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Recrystallization or Not ?: Formation Mechanism of Ultrafine Grains in Aluminum through Severe Plastic Deformation and Subsequent Annealing

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In order to understand how the ultrafine grained structures form, microstructure evolution in a pure aluminum during ultrahigh straining and subsequent annealing processes is studied in the present paper. The microstructures and crystallographic features of a commercial purity aluminum heavily deformed by ARB process and then annealed were precisely analyzed by FE-SEM/EBSD and TEM. It was confirmed that the formation of ultrafine grains during heavy deformation could be understood in terms of *grain subdivision*, where deformation induced boundaries subdivided the original crystal. In the pure Al deformed to ultrahigh strains, rather uniform changes of microstructures continuously happened during subsequent annealing. The process might be called *continuous recrystallization*, but it was rather close to *normal grain growth* accompanied with recovery of the elongated ultrafine grains formed by ultrahigh-strain plastic deformation.

Keywords: *severe plastic deformation, grain subdivision, recovery, continuous recrystallization, grain growth.*

1. Introduction

It is well known nowadays that ultra-high straining, or severe plastic deformation (SPD), of metallic materials realizes ultrafine grained (UFG) microstructures with mean grain sizes much smaller than one micrometer [1,2]. On the other hand, the formation mechanism of the UFGs has not yet been fully and commonly understood. Especially in Al and Al alloys, UFG structures are often realized in the as-deformed state without annealing, which is completely different from conventional deformation and recrystallization mechanism for producing fine grained structures. In the present paper, detailed results of microstructure evolution during SPD and subsequent annealing processes are shown and the mechanisms of the UFG structure evolution are discussed.

2. Experimental

A commercial purity aluminum (JIS-1100, 99% purity) was used in the present study. Fully recrystallized 1100 sheets 1 mm in thickness, 40 mm in width and 250 mm in length were provided to ultra-high straining by accumulative roll bonding (ARB) process. The ARB is a SPD process using rolling deformation, where stacking, roll-bonding and cutting of sheet are repeated [3,4]. The detailed procedures of the ARB process have been reported elsewhere [1-4]. Two pieces of the 1mm thick sheets of 1100-Al were stacked after surface treatments, and the ARB process using 50% one-pass roll-bonding per cycle was carried out at room temperature without lubrication. A two-high rolling mill with a roll diameter of 310 mm was used. The ARB process was repeated up to 6 cycles. The sheets ARB processed by 6 cycles were annealed at various temperatures for various periods. The microstructures and microtextures were observed by transmission electron microscopy (TEM) using Hitachi H-800 operated at 200 kV and by electron back-scattering diffraction (EBSD) analysis in a scanning electron microscope equipped with a field-emission type gun (FE-SEM) operated at 15 kV. TSL-OIM system was used for the EBSD measurement and analysis.

3. Results and Discussions

3.1 Microstructure evolution during ultrahigh strain deformation

Figure 1 shows TEM microstructures of the 1100-Al sheets ARB processed by 1, 3, 5 and 6 cycles at room temperature (RT). The microstructures were observed from the transverse direction (TD) of the sheets and the observed locations were near the thickness center of the sheets. Selected area diffraction (SAD) patterns are also represented in the figures. The 1-cycle ARB specimen, which is equivalent to 50% cold-rolled material, shows typical dislocation cell structures (Fig.1(a)). The SAD pattern is still spotty, though some degrees of striation are recognized. With increasing the number of the ARB cycle, i.e., equivalent strain, lamellar structures elongated to the rolling direction (RD) develop more and more. After 6 cycles of the ARB process, the specimen is filled with the elongated lamellar structure (Fig.1(d)). The lamellar structure is fairly homogeneous, and the mean interval of the lamellar boundaries along the normal direction (ND) of the sheets is about 200 nm. The SAD pattern is ring-like, which suggests large misorientation has developed in this structure. In fact, detailed crystallographic analysis using Kikuchi-line diffractions in TEM has

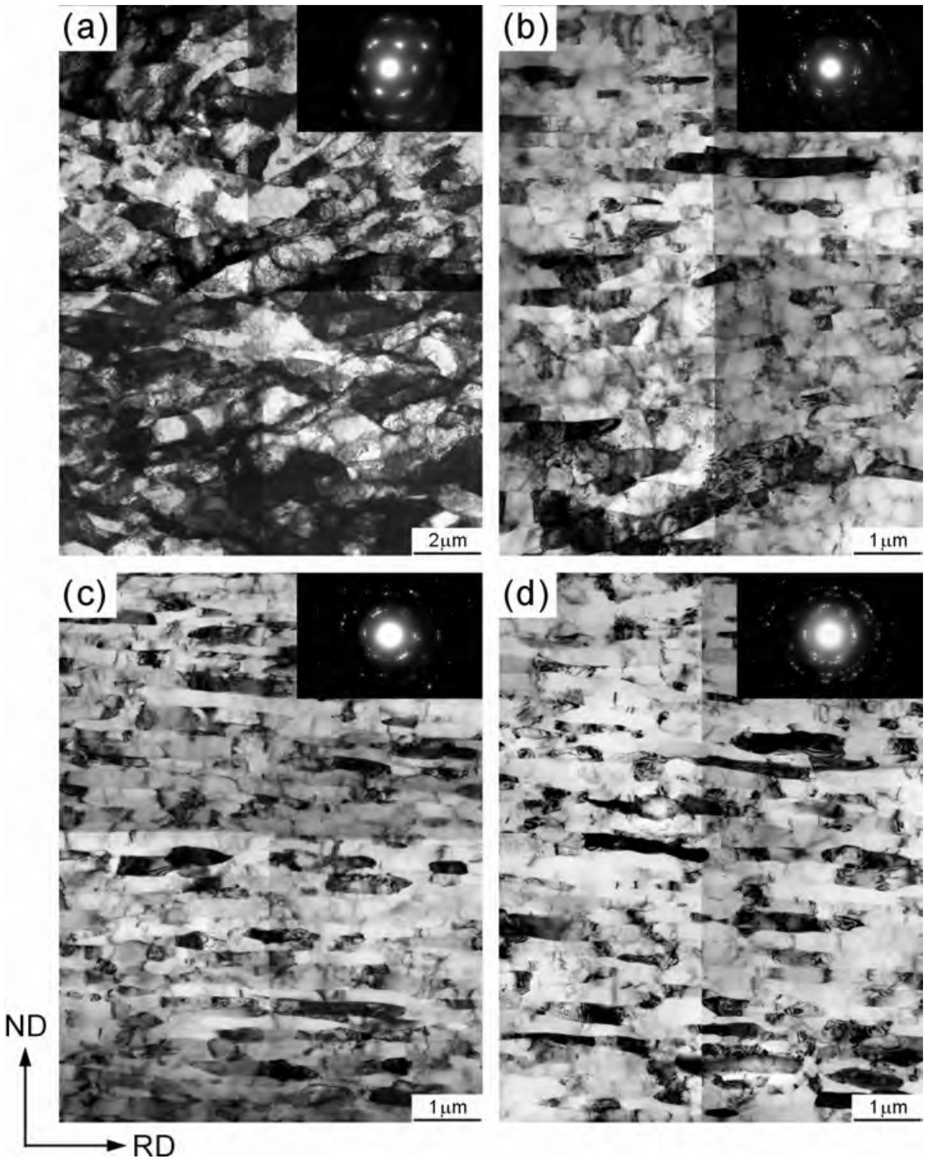

Fig.1 TEM microstructures and corresponding SAD patterns of the 1100-Al ARB processed by (a) 1 cycle, (b) 3 cycles, (c) 5 cycles and (d) 6 cycles at room temperature without lubrication.

clarified that the lamellar boundaries formed after 5-6 ARB cycles are mostly high-angle grain boundaries having large misorientation [5]. Thus, from a viewpoint of misorientation, the microstructure can be considered as ultrafine 'grains'. This is a typical UFG structure observed in as-ARB processed specimens. It should be emphasized, however, that the microstructure is essentially a deformation microstructure, as the morphology is elongated along the deformation direction and there are many dislocations and sub-boundaries (low-angle boundaries) inside the elongated grains.

The evolution of the structure can be understood in larger scales through FE-SEM/EBSD analysis. Figure 2 shows boundary maps obtained from EBSD analysis of the 1100 specimens ARB processed by 1, 2, 4 and 6 cycles at RT. The orientation mapping was carried out at different thickness locations in the identical specimen. The thickness locations are indicated as t/t_0, which indicates the position from the center normalized by the total thickness (t_0) of the sheet (~1 mm) [6]. Therefore, $t/t_0=0$ means thickness center, $t/t_0=0.25$ the quarter thickness, and $t/t_0\sim0.5$ the subsurface. The green lines in Fig.2 represent high-angle grain boundaries having misorientation larger than 15°, while red lines low-angle boundaries with misorientation between 2° and 15°. The boundaries having misorientation smaller than 2° were cut off in Fig.2, in order to remove inaccuracy due to automatic EBSD measurement and analysis. When looking at the quarter thickness ($t/t_0=0.28$-0.29),

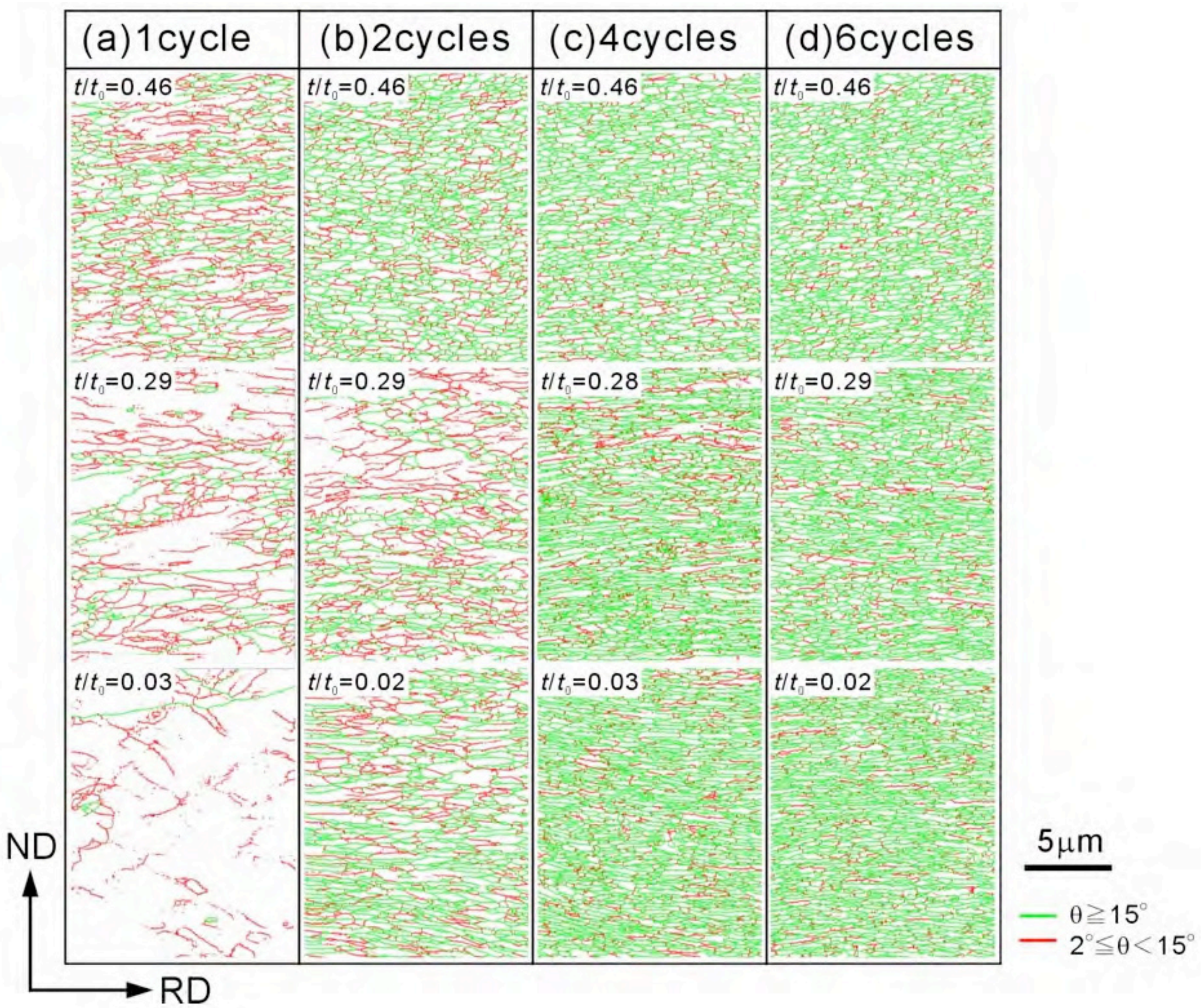

Fig.2 EBSD boundary maps at various thickness locations (t/t_0) in the 1100-Al ARB processed by various cycles at room temperature without lubrication.

the microstructure evolution with increasing strain can be typically understood. In the specimen ARB processed by 1 cycle, the boundary map is mostly composed of low-angle boundaries (red lines). Most of the high-angle boundaries (green lines) are considered to be initial grain boundaries. That is, this boundary map shows typical deformation microstructure after 50% rolling, which is composed of initial grains elongated to RD and some low-angle boundaries developed within the initial grains. The TEM microstructure, Fig.1(a), shows a more detailed view of the dislocation structures within an initial grain. After 2 ARB cycles, the boundary map seems to be a deformation microstructure still, but the number of high-angle grain boundaries increases. Especially, some new high-angle boundaries are introduced near initial grain boundaries. With increasing the ARB strain, the density of high-angle boundaries (green boundaries) increases, and eventually the boundary map is uniformly filled with the lamellar green boundaries elongated to RD. This is totally the same as the lamellar boundary structure observed in TEM (Fig.1(d)). The average interval of high-angle boundaries (green boundaries) in the EBSD boundary map (Fig.2) is also ~200 nm.

The evolution of ultrafine structures is much more accelerated at subsurface location (t/t_0=0.46) than that at quarter thickness in Fig.2. This is because the present ARB process was carried out without lubrication, and large amount of redundant shear strain caused by friction between the rolls and the sheet was introduced in the subsurface regions [7,8]. The microstructure evolution at center location is also faster than the quarter thickness after the second cycle, since in the ARB process the roll-bonded sheet is cut and stacked so that half of the surfaces come to the center in the next cycle. The final structures after large plastic strains are, however, the similar microstructures composed of the elongated UFGs, independent of the thickness locations.

After the observations shown in Figs.1 and 2, we conclude that the evolution of the UFG structure during the ARB (during SPD processes in general) can be understood in terms of *grain subdivision* [9,10]. The process of *grain subdivision* is illustrated in Fig.3. Plastic deformation of metallic crystals are borne basically by dislocation slips. In polycrystalline materials, the slip patterns (combination and number of slip systems operated) can differ depending on locations even within an identical crystal. Different slip patterns result in different crystal rotation, to produce misorientation between neighboring regions. The boundaries between such regions to bear the misorientation is called *geometrically necessary boundaries* (*GNBs*) [9,10]. In addition, dislocations stored in the crystal tend to form low-energy configurations. The dislocation

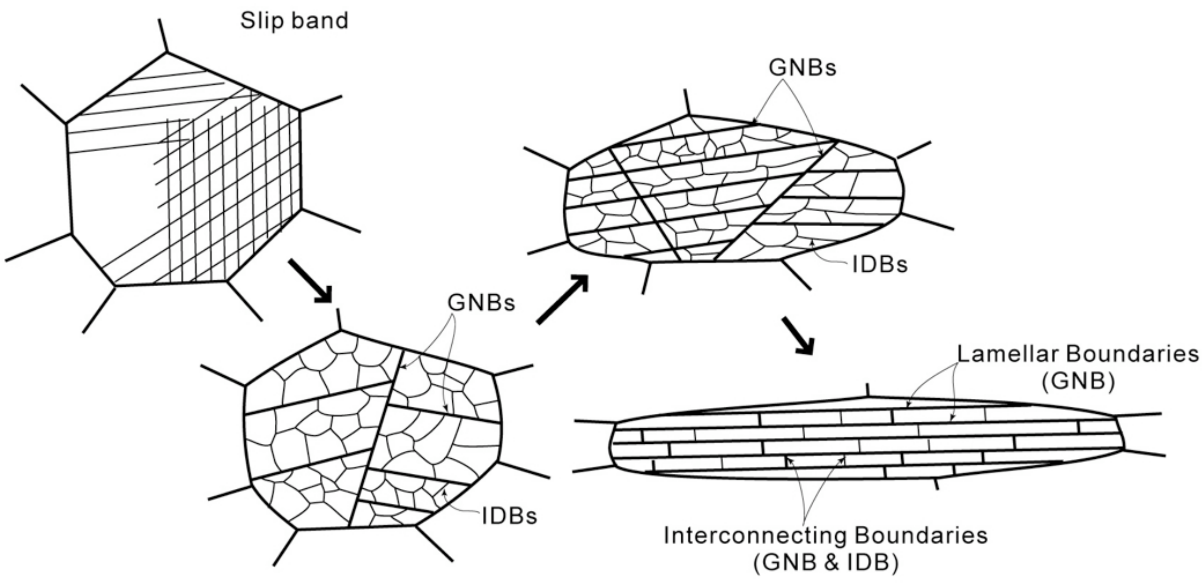

Fig.3 Schematic illustration showing *grain subdivision* mechanism during plastic deformation.

boundaries formed by such dislocations are called *incidental dislocation boundaries* (*IDBs*) [9,10]. The original grain (crystal) is finely subdivided by the *GNBs* and *IDBs* with increasing plastic strain, that is the *grain subdivision* mechanism. Especially *GNBs* are important for the formation of UFGs, as their misorientation increases with increasing plastic strain applied [9,10]. So-called lamellar boundary structure has been reported in heavily cold-rolled aluminum [9], where lamellar boundaries are *GNBs*. The ultrafine lamellar structures observed in the present ARB specimens (Fig.1(c,d)) are quite similar to the lamellar boundary structure reported in heavily cold-rolled aluminum, though the fraction of high-angle boundaries in the ARB specimens is much higher than that in the cold-rolled one [5]. Because the mechanism of microstructure evolution is *grain subdivision*, it can be also said that the UFG structures produced by ultrahigh straining of aluminum are essentially deformation microstructures, though the subdivided regions (grains) have large misorientation to each other. This agrees well with the fact that the UFG microstructures obtained through the ARB involves many dislocations and low-angle grain boundaries (Figs.1 and 2).

3.2 Microstructure evolution during subsequent annealing

Change in TEM microstructures during annealing of the 1100-Al ARB processed by 6 cycles are shown in Fig.4. The specimens were annealed at various temperatures for 1.8 ks. In low-temperature annealing at 150°C (Fig.4(b)), recovery happens to decrease dislocation density within the elongated grains, and the elongated structure slightly coarsens through grain boundary migration. The microstructure coarsens with increasing the annealing temperature, and after annealing at 250°C, nearly equiaxed grain structure is observed (Fig.4(d)). The grains are free from dislocations, so that it is difficult to distinguish them from conventionally recrystallized grains. However, the mean grain size in Fig.4(d) is still around 1 μm, which cannot be realized through conventional deformation and recrystallization. It is clear from Fig.4 that the microstructural change during annealing is uniform and continuous, and no nucleation and growth of particular grains are observed.

The tendency is the same in more macroscopic grain boundary maps obtained by EBSD analysis. Figure 5 summarizes the orientation color maps indicating crystallographic orientation parallel to ND or RD of the sheets and the grain boundary maps drawn in the same way as Fig.2 of the 1100-Al specimens ARB processed by 6 cycles and then annealed at various temperatures for 1.8 ks. The boundary maps in Fig.5 indicate that the change in microstructures during annealing is similar to that observed in Fig.4, also in larger areas. The ND and RD color maps maintain similar tendency of colors, respectively, independent of annealing temperatures, which suggests that the texture of the material does not change significantly. Such a continuous change of the grain structure is sometimes called *continuous recrystallization* [11], since it is greatly different from

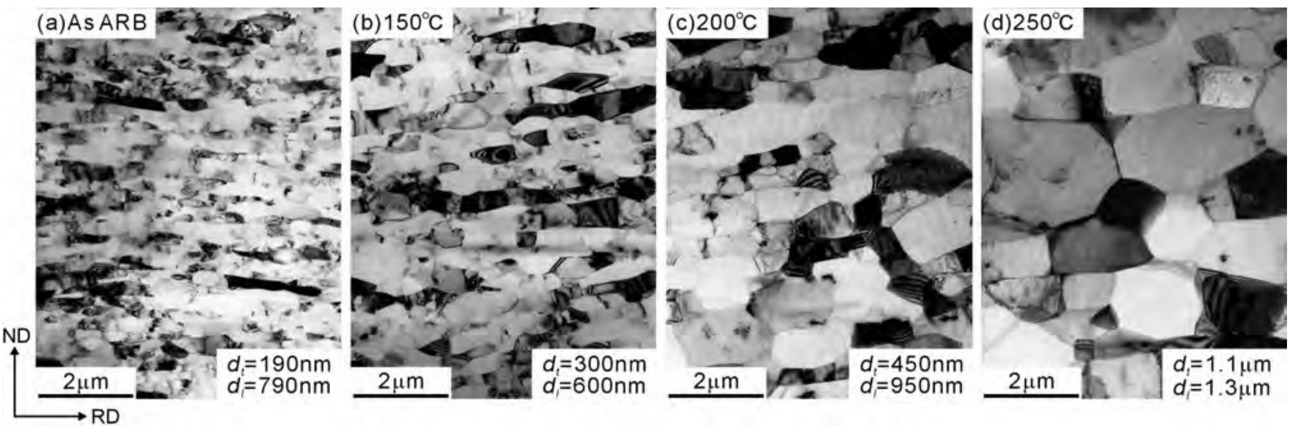

Fig.4 TEM microstructures of the 1100-Al ARB processed by 6 cycles at RT without lubrication and then annealed at various temperatures for 1.8 ks.

conventional recrrystallization characterized by nucleation and growth of particular grains (*discontinuous recrystallization*). To keep texture is a characteristic of *continuous recrystallization*. However, even in conventional recrystallization (*discontinuous recrystallization*), it is considered that no new recrystallized grains nucleate through thermal fluctuation of atoms but potential nuclei existed in the deformed microstructure preferentially grow in annealing [12]. That is, recrystallization is essentially a continuous process from the deformed microstructure. In that sense, it might be confusing to use the term, *continuous recrystallization*. In the materials deformed to ultrahigh strains, the structure finely subdivided in submicrometer dimensions is already full of high-angle grain boundaries, as is shown in Fig.2 or Fig.5(a). Thus, it is rather suitable to consider that the microstructural change during annealing of SPD processed materials (Figs.4 and 5) is a kind of grain growth accompanied with recovery at grain interior. Actually, growth of particular grains can happen even in the same ARB processed 1100-Al under certain annealing conditions, as shown in Fig.6. This looks like conventional (discontinuous) recrystallization by nucleation and growth, or *abnormal grain growth*. The microstructural change shown in Figs.4 and 5 are much more like *normal grain growth*.

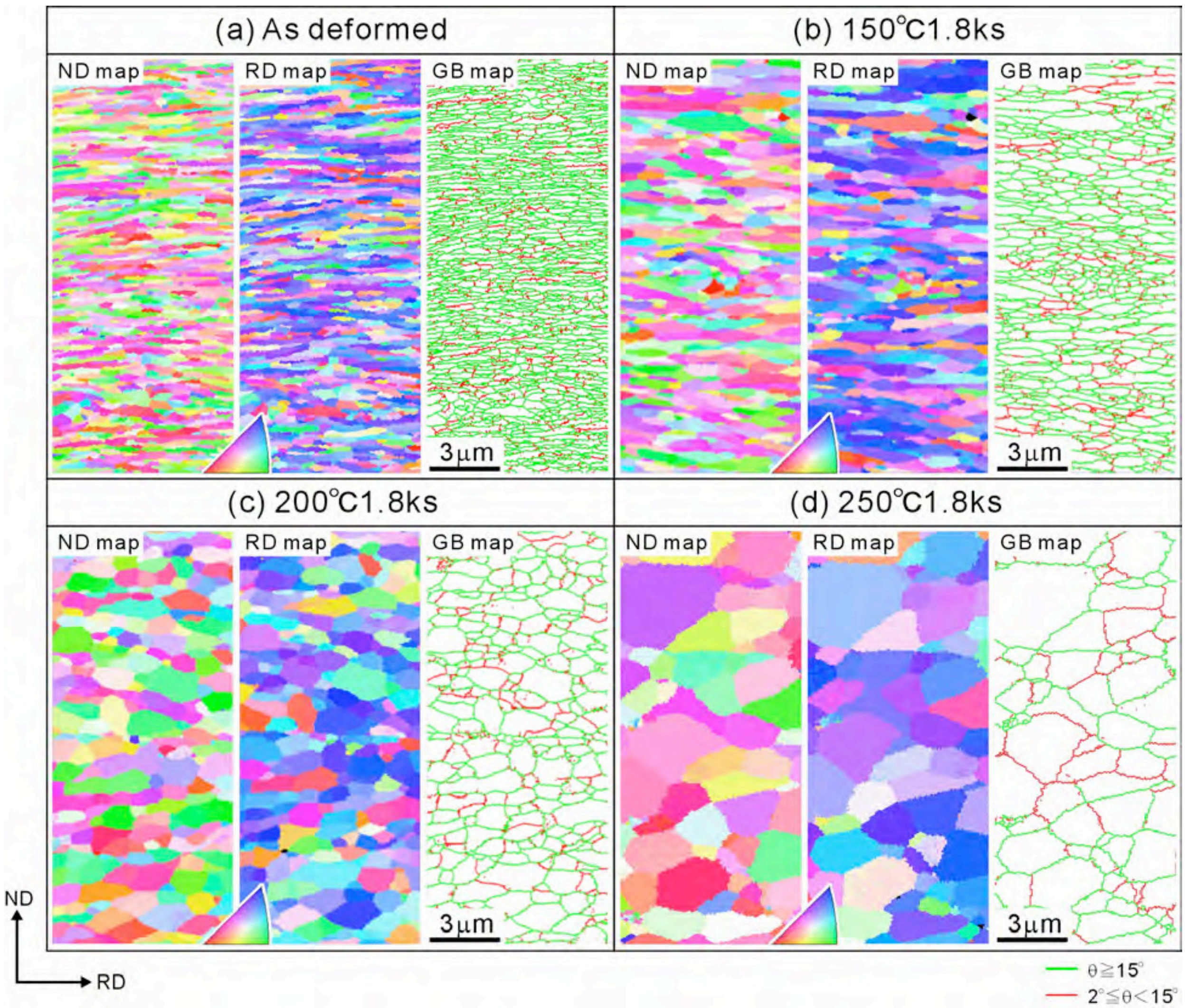

Fig.5 ND and RD orientation color maps and boundary maps obtained from the EBSD measurement of the 1100-Al ARB processed by 6 cycles at RT without lubrication and then annealed at various temperatures for 1.8 ks.

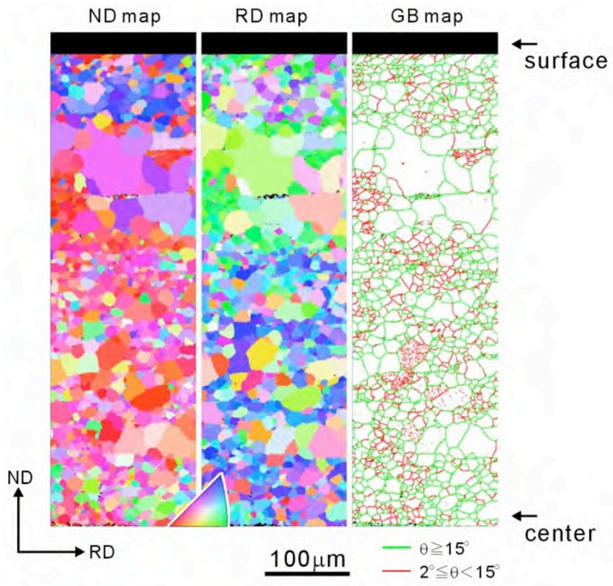

Fig.6 ND and RD orientation color maps and boundary map obtained from the EBSD measurement of the 1100-Al ARB processed by 6 cycles at RT without lubrication and then annealed at 400°C for 1.8 ks.

4. Conclusions

Microstructural evolution of a commercial purity aluminum during ultrahigh straining and subsequent annealing processes was studied in details. The formation of ultrafine grains in severe plastic deformation (ultrahigh straining) can be understood in terms of *grain subdivision*. As a result, the obtained ultrafine grains essentially had characters of deformation microstructures, like elongated morphologies and large amount of dislocations and sub-boundaries, although the elongated ultrafine grains have large misorientation to each other. When the highly deformed pure aluminum is annealed, recovery and grain boundary migration gradually occurred to result in rather continuous changes in the grain structures. This may be called *continuous recrystallization*. However, it is better to consider that the microstructural change during annealing is a kind of *normal grain growth* accompanied with recovery at grain interior. Under certain annealing conditions, inhomogeneous growth of particular grains (*abnormal grain growth*) happened even in the ultrahigh strained pure aluminum.

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