Microstructures and Mechanical Properties of Ultra Low Carbon Interstitial Free Steel Severely Deformed by a Multi-Stack Accumulative Roll Bonding Process*

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An ultra low carbon interstitial free (IF) steel was severely deformed by the six-layer stack accumulative roll-bonding (ARB) process for improvement of the mechanical properties. As-received material with 1 mm in thickness showed a recrystallization structure with average grain diameter of 27 μm. The ARB was conducted at ambient temperature after deforming the as-received material to 0.5 mm thick by cold rolling. The ARB was performed for six-layer stacked, i.e. 3 mm thick sheet, up to 3 cycles (an equivalent strain of ~7.1). In each ARB cycle, the stacked sheets were, first, deformed to 1.5 mm thick by the first pass, and then reduced to 0.5 mm thick, equals to the starting thickness, by multi-pass rolling without lubrication. The specimen after 3 cycles of ARB was annealed for 1.8 ks at various temperatures ranging from 673 K to 1073 K. The tensile strength of the ARB processed materials increased largely with the number of ARB cycles, after 3 cycles it reached a maximum of 1.12 GPa, which is about 4 times larger than that of the initial material. The elongation dropped largely after the cold rolling prior to the ARB, however it remains almost constant during the subsequent ARB process. Transmission Electron Microscopy revealed that the ARB processed materials exhibited a dislocation cell and/or subgrain structure with relatively high dislocation density. The selected area diffraction (SAD) patterns suggested that the orientation difference between neighboring cells was very small. The annealing up to 873 K resulted in gradual decrease in the strength due to the static recovery. The annealing above 873 K resulted in recrystallization and normal grain growth, and thereby a significant drop in the strength and recovery in ductility.

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1. Introduction

In recent years, ultrafine grained (UFG) materials have received considerable attention from many researchers for their potential improvements in mechanical properties. These materials have been processed using severe plastic deformation techniques such as equal channel angular pressing (ECAP),1–3) high-pressure torsion (HPT),4) conshearing,5) and accumulative roll-bonding (ARB).6–20) etc. Among such techniques, the ARB process is mostly appropriate for practical applications to flat products because it can be carried out readily by the conventional rolling system. It has been demonstrated that the significant strengthening through the ARB process could be achieved in various kinds of aluminum alloys and steels. In the way, to date, the ARB has been done mostly by two-layer stacking method where two sheets with same dimensions was roll-bonded by 50% reduction in thickness. In that case, the ARB of at least 5 cycles (an equivalent strain of 4.0) was required for ultrafine grains to cover almost all over the materials. However, there is no restriction, in principle, on the number of stacking layers in ARB process. If the number of the stacking layers would be increased, some advantages as follow are expected,18) first, the productivity would be increased due to the simplification of ARB processing, secondly, the bonding strength between the stacked layers would be improved due to heavy draft, thirdly, it is possible to introduce more severe shear deformation to the materials. Therefore, multi-stack ARB is expected to be a more powerful and practical process for attaining ultra grain refinement and high strengthening in bulk metallic materials. Actually, it has been demonstrated that the six-layer stack ARB is an effective process for ultra grain refinement and strengthening in the case of commercial purity aluminum.18)

In this study, an ultra low carbon interstitial free (IF) steel is severely strained by six-layer stack ARB process. Ultra low carbon IF steel is used widely for automotive industry because of its excellent deep drawability. Application of the ARB process to the IF steel would result in increase in the strength, and thereby weight-saving of the automotive. Tsuji et al. have applied the ARB process to IF steel at 773 K and succeeded in ultra grain refinement and high strengthening,9) and furthermore clarified the effects of strain on microstructures and mechanical properties of the ARB-processed and annealed IF steel.10) However, their studies are focused on the ARB at intermediate temperatures. In the ARB, the processing temperature is an important factor in formation of the UFGs. The author et al.11) have studied on the ARB of the IF steel at ambient temperature and clarified the difference in microstructure and mechanical properties between IF steels ARB-processed at ambient temperature and 773 K. In addition, it was found that the ARB at ambient temperature is more effective in strengthening than that at 773 K. However, in the case of two-layer stack ARB, it was difficult to introduce a strain above ~4.0 to the IF steel due to insufficient workability. The multi-layer stack ARB would let the larger amount of strain be able to introduce into the IF steel, resulting in further improvement in strength. Besides that, multi-layer stack ARB has some advantages mentioned.

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before. Therefore, this study is aimed to investigate systematically the microstructure and mechanical properties of the IF steel processed by the six-layer stack ARB and subsequently annealed.

2. Experimental Procedures

2.1 Specimen preparation

A Ti-added ultra low carbon IF steel (0.002 mass% C-0.003N-0.072Ti) was used. The as-received material was in a fully annealed state and its dimensions were 1 mm in thickness, 30 mm in width and 300 mm in length. The average grain size of the recrystallized specimen was 27 μm in diameter. The ARB was conducted after deforming the as-received material to 0.5 mm thick by 50% cold rolling. The schematic illustration of the six-layer stack ARB process is shown in Fig. 1. In the beginning of the ARB, degreasing and wire-brushing were treated on the surfaces of each sheet for sufficient bonding. The six sheets were then stacked to each other. The six-layer sheet 3 mm thick was deformed to 1.5 mm thick by 50% cold rolling, and then reduced to 0.5 mm thick, as the starting thickness, by multi-pass rolling using a 2-high mill with roll diameter of 310 mm at roll speed of 0.29 m/s without lubrication. The rolled specimen was cut to six pieces of same length with a guillotine, and then supplied to the next cycle. The same procedure was then repeated, up to three cycles (an equivalent strain of 7.1) at ambient temperature. The equivalent strain is estimated from the change in thickness under plane-strain condition. The specimen after 3 cycles was annealed for 1.8 ks at various temperatures ranging from 673 to 1073 K.

2.2 Testing and characterization

The microstructure was investigated by transmission electron microscopy (TEM). TEM studies were conducted with a HITACHI H-800 microscope operated at 200 kV. Thin disk-shaped foils parallel to the rolling plane were prepared by spark machining and twin-jet polishing for TEM observation. The mechanical properties of the ARB-processed and subsequently annealed specimens were determined at ambient temperature by an Instron-type tensile testing machine. The test pieces were spark-machined so that the tensile direction was parallel to the rolling direction. The gauge length was 10 mm and the gauge width was 5 mm. The initial strain rate was \(8.3 \times 10^{-4} \text{s}^{-1}\).

3. Results and Discussion

3.1 Microstructure

3.1.1 As ARB-processed IF steel

Figure 2 shows TEM micrographs and the corresponding selected area diffraction (SAD) patterns observed at the plane perpendicular to normal direction (ND plane) of IF steel before and after six-layer stack ARB process. The SAD patterns were taken from the center of the bright field images by using an aperture with diameter of 1.6 μm. The specimen before the ARB exhibits a dislocation cell structure with relatively very high dislocation density because it was deformed by 50% cold rolling, as shown in Fig. 2(a). The corresponding SAD pattern, viewed from the \((111)\) zone axis, shows a single net pattern. This suggests that the orientation difference between neighboring cells hardly exists. The specimen after 1 cycle (an equivalent strain of \(\sim 3.0\)) also exhibits a dislocation cell structure similar to the specimen before ARB [Fig. 2(b)]. However, the cell walls became sharper and the corresponding SAD pattern, viewed from the \((011)\) zone axis, is characterized by slightly diffused spots. The specimen after 2 cycles (an equivalent strain of \(\sim 5.0\)) exhibits still a dislocation cell structure in which the walls became quite sharp and at the same time the interiors became even more free of dislocations. In addition, the cell size is smaller than those of the previous ones and the subgrains are seen partially in the structure, as indicated by the arrow [Fig. 2(c)]. In addition, the SAD pattern exhibits many extra spots, indicating that the misorientation between the cells and/or subgrains is larger than those of the previous specimens. The specimen after 3 cycles (an equivalent strain of \(\sim 7.1\)) has also a microstructure similar to that after 2 cycles, as shown in Fig. 2(d). Well-defined UFGs often seen at ARB-processed metals were not observed in this study. This result is very similar to that of IF steel processed by two-layer stack ARB at ambient temperature.\(^{11}\) However, it is quite different from the case of aluminum in which the UFGs developed even at the first cycle.\(^{18}\) Where did the difference between two materials come from? The formation mechanism of the UFGs by the ARB process is known as the continuous recrystallization (the recrystallization \(\text{in situ}\)) characterized by subdivision to UFG by severe deformation, recovery to form clear UFGs, and short-range grain boundary migration.\(^{20}\) That is, the recovery is necessary for formation of the UFGs by the ARB process. In the case of aluminum, the recovery can occur due to temperature rise by severe plastic working and relatively high homologous temperature even at ambient temperature. However, in the case of IF steel, the recovery is hard to occur at ambient temperature because the temperature rise by working is not enough to induce the recovery in IF steel. Therefore, the results of the present study support the indispensability of the recovery for formation of the UFGs.

3.1.2 After annealing

Change in TEM microstructure observed at ND plane of the specimen ARB-processed by 3 cycles with annealing temperature is shown in Fig. 3. As shown in Fig. 3(a), the UFGs with the average grain size of 400 nm and clear grain boundaries formed partially after annealing at 773 K, as indicated by the arrow. However, lots of dislocations are seen inside the UFGs and the grains are elongated along a certain
direction, supposedly the rolling direction, suggesting that
their formation mechanism is different from that of the
conventional recrystallization. The sample annealed at 823 K
also shows a structure containing the UFGs, however the
fraction of the UFGs is higher and the grain diameter
(\sim 800 \text{ nm}) is larger than those of the sample annealed at
773 K. In addition, it still exhibits very high dislocation
density. However, as shown in Fig. 3(c), the specimen
annealed at 873 K, exhibits quite different structure, com-
pared to the samples annealed at temperatures below 823 K.
That is, it has a structure similar to conventional recrystal-
lization structure, i.e. equiaxed and dislocation-free grains.
The average grain size (\sim 1.4 \mu m) is larger than that of the
sample annealed at 823 K. As shown in Fig. 3(d), the
annealing at 923 K resulted in normal grain growth.

3.2 Mechanical properties
Nominal stress-strain curves of the as-received and as
ARB-processed IF steel are shown in Fig. 4. The as-received
one shows relatively low strength and large elongation. The
tensile strength increases with increasing a number of ARB
cycles and reaches a maximum of 1.12 GPa after the 3rd
cycle, which is 4 times larger than the initial material. It is
notable that even the IF steel with less alloying element
shows such outstanding strength above 1 GPa and that it can
be attained by 3 cycles of the ARB. This increase in strength
is considered to be primarily due to work hardening. On the
other hand, the elongation dropped largely by 50\% cold
rolling and remained almost constant during the ARB. It is
found from the s-s curves that the uniform elongation is very
low, which is a common characteristic of severely deformed
materials. However, the phenomena like “yield-drop” often
appeared at the ARB processed materials\cite{14,20} did not appear
in this study. This let us suppose that the yield-drop
phenomena are associated with the existence of the UFGs.

Figure 5 shows the mechanical properties plotted as a
function of the equivalent strain (the number of ARB cycles).
For comparison, the results of the IF steel two-layer stack
ARBed at 773 K\cite{9} and ambient temperature\cite{11} are plotted
together in Fig. 5. It is found that the tensile strength
increases with increasing the equivalent strain at any cases.
The strength increases rapidly with the equivalent strain at
the initial stage, primarily due to strain hardening at any
cases. However, the strength increment with the strain is
different in each case, especially at large equivalent strain
above \sim 2. The difference in the strength between the two
layer stack ARBed ones would be primarily due to the
different strain hardening rate associated with ARB process-
ing temperature.\cite{11} The difference in the strength due to the
different ARB stack-layer number is thought to be caused by
the additional shear strain, so-called, the redundant shear
strain, introduced beneath the surface of material due to the

![Fig. 2 TEM micrographs of IF steel severely deformed by six-layer stack ARB. Observed from ND plane before ARB (a), after 1 cycle
(b), 2 cycles (c) and 3 cycles (d) respectively.](image-url)
large friction between rolls and workpiece. On the other hand, it is found at any cases that the elongation dropped largely to below 10% above an equivalent strain of $\varepsilon_{0}$.

Figure 6 shows the change in nominal stress-strain curves with the annealing temperature. The tensile strength decreased and the total elongation increased gradually with raising the annealing temperature up to 873 K. Annealing at 923 K caused the drastic increase in the elongation, accompanying with the large decrease in the strength. This corresponds well with the grain growth at 923 K, as shown in Fig. 3(d). This indicates that it is necessary to anneal above 923 K for obtaining sufficient uniform elongation. The softening curves of the as-ARB processed IF steel against annealing temperatures is plotted in Fig. 7. For comparison, the results for two-layer stack ARB at ambient temperature are plotted by dotted lines in the figure. The strength of specimen annealed at 673 K is lower than that of the as-ARB processed one. Annealing at temperatures above 673 K resulted in gradual decrease in the strength. As a result, the strength dropped largely to about 200 MPa after annealing at 973 K. As shown in the figure, a decrease in strength with annealing temperature is larger in six-layer stack ARBed one than in two-layer stack ARBed one. This is probably caused by the difference in total strain (or stored energy) introduced by the ARB process. That is, an amount of total strain introduced by the ARB was larger in the six-layer stack ARB (~7.1), comparing to the two-layer stack ARB (~4.0). Therefore,

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**Fig. 3** TEM micrographs of IF steel annealed at various temperatures after 3 cycles of six-layer stack ARB. Observed from ND plane after annealing at 773 K (a), 823 K (b), 873 K (c) and 923 K (d) respectively.

**Fig. 4** Nominal stress-strain curves of IF steel processed by six-layer stack ARB.

**Fig. 5** Changes in mechanical properties of IF steel with equivalent strain applied by six-layer stack ARB.
the released energy during annealing would be larger in the six-layer stack ARBed one, resulting in larger decrease in the strength. In the way, these softening behaviors are very different from the conventional behaviors accompanying drastic drop in strength at a certain annealing temperature. The turning point in softening behaviors is 873 K in both ARB processes. That is, the decrease in the strength at annealing temperatures ranging from 673 to 873 K is due to the static recovery, however it at temperatures above 873 K is caused by recrystallization and grain growth. On the other hand, the change in total elongation with annealing temperature is very similar to each other. That is, the total elongation hardly changed during annealing up to 673 K, however it increased gradually at temperatures above 773 K and rise considerably above 873 K. In addition, it is found from the figure that the annealing above 873 K is required for recovery of the uniform elongation. This mechanical behavior corresponds well with the microstructural change with the annealing temperature.

4. Conclusions

Six-layer stack accumulative roll bonding (ARB) process was applied to an ultra low carbon IF steel, the results obtained are summarized as follows,

(1) The six-layer stack ARB up to 3 cycles was successfully performed at ambient temperature, and large equivalent strain of ~7.1 was introduced to the specimen.

(2) The tensile strength of the as-ARB processed IF steel increases largely with the number of the ARB cycles, reaches a maximum of 1.12 GPa at the 3rd cycle, which is about 4 times greater than that of the starting material. On the other hand, the elongation drops largely by the 50% cold rolling prior to the ARB, and it remains almost constant during the subsequent ARB process.

(3) The as-ARB processed materials show primarily a dislocation cell and/or subgrain structure with high dislocation density, which is very similar to those processed by two-layer stack ARB at ambient temperature.

(4) The annealing up to 873 K results in gradual decrease in the strength due to the static recovery. At higher temperatures above 873 K, the conventional recrystallization and normal grain growth occurred, resulting in a significant drop in the strength and recovery in ductility. This softening behavior is very similar to that of two-layer stack ARB. However, a decrease in strength with annealing temperature is larger in six-layer stack ARBed one than in two-layer stack ARBed one.

REFERENCES