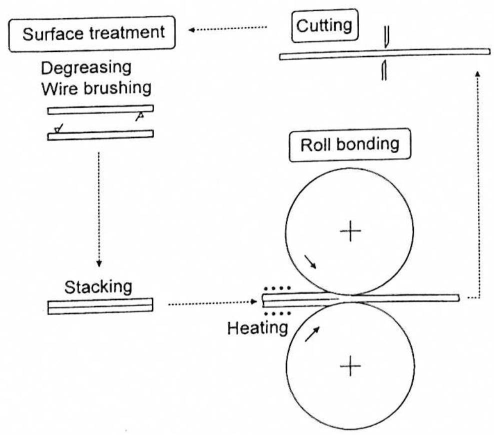

Figure 1 Schematic illustration showing the principle of Accumulative Roll-Bonding (ARB).

microstructural evolution and the change in mechanical properties of 1100 and 5083 aluminum alloys during ARB.

2. EXPERIMENTAL

1100 and 5083 aluminum alloys were used in this study. The chemical compositions of the materials are summarized in **Table 1**. Fully annealed 1100 and 5083 sheets 1mm in thickness, 20mm in width and 300mm in length were provided for ARB. The mean grain sizes in the starting sheets were 37μm and 18μm for 1100 and 5083, respectively. ARB was conducted on the condition that the reduction in thickness per cycle was 50% (equivalent strain of 0.8) and the rolling temperature was 473K. Two pieces of the sheets were stacked after degreasing and wire-brushing, held in an electric furnace set at 473K for 300s, and then roll-bonded. The roll-bonding was done without lubricant using the two-high mills whose roll diameters are 255mm for 1100 and 310mm for 5083. The mean strain rates were $12s^{-1}$ for 1100 and $43s^{-1}$ for 5083, respectively. The 50% reduction at 473K was quite enough to bond the sheets. The sheet was air-cooled after roll-bonding. The rolled sheet 1mm in thickness was cut into two pieces, and the above mentioned procedures were repeated up to 8 cycles or 7 cycles for 1100 or 5083, respectively.

Tensile specimens 10mm in gage length and 5mm in width were electrospark-machined from the ARBed sheets. Tensile direction was parallel to the rolling direction. Tensile tests at ambient temperature were carried out using an instron-type testing machine at a cross-head speed of 0.5 mm min^{-1}. The microstructural observation by transmission electron microscopy (TEM) was done for the specimens after ARB. Thin foils parallel to the rolling plane were prepared by twin-jet electropolishing using a 400ml HNO_3 + 800ml CH_3OH solution.

Table 1 Chemical compositions of the alloys used in this study. (mass%)

	Si	Fe	Cu	Mn	Mg	Cr	Zr	Ti	V	Al
1100	0.10	0.53	0.11	0.01	0.02	0	0.0012	0.02	0.01	bal.
5083	0.08	0.19	0.02	0.57	4.45	0.06	0.01	0.02	0.01	bal.

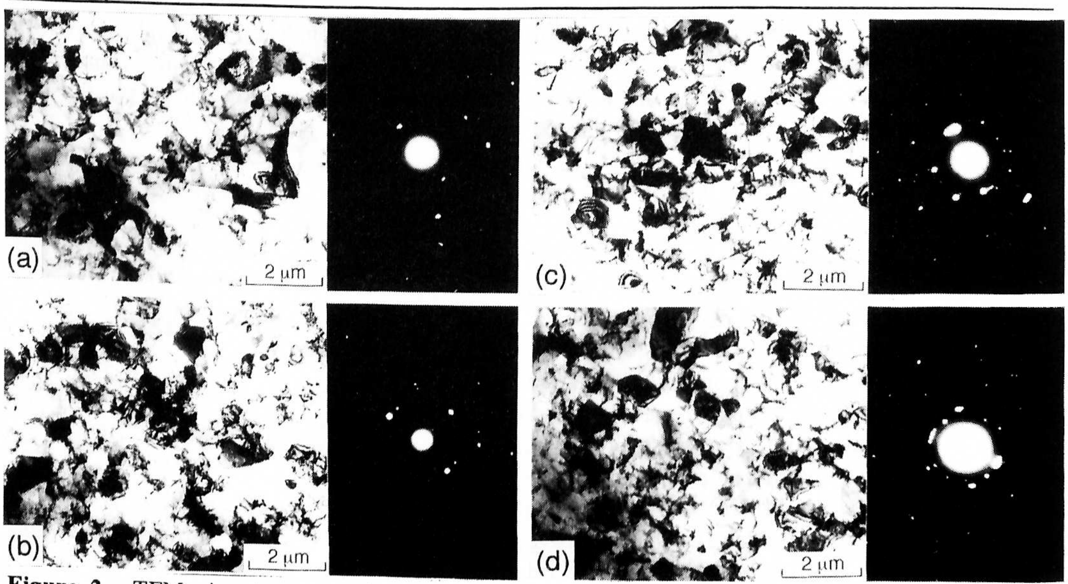

Figure 2 TEM microstructures and corresponding SAD patterns of 1100 alloy ARBed at 473K by 1 cycle (a), 3 cycles (b), 5 cycles (c), or 8 cycles (d).

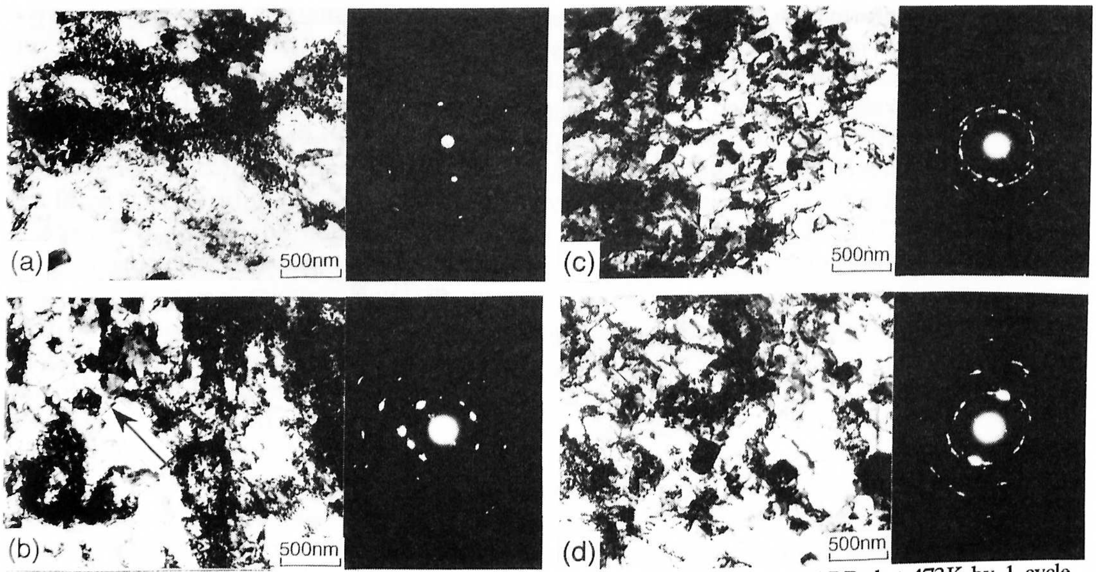

Figure 3 TEM microstructures and corresponding SAD patterns of 5083 alloy ARBed at 473K by 1 cycle (a), 3 cycles (b), 5 cycles (c), or 7 cycles (d).

3. RESULTS

Figure 2 shows the TEM microstructures of 1100 alloy after various cycles of ARB. Every selected area diffraction (SAD) pattern in this study was taken from the center of the left image using an aperture with a diameter of 1.6μm. After 1 cycle, grain structures can be already seen. They are, however, subgrains, because the SAD pattern shows a nearly single net pattern indicating small misorientation. Actually most of the boundaries were composed of dislocation networks. Fine grains surrounded by clear but irregular-shaped boundaries with diameters smaller than 1μm partially appeared after 3 cycles of ARB (Fig.2(b)). SAD pattern of the region including the ultra-fine grains shows that large misorientation starts to generate. The fraction of the ultra-fine grains increased with increasing the number of cycles (strain), while the grain size kept a constant value of about 700nm. After 6 cycles, the whole specimen was covered with the ultra-fine grains and every SAD pattern was quite complicated ones which indicates that the ultra-fine grains are polycrystals having large misorientations to each other.

The microstructure of the 5083 alloy showed the similar process during ARB (**Fig.3**). Different from 1100, subgrains were scarcely observed in 5083 alloy but only the cell structures with tangled dislocations were seen at the early stages of ARB (Fig.3(a)). Though most

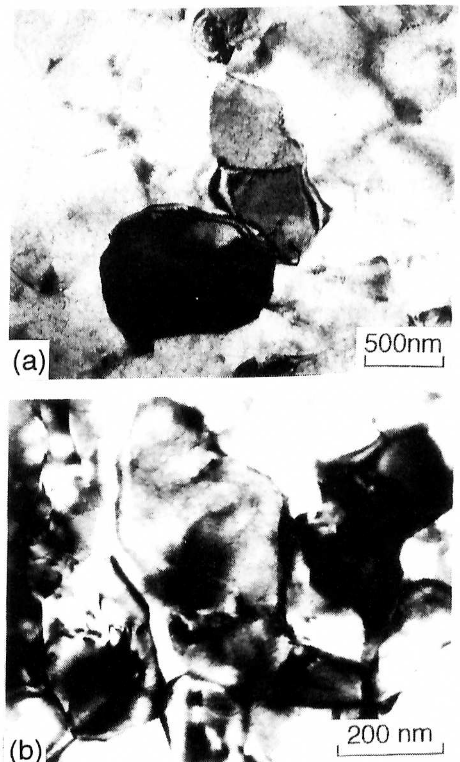

Figure 4 TEM microstructures of 1100 alloy ARBed by 8 cycles (a) and 5083 alloy ARBed by 7 cycles (b).

parts showed deformation cell structures after 3 cycles, ultra-fine grains appeared in part, as is pointed out by an arrow in Fig.3(b). The number of ultra-fine grains increased with increasing strain. Most areas were filled with the ultra-fine grains after 7cycles (Fig.3(d)), though some areas still showed deformation structures. The SAD pattern in Fig.3(d) showed quite complicated ones, indicating that the ultra-fine grains are the polycrystals. The ultra-fine grains in 5083 alloy were finer than those in the 1100 alloy. **Figure 4** shows the TEM micrographs of the both alloys after a number of cycles. The mean grain sizes were 670nm and 280nm for 1100 and 5083, respectively. It should be noted that the ultra-fine grains formed locally inhomogeneously in the both ARBed materials at the middle stages of ARB (3 to 5 or 7 cycles). That is, some areas were covered with the ultra-fine grains, while the other areas had scarcely the ultra-fine grains.

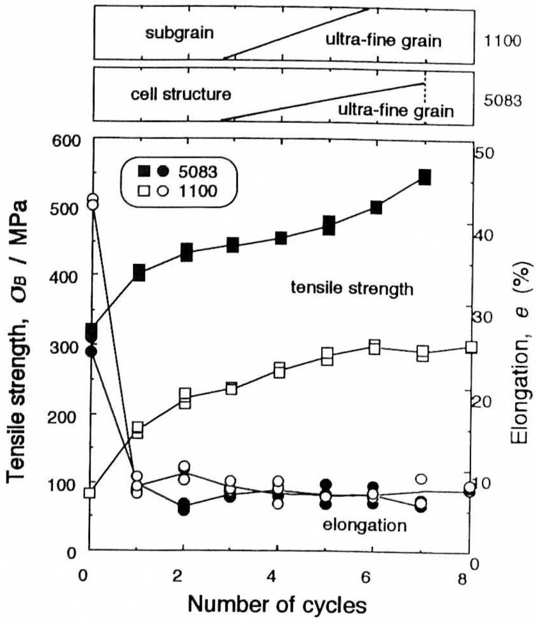

Figure 5 Change in mechanical properties of 1100 and 5083 during ARB. The microstructural changes are schematically shown above the figure.

The change in mechanical properties of the both alloys during ARB is shown in **Fig.5**. The microstructural changes are also schematically illustrated above the figure. The tensile strength increased with increasing the number of cycles (strain). In 1100, it reached 300MPa after 6 cycles, which is surprisingly 3.7 times greater than that of the starting material (80MPa). The tensile strength did not change after 6 cycles and showed a constant value, which corresponds well with the fact that the whole material was filled with the ultra-fine grains after 6 cycles. In the case of 5083, the tensile strength was still increasing after 7 cycles. It corresponds with the microstructural result that the material was still not completely covered with the ultra-fine grains even after 7 cycles. The material ARBed by 7 cycles showed a strength of 550MPa which is 1.7 times larger than that before ARB. The elongation largely decreased to about 8% by 1 cycle of ARB, and kept a constant value of six to ten percents even when the strain further increased in the both alloys.

4. DISCUSSIONS

The present study clarified that the novel ARB process can produce ultra-fine grained bulk aluminum alloys with surprising strength. This high strength is presumably due to grain refinement, because the increase in tensile strength after 3 cycles corresponded with the formation of ultra-fine grains well. It has been also clarified in parallel investigations that the ultra-fine grains form not only in aluminum alloys but also in a steel by ARB [11] and the ARBed 5083 alloy with ultra-fine grains performs low temperature superplasticity at 473K [12]. The ultra-fine grains in the ARBed aluminum alloys have a quite similar feature to those reported in the studies about torsion straining[1,4] or ECAP [4,5] processes. In the present ARB, however, the rolled material was reheated at 473K for 300s before next roll-bonding. It is possible that a microstructural change occurred during this reheating. In order to clarify the microstructural change, the ARBed 5083 specimens were only reheated at 473K for 300s, and then observed by TEM. **Figure 6** shows examples of the results, which are the TEM microstructures of the 5083 alloy ARBed by 3 cycles and then reheated. They are equivalent to the microstructures just before the 4th roll-bonding. Although the ultra-fine grains were observed only in part in the material ARBed by 3 cycles (Fig.3(b)), the deformed structure was obviously rearranged by reheating and the number of the ultra-fine grains increased (Fig.6(a)). Such microstructural changes were recognized in every reheated material when it had been processed by more than 2 cycles. This indicates that the formation of the ultra-fine grains proceeds during reheating. It should be noted, however, that this result does not deny the possibility of dynamic change in microstructure during rolling. Actually the materials after roll-bonding showed equiaxed ultra-fine grains with the similar grain sizes to those after reheating in spite of 50% reduction. Further, it is noteworthy that the formation of ultra-fine grains are certainly inhomogeneous in the same sample, as Fig.6(b) scarcely shows the ultra-fine grains. The areas without ultra-fine grains always showed small local misorientations (see SAD pattern in Fig.6(b)). This must be a key to solve the mechanism of the formation of ultra-fine grains.

The above results suggest that large local misorientation is necessary for the ultra-fine grains to form. On the basis of those observations, we propose a hypothetical model for the formation of ultra-fine grains. Intense straining introduces large local misorientations (in other words large local lattice curvatures) into the material. The upper illustration in **Fig.7** indicates schematically the orientation profile in such a region. The local misorientations can be described by geometrically necessary (g-n) dislocations proposed by Ashby [13], so that the region with large local misorientation involves locally high dislocation density as well. Although only diffusion within a short range can occur in aluminum alloys at 473K, the steep change in orientation described continuously by g-n dislocations could be converted into a kind of "boundary" structure by such a limited diffusion, resulting in the ultra-fine grains (see the lower illustration in Fig.7). Those boundaries illustrated in the lower figure in Fig.7 might have somewhat diffuse and unplane structure. Valiev et al. [3,4] called

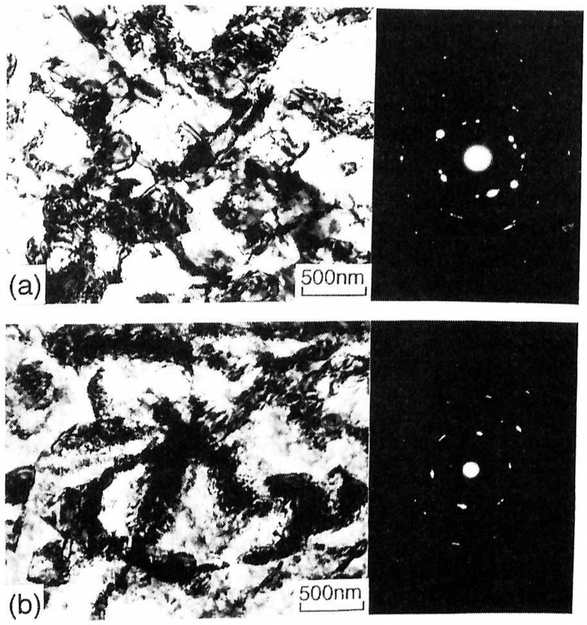

Figure 6 TEM microstructures of a 5083 alloy ARBed by 3 cycles and reheated at 473K for 300s.

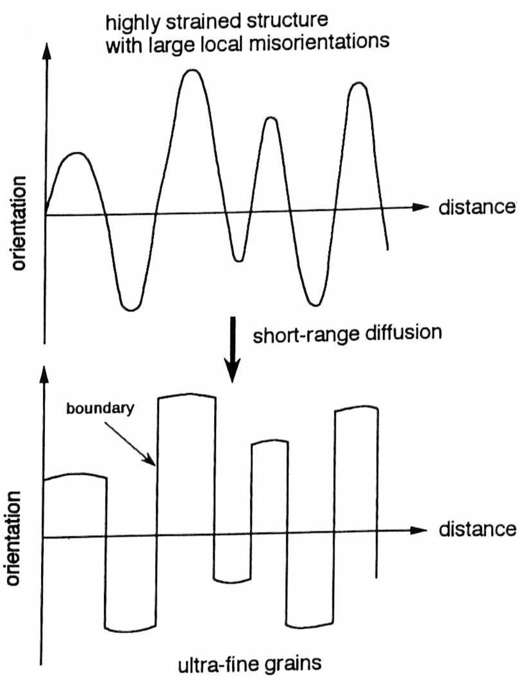

Figure 7 Schematic illustrations showing a model for the formation of ultra-fine grains.

such a boundary "nonequilibrium grain boundary". Anyway, this structural change could occur because the orientation change is very steep and the driving force, i.e., the dislocation density, to drive the formation of the boundaries is very large in highly strained materials. On the other hand, it would be difficult for ultra-fine grains to form in the regions with small local misorientations like Fig.6(b). The process shown in Fig.7 might also occur locally in a material deformed to an ordinary extent of strain. For example, the nucleation of recrystallization at the shear bands, where a large strain localizes, in cold-rolled and annealed sheets [14] might be produced in the same process described above. In the materials strained by a relatively low degree, however, the grains formed would grow immediately at high temperature or the number of the grains are too small to be detected at low temperature, since the regions with large local misorientations are only the minor parts in the deformed microstructures.

5. CONCLUSIONS

A novel intense straining process, ARB, was applied to 1100 and 5083 aluminum alloys, and ultra-fine grained bulk materials were successfully obtained. The mean grain sizes after 7 or 8 cycles of ARB at 473K were 670nm and 280nm for 1100 and 5083, respectively. It was clarified that the formation of ultra-fine grains proceeds during reheating before roll-bonding. The ARBed sheets with ultra-fine grains showed very high strength two or three times larger than that of the starting materials, i.e., 300 MPa for the 1100 alloy ARBed by more than 6 cycles and 550MPa for the 5083 alloy ARBed by 7 cycles. The elongation of the ARBed materials were several percents regardless of the cycles.

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