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Thermal stability of aluminum cold rolled to large strain

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Abstract Common to metals deformed to high strains is a very fine microstructure, high strength, and limited ductility. Structure and property optimization by annealing after deformation must, therefore, be explored. In the present study, commercial purity aluminum has been annealed after cold rolling to ultrahigh strains up to $\varepsilon_{vM} = 6.2$ and annealing processes have been studied in terms of recovery and conventional recrystallization. These processes have been analyzed by isochronal and isothermal annealing in the temperature range 140–420 °C. It has been found that the recrystallization temperature is a little affected by the rolling strain, whereas the rate of recovery and the temperature range over which recovery takes place increase significantly as the strain is increased. These observations are discussed as to how they can guide studies of nanostructured metals processed by plastic deformation.

Introduction

Nanostructured metals or nanometals produced by plastic deformation to very high strains are characterized by a fine microstructure and a large content of stored energy [1–3]. Their thermal stability is, therefore, reduced, which can be illustrated by the observation that a heavily cold drawn copper wire partly recrystallizes when stored for long time at ambient temperature [4]. The deformed nanometals are also characterized by a high strength and low ductility i.e., their thermal stability is important as optimization may be sought through an annealing treatment which, however, must not lead to an excessive loss in strength. An important step on the route to application of deformed nanometals is therefore to characterize and model the effect of annealing conditions on their structure and properties. Such studies may involve many material and process parameters, but the present article will focus on commercial purity aluminum deformed by rolling from medium to a maximum strain of $\varepsilon_{vM} = 6.2$ (99.5% reduction in thickness). Isochronal annealing treatments have been carried out in air for 2 h at temperatures from 180 to 420 °C and isothermal heat treatments have been carried out for extensive lengths of time at temperatures from 140 to 220 °C and from 245 to 290 °C [5–9]. The structural characterization has been performed by the use of electron back scattering diffraction (EBSD) and transmission electron microscopy (TEM) and the mechanical properties have been determined by hardness measurements and tensile testing. The experimental observations are combined in an analysis of the recovery and recrystallization behavior including the kinetics of such processes.

Experimental

The materials used are aluminum cold rolled (CR) and accumulative roll bonded (ARB), see Table 1. The sample designations given in this table correspond to designations given in previous papers [2, 5, 9] where more detailed annealing studies have been reported. The microstructures

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Table 1 Materials

	Sample	Strain (ε_{vM})	Supplier
AA1200 (99% purity)	CR2	2.3	ALCAN Int.
	CR4	4.6	
AA1050 (99.5% purity)	$\varepsilon = 2$	2.3	Risø National Laboratory
	$\varepsilon = 4$	4.6	
AA1050 (99.5% purity)	ARB1	0.8	Osaka University
	ARB6	6.3	

CR: Cold rolled

ARB: Accumulative roll bonded

of the deformed samples have been characterized in the longitudinal section by EBSD and TEM. The same sample section has been used for the annealed specimens where orientation maps have been taken using an EBSD system attached to a field emission gun scanning electron microscope (SEM) and applying commercial software (channel 5, HKL Technology). The deformed samples have been subjected to isochronal and to isothermal annealing heat treatment in an air furnace at temperatures in the range 140–420 °C. After annealing 10 Vickers hardness measurement have been made on each sample at room temperature with a miniload hardness tester employing a load of 200 g for 10 s. The tensile test data have been supplied by ALCAN International.

Microstructural evolution—deformation

The structural evolution during rolling follows a similar pattern in all the samples. With increasing strain the structure is subdivided by low-to-medium angle dislocation boundaries and high angle boundaries that are the original grain boundaries supplemented with deformation-induced high angle boundaries. The deformation structure evolves with increasing strain from a cellblock structure to a typical lamellar structure with extended boundaries almost parallel to the rolling plane and short interconnecting boundaries. The average spacing between the boundaries decreases and their misorientation angle increases as the strain increases and the fraction of high angle boundaries ($\theta > 15°$) increases and may reach values as high as 60–80% [10]. At all strains the interconnecting boundaries are low-to-medium angle dislocation boundaries. The dislocation density in these boundaries together with the dislocation density of loose dislocations and tangles present between the boundaries is of the order of 10^{14}–10^{15} m^{-2}. This means that although the fraction of high angle boundaries is high the samples have the characteristics of a deformed structure rather than being an ultrafine grain material. The various structural features contribute to the energy stored [1, 3] in

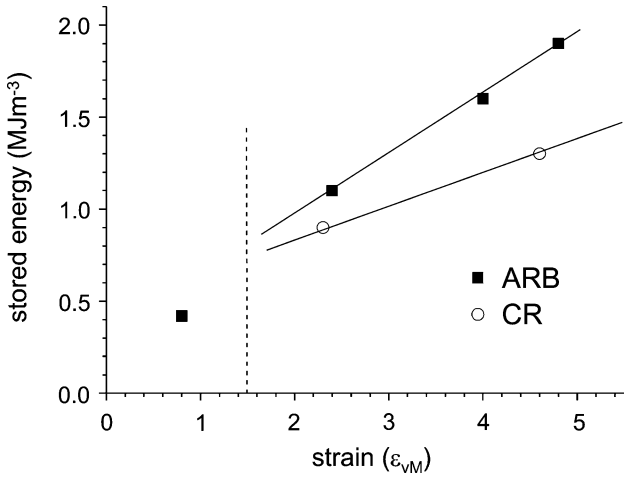

Fig. 1 Stored energy calculated from microstructural measurements as a function of the rolling strain (ε_{vM}) for samples deformed by cold rolling (AA1200) and by accumulative roll bonding (AA1050). Lines have been fitted only for the large strain region $\varepsilon_{vM} > 1.5$ as it has been reported elsewhere that a transition is found in the stored energy/strain relationship at approximately this level [2]

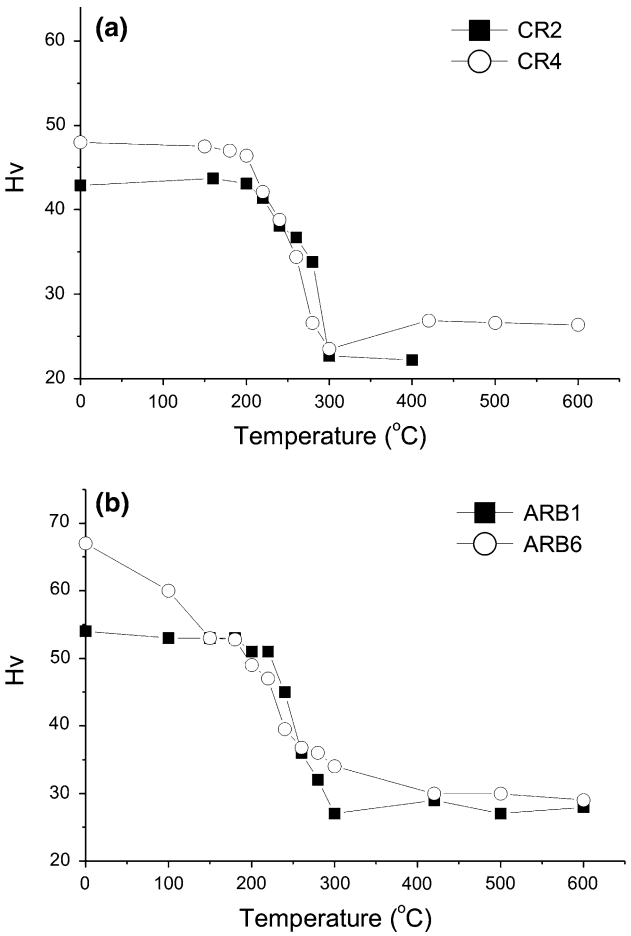

Fig. 2 Change in hardness with annealing temperature for isochronal annealing for 2 h. The hardness in the deformed state is also shown (0°C value) [2]. (**a**) CR2 and CR4. (**b**) ARB1 and ARB6 (see Table 1 for designations)

Table 2 Structural and texture changes during annealing [2]

Sample	F_{HAB} (%)[a]		Rolling texture %	
	As def.	Ann. (2 h/420 °C)	As def.	Ann. (2 h/420 °C)
CR2	34	88	50	15
CR4	54	74	82	59
ARB1	19	89	73	33
ARB6	65	76	81	68

[a] F_{HAB}: Fraction of high angle boundaries

the deformed sample. This stored energy increases linearly with increasing strain [1, 2], see Fig. 1. The combination of a high fraction of high angle boundaries, a high density of dislocations and a large content of stored energy are characteristics of metals deformed to large strains. Another characteristic is that the microstructure is fairly heterogeneous [11]. These structural characteristics are crucial in an analysis of the annealing behavior.

Annealing—recovery and recrystallization

Isochronal annealing treatments for 2 h at increasing temperature lead to softening, microstructural coarsening and a change in the crystallographic texture. The softening is illustrated in Fig. 2 based on the change in hardness with increasing temperature. These figures show that in the low temperature region the softening is more pronounced in CR4 than in CR2 and significantly higher in ARB6 than in ARB1. However, for all samples annealing in the interval 200–300 °C results in rapid softening. The strain level also affects the changes in texture and in the fraction of high angle boundaries when the material passes from the deformed to the annealed state at 420°C. This is illustrated in Table 2, which shows for small and medium strains (CR2 and ARB1) that the annealing leads to a significant increase in the fraction of high angle boundaries and a significant decrease in the concentration of rolling texture components. At large strain (CR4 and ARB6) such changes are much less pronounced. This has led to a suggestion of a possible change from conventional (discontinuous) recrystallization at low-to-medium strains to extended recovery or continuous recrystallization at very large strains [12, 13]. An analysis based on detailed microstructural evidence as exemplified by Fig. 3, however, led to the conclusion for all samples in the present study that the annealing behavior can be characterized as nucleation and growth, i.e.,

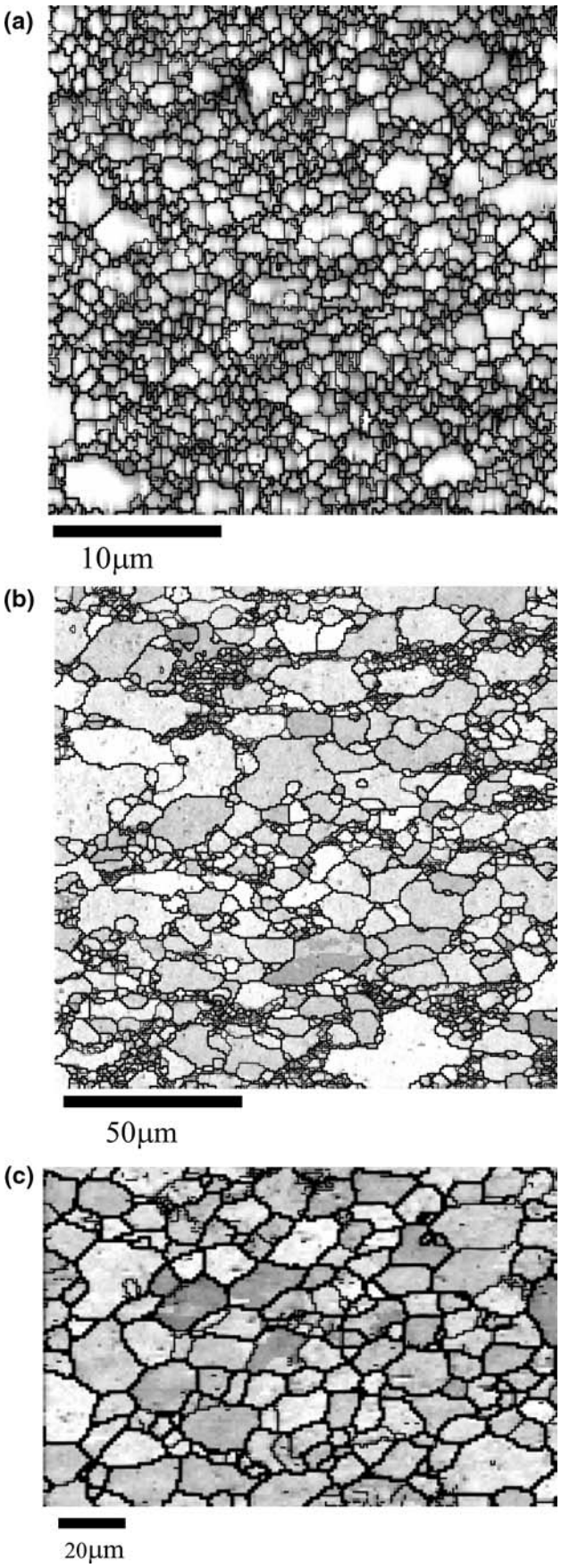

Fig. 3 Microstructure from EBSD measurements for CR4 sample annealed for 2 h at (**a**) 260 °C, (**b**) 280 °C, (**c**) 420 °C. The grey shading represents the EBSD band quality (white = highest pattern quality). (**a**) shows initiation of recrystallization and (**c**) shows a fully recrystallized structure [2]

conventional recrystallization [2, 9], however, with a softening rate which is significantly affected by the strain. This recrystallization leads to a polycrystalline structure with a grain size, which decreases with increasing strains as follows: ARB1 (15.0 μm), CR2 (9.7 μm) CR4 (8.1 μm), ARB6 (7.0 μm) [2]. The isochronal experiments have led to a study of the recrystallization process by carrying out isothermal annealing experiments from 245 to 280 °C. The recrystallization kinetics have been analyzed based on microstructural observations of nucleation and growth and a similar behavior has been found for AA1200 and AA1050. For both strains $\varepsilon = 2$ and $\varepsilon = 4$ the activation energy has been calculated to be about 178 kJ/mol [5, 14] in good agreement with the results in Ref. [15] where the activation energy for recrystallization has been determined to 170 kJ/mol for commercial purity aluminum cold rolled to strain $\varepsilon = 1$ and $\varepsilon = 3$ and annealed in the temperature interval 250–375 °C.

Annealing—recovery only

To study in more detail changes in hardness and microstructure during recovery preceding nucleation and growth, isothermal heat treatment have been carried out at temperatures from 140 to 220 °C over time intervals that range from 2 to 672 h. The results from these studies are summarized in the following for AA1050 cold rolled to $\varepsilon_{vM} = 2.3$ and $\varepsilon_{vM} = 4.6$. The softening during annealing is presented in terms of the fraction R by the expression:

$$R = \frac{H - H_r}{H_d - H_r}$$

where H is the instantaneous hardness after deformation and partial annealing, H_r is the hardness of the fully annealed aluminum, i.e., completely recrystallized and H_d is the hardness of the as-deformed material, i.e., before annealing is carried out. The R versus time curves of the type illustrated in Fig. 4 have been analyzed in order to study recovery only, i.e., the changes in the matrix which do not contain recrystallization nuclei [14]. It has been found that recovery only characterizes specimens which have been heated up to 100 h in the temperature interval 200–220 °C. For longer times recrystallization nuclei start to form with an increasing rate with increasing temperature [14] in the temperature interval investigated. In this experiment also the activation energy for recovery-only has been derived to be significantly lower than the activation energy for recrystallization. It has also been shown that the recovery rate increases significantly as the strain is increased as also illustrated in Fig. 2. The structural changes taking place during recovery have been followed by EBSD and TEM both globally and locally [2, 6–8, 16].

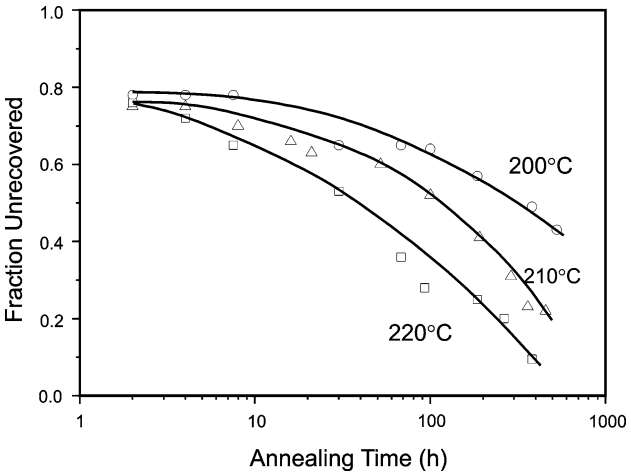

Fig. 4 Fraction unrecovered for aluminum of commercial purity (99.5%) cold rolled to $\varepsilon_{VM} = 4.6$ and heat treated at temperatures 200, 210 and 220 °C

Qualitatively these studies show different recovery processes such as annihilation of dislocations present in low angle boundaries and in the volume between boundaries in parallel with some coarsening and evolution of a more equiaxed structure [2, 8]. These observations point to recovery processes such as coalescence and boundary migration. A coalescence process is illustrated in Fig. 5, where a low angle boundary coalesces into a high angle boundary [17]. The rate of such processes may be accelerated as the strain is increased due to an increase in the stored energy and in the fraction of high angle boundaries leading to the observed increase in the recovery rate. This

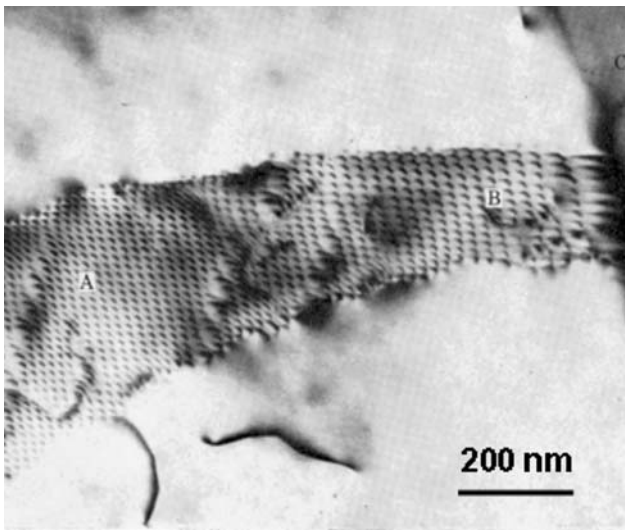

Fig. 5 A bright field TEM micrograph of pure aluminum cold rolled and annealed. A low angle twist boundary (**a**, **b**) is linked to a high angle boundary (**c**, **d**). The spacing within the net of the low angle boundary is found to increase form ca. 27 nm near A to ca. 33 nm in the vicinity of B. The spacing of the dislocation net is even larger in the immediate vicinity of the high angle boundary [17]

correlation between structural parameters and recovery rate leads to a consideration of the effect of structural heterogeneities for example in the form of shear bands and local variations in the deformation texture [2, 8, 11]. Such heterogeneities may lead to local differences in the recovery rate. As an example it has been found in CR2 and CR4 [2, 8] that regions where crystallites have rolling texture orientations recover with a slower rate than regions of other orientations due to a smaller fraction of medium and high angle boundaries in the former.

Mechanical properties

The significant increase in strength following plastic deformation to high strains is counterbalanced by a large decrease in ductility see Table 3 where tensile properties of CR4 are given. In order to optimize properties, annealing after deformation has been explored by heat treatments at temperatures from 150 to 400 °C. This temperature range encompasses both recovery and recrystallization, where 260 °C is the temperature where recrystallization starts and 300 °C is the temperature for completion of recrystallization [2, 5]. The numbers in Table 3 show that to reach a good ductility annealing has to be carried out at temperatures in the range 200–250 °C. However, at such temperatures the softening is pronounced as also observed in previous experiments where commercial purity aluminum has been annealed after large strain deformation by rolling [2, 15] and by accumulative roll bonding [18].

Concluding remarks

High strain deformation of aluminum followed by annealing has extended our general knowledge about processes such as recovery and recrystallization. It has for example been found that conventional or discontinuous recrystallization takes place at all strains in the range $\varepsilon_{VM} = 2.3$ to $\varepsilon_{VM} = 6.2$ and also that the recrystallization temperature is a little affected by the strain in this range. The common finding that the recrystallized grain size decreases with increasing strain is confirmed. As to recovery, an observation of both scientific and technological relevance is that the recovery rate increases with increasing deformation strain and that recovery may take place over a large temperature range (see Figs. 2 and 4). This gives a large window for recovery heat treatments without the initiation of recrystallization. However, the use of a recovery process to optimize strength and ductility is not straightforward and different annealing routes must be explored. One such route is two-step annealing as discussed in another paper at this conference [16]. Also it is a possibility to replace the annealing steps by a slight reduction by cold deformation, which has led to a very satisfactory combination of strength and ductility in aluminum deformed by accumulative roll bonding [19]. This process adds to a number of other routes aimed at optimizing the strength and ductility of nanostructured metals. Common to these different routes is a need for quantification of structural parameters and to explore relationships between structure and mechanical properties. An important part of this research and development will be to investigate the effect of solutes and fine particles on the structure evolution both during deformation and annealing.

Table 3 Tensile strength and total elongation of CR4 after annealing for 2 h

Temperature °C	UTS (MPa)	Elongation (%)
RT	211.4	7.1
150	188.1	3.3
200	157.3	2.5
250	120.4	20.1
300	95.6	29.8
400	96.2	27.6

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