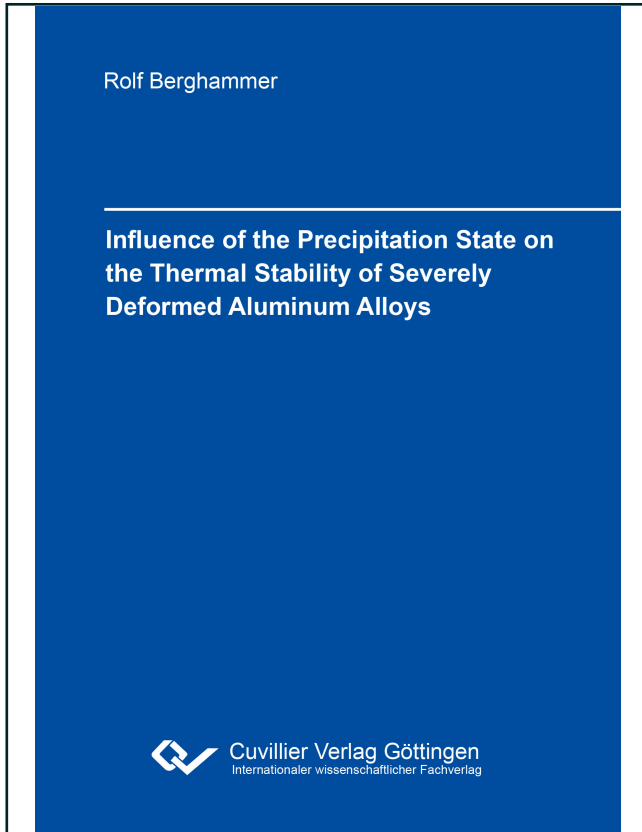Rolf Berghammer (Autor)
Influence of the Precipitation state on the Thermal Stability of Severely Deformed Aluminum Alloys

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1 Introduction

The history of ultra fine-grained materials (UFG) and severe plastic deformation (SPD) dates back to the 1950's [Bridgman 1952], but when Valiev reported outstanding mechanical properties as a result of the refined microstructure in the late 1980's [Valiev 1988], the interest in UFG grew enormously. A lot of improved properties, such as increased strength in combination with good ductility, improved fatigue resistance, fracture toughness and superplastic flow [Estrin 2002; Valiev 2000] were reported since then. UFG structures are defined as structures having grain sizes below 1 µm, being predominantly equiaxed with high angle grain boundaries (HAGB) being dominant [Valiev 2000].

UFG structures can be produced either by consolidation of powders or by severe plastic deformation (SPD) of bulk material. The so-called bottom-up approaches, where material is assembled from nanoparticles are inevitably connected with contamination and porosity [Gleiter 1989]. SPD processes deform materials without changing their overall geometry, thus they can be repeated for several times and thereby large strains can be accumulated in the material. During deformation dislocations are created, which will rearrange into a cell structure and with increasing absorption of dislocations, the misorientation of the boundaries increases and gradually high angle grain boundaries are formed [Hockauf 2010]. Grains will be refined down to a submicron level.

One of the most commonly used SPD processes is equal channel angular pressing (ECAP). At the Institute of Physical Metallurgy and Metal Physics at the RWTH Aachen University confined channel die pressing (CCDP) is conducted, which is a simple and fast method for producing small volumes of UFG structures.

Although a lot of publications deal with UFG materials and their improved mechanical properties [Estrin 2002; Valiev 2000] only rarely their thermal stability was examined [Etienne 2010; Adamczyk-Cieslak 2010; Hasegawa 1999]. But as the material properties are directly linked to the UFG structure, it is essential for technical application to preserve the structure through processing and application of the material even at elevated temperatures.

There are different opinions on the fundamental mechanisms of microstructure change during annealing of SPDed samples, it is still under discussion whether discontinuous recrystallization or discontinuous grain growth is responsible [Morris 2002; Vinogradov 2002; Poortmans 2004; Yu 2004].

One way to influence the thermal stability of the UFG structure is by different precipitation states. Precipitates are known to have a very complex influence on recrystallization, mostly depending on their size and their volume fraction, where small precipitates are capable to retard recrystallization and large precipitates can improve the formation of nuclei and thereby accelerate recrystallization. On the other hand, when alloying elements are in solid solution they can impede the movement of grain boundaries and suppress recrystallization [Gottstein 2007].

The scope of this work was to determine the influence of different precipitation states on the formation of microstructure during deformation and the thermal stability of this structure in a succeeding heat treatment.

Two different approaches were chosen to answer this question:

In the first approach an AlMnFe alloy was produced with two different precipitation states. Therefore one part of the samples was quenched from the melt to produce a supersaturated solid solution and the second part of the samples was thermo-mechanically treated to form precipitates. These samples were deformed by CCDP and ECAP. These methods were compared and as ECAP was more effective in grain refinement and resulted in more sample material ECAPed samples were chosen over CCDPed material.

For the second approach two different alloys were selected, one of them is a non-heat treatable alloy; the other one is heat treatable. When the second alloy is aged, the composition of the matrix is similar to the composition of the first alloy, but contains additionally precipitations. For this purpose the AlMg alloy AA5005 and the AlZnMg alloy AA7023 were chosen. Due to difficulties producing ECAP samples, for the last part of this work samples were heavily cold rolled.

The microstructure of the samples was characterized by optical and electron microscopy (EBSD and TEM), mechanical properties were determined by microhardness measurements and tensile tests, and in addition the texture was determined using an x-ray goniometer.

With a better understanding of the ongoing softening mechanisms a controlled combination of SPD and heat treatment gives the opportunity to improve the ductility and still preserve an increased strength. With optimized properties a broad field of possible applications for UFG materials opens up.

2 Theoretical Background

Metals are of high technical importance as they show a good formability and thereby can be deformed by various production processes, their high strength makes them interesting for numerous applications. While deformation leads to an increase of strength but a loss in ductility, softening mechanisms can increase ductility but on the same hand decrease strength. By a combination of deformation and annealing large changes in geometry can be realized, to control these properties a good understanding of the mechanisms is essential.

2.1 Fundamentals of Plastic Deformation
Mechanism

The formability of metals can be explained by the underlying mechanisms on a crystalline scale. Metals do have a highly ordered crystal structure and it can be shown by diffraction experiments that this structure is not altered during deformation (fig. II. 1). As the movement of complete crystal planes against each other would lead to higher – as actually measured – theoretical shear forces, it can be ruled out as a deformation mechanism. Rather than moving a big part of the crystal, a 1-dimensional defect will move through the lattice structure. The movement of this so-called dislocation will shear the lattice step by step. Dislocations are 1-dimensional defects in the crystal structure, which can be considered – in the easiest case – as an inserted half plane (fig. II. 2).
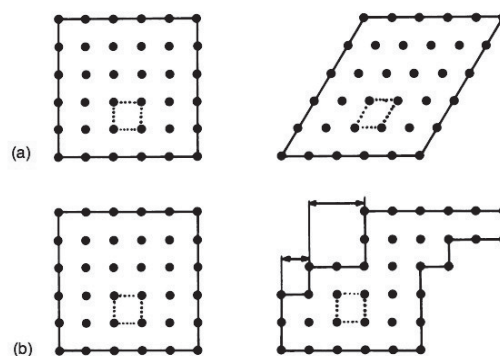

Figure II. 1: Scheme of possible deformation mechanisms on a crystalline scale [Gottstein 2007].

From experiments on single crystals glide lines can be seen which indicates glide concentrates on certain crystallographic planes.
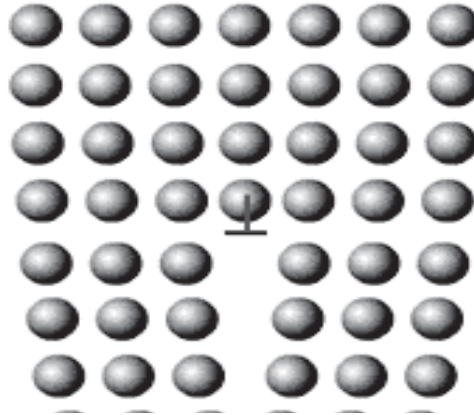

Figure II.2: Atomistic order of an edge dislocation [Alumatter 2013].

To move a dislocation through the crystal the Peierls-stress τ_P has to be exceeded.

$$\tau_P = \frac{2G}{1-\nu} \cdot \exp\left(-\frac{2p}{1-n}\frac{d}{b}\right) \qquad\qquad \text{equation II. 1}$$

This stress is proportional to the shear modulus G and exponentially proportional to the glide plane distance d divided by the Burger's vector b; ν is the Poisson ratio. It can be concluded from equation II. 1 that glide will occur on the planes with a high distance and in the closest packed direction. In the case of aluminum the {111}<110> slip systems will be activated, of which 12 exist.

The calculated shear stress under the assumption of dislocation movement is in the same order as the experimental value.

Stages of Deformation

The deformation changes as it progresses. Not only can elastic and plastic deformation be differentiated but also up to five different stages of plastic deformation can be distinguished [Sevillano 1993]. These stages can be visualized in a "Kocks-Mecking-Plot" (fig. II. 3), as they differ in their hardening coefficient.
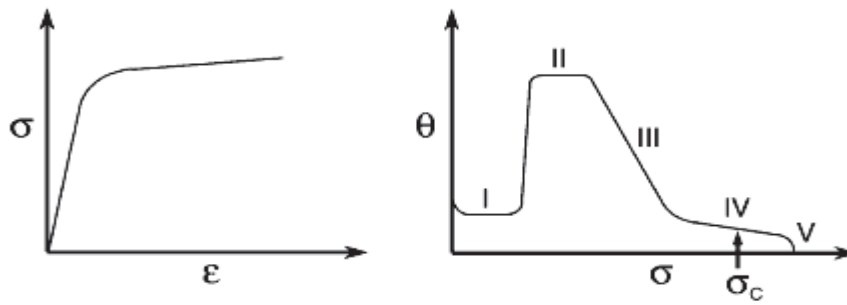
Figure II. 3: Kocks-Mecking-plot [Gottstein 2004].

Stage I is the stage of single slip and is also called easy-glide. It's characterized by a low hardening coefficient. As there is virtually no interaction of dislocations on the primary slip system with dislocations on different, secondary slip systems, dislocations can travel long distances without meeting obstacles. It should be mentioned that this stage only occurs in single crystals.

Stage II is initiated by the interaction of primary dislocations with dislocations on secondary slip systems. When meeting they form so-called Lomer-Cottrell-locks, which are immobile. Succeeding dislocations cannot bypass these obstacles; the internal stress increases. But to accommodate deformation the number of mobile dislocations has to stay constant, therefore new dislocations have to be created, this can happen through the activation of Frank-Read sources. The increasing number of dislocations accordingly leads to an in-creased flow stress (equ. II. 2).

$$\tau = \alpha\, G b \sqrt{\rho} \qquad\qquad\qquad\qquad \text{equation II. 2}$$

With α being a constant; G being the shear modulus; b the Burger's vector and ρ the dislo-cation density.

The hardening coefficient has considerably increased compared to stage I and is virtually independent of crystal orientation and even structure (equ. II. 3).

$$\Theta_{II} = \frac{d\sigma}{d\varepsilon_{II}} \approx \frac{G}{300} \qquad\qquad\qquad\qquad \text{equation II. 3}$$

In stage III the hardening rate decreases, which can be explained by dynamic recovery processes. By cross slip screw dislocations can change their glide plane and bypass obstacles. They are not restricted to a certain slip system, but the Schmid-factor is lower for the new slip system, thus a higher shear stress is necessary – but in stage III the external stresses are sufficient. Dislocations are now capable of traveling longer distances to annihilate each other [Gottstein 2007].

Stage IV shows a small and almost constant hardening rate. A virtually unchanged number of dislocations is leaving the cell interiors and is forced into a decreasing volume of cell walls. The cell interiors become almost dislocation free as compared to the cell walls [Estrin 1998].

The transition to stage V can be related to the generation and displacement of mobile sub-boundaries. As the cell walls get freed from debris and only consist of geometrical necessary dislocations they can be considered as low-angle grain boundaries (LAGB), which can move and thereby adjust to force imbalances [Gottstein 2004].

The Deformed State

When metals are deformed their microstructure will change. Not only the shape of the grains changes but also the total area of grain boundaries increases significantly. Dislocations created by deformation will be integrated in to the grain boundaries and increase their total area. Additionally these newly formed dislocations lead to the formation of an internal structure. Both the increase in grain boundary area and the accumulation of dislocations within the grains lead to a rise in stored energy, which later provides the driving force for recrystallization.

The shape change of grains is roughly in accordance with the overall change of the sample geometry; this leads to an increase of the surface of the grains where the dislocations, generated during deformation, are integrated. To be more specific the shape of the deformed grains relies also on the deformation mode; rolling leads to laths whereas compression results in disc shaped grains. Accordingly the increase of grain boundary area is not only dependent on the strain but also on the deformation mode. I.e. compression leads to a slightly higher grain boundary area than rolling [Humphreys 2004].

The dislocation density and thereby the stored energy is also influenced by the stacking fault energy (SFE). Low SFE promotes the dissociation of screw dislocations into partials. Partial dislocations have a smaller Burger's vector, which contributes by square to the energy and thereby the total energy can be reduced for the case that SFE is low enough that the partials can dissociate over a large distance. As the Burger's vector and the line element are not parallel anymore, the partial dislocations now are restricted to a defined glide system, so that cross slip is only possible when the partials reunite. This hinders dynamic recovery, which leads to an increase in dislocation density [Humphreys 2004; Gottstein 2007]. While copper has a low SFE and hardly recovers, aluminum has a high SFE and shows pronounced recovery.

Cell Formation

Dislocations rearrange into more stable configurations; the favorable distribution is a cell structure. Cell walls are formed by a three-dimensional network of complex dislocation tangles. With increasing strain cells decrease in size, usually down to a limit given by the shear stress. In aluminum the limiting cell size is in the range of 0.4 μm (see equation II. 4). With progressing deformation the cell walls sharpen and misorientation increases until the cell structure merges into a subgrain structure.

$$\frac{\sigma}{\mu} = k\,\frac{b}{d} \qquad\qquad\qquad\qquad \text{equation II. 4}$$

with d: subgrain size; b: Burgers vector; σ: stress; μ: shear modulus; $\frac{\sigma}{\mu}$: normalized shear stress; k: constant of about 10.

The deformation is not homogeneously distributed in the material; some grains of certain orientations undergo stronger deformation. Grains are subdivided and subsequently these subgrains rotate and change their orientation. By this way the deformation microstructure and texture are formed.

The cold worked microstructure is usually comprised of a high number of dislocations and a large fraction of low angle grain boundaries. In the case of strong plastic deformation a higher fraction of HAGB can be formed, and according to Humphreys the structure can be

stable against discontinuous growth (recrystallization), when a fraction of 0.6-0.7 of high angle grain boundaries is exceeded [Humphreys 2004].

Non-Cell-Forming Metals

As the formation of cells is based on the rearrangement of dislocations it can be referred to as a recovery process. But magnesium in solid solution (3-5%) can hinder dynamic recovery in aluminum by the pinning of dislocations and thereby cell structures cannot be formed. Most of the dislocations are arranged in diffuse patterns along {111} slip planes and microbands are formed.

Microstructure Development of Two-Phase Materials

Particles can have different influence on the microstructure development according to their size, appearance and properties. They can lead to an increased dislocation density (non deformable particles lead to the accommodation of geometrically necessary dislocations, but at large strains dynamic recovery usually limits the number of dislocations anyway, so that the effect of small non shearable particles is only noticeable at small strains). Around non deformable and coarse particles a zone of inhomogeneous deformation with high dislocation density, high misorientations and relatively small subgrains (<0.1 µm) is formed, this will trigger the so-called particle stimulated nucleation (PSN), which will be explained in chapter 2.2.

Severe Plastic Deformation

Going to very high deformations is one way to drastically refine the microstructure. By so-called severe plastic deformation ultra fine grained structures can be produced, which are reported to show improved properties, such as high strength in combination with good ductility, improved fatigue resistance, fracture toughness and superplastic flow [Estrin 2002; Valiev 2000].

The improved strength can – at least partially – be explained by the Hall-Petch equation, which correlates an increased strength with a decreasing grain size (d).

$$\sigma_y = \sigma_0 + k_y \sqrt{d} \qquad\qquad\qquad \text{equation II. 5}$$

k_y: Hall-Petch constant; d: grain size; σ_y: yield strength; σ_0: strength of single crystal

UFG refers to structures with grain sizes below 1 µm and a predominantly equiaxed grain structure. Valiev proposed that UFG materials must meet 3 requirements [Valiev 2000]:

1. HAGB should prevail in the UFG structures since only in this case a qualitative change in properties of materials occurs.

2. The uniform formation of nanostructures within the whole volume of a sample is necessary to provide stable properties of the processed materials.

3. Samples should not have any mechanical damages or cracks although they are exposed to large plastic deformations.

The fundamental ideas of SPD go back to the 1950s [Bridgeman 1952], but the process really gained momentum in the early 1980s, when Segal developed equal channel angular pressing (ECAP); and by the end of the 1980s, when the works of Valiev pointed out the potential to improve various properties by SPD [Langdon 2010]. It was shown by Langdon that SPD is by now one of the most worked on topics, according to the number of publications and citations, in materials science [Langdon 2010].

In contrast to conventional metal forming processes, where the strain is limited simply because of the reduction of the cross section of the work pieces, SPD processes can accumulate high strains without large changes in geometry, as the process can be repeated multiple times. A UFG structure is formed by the accumulation of dislocations and the formation of cells, subgrains and grains.

SPD techniques do have different strain paths, for example in high pressure torsion strain is monotonic, whereas in ECAP or confined channel die pressing (CCDP) the strain path is reversed [Horita 2000; Gholinia 2000].

During SPD coarse grains are fragmented and a large number of dislocations is arranged in a cell structure. Already after one pass an elongated, cellular structure can be observed, with its longitudinal direction aligned in the direction of the primary glide system [Iwahashi 1998]. At a certain point the ability of the cell boundaries to absorb more dislocations slows down and the dislocations form a structure within the cell, which will split the elongated cells into equiaxed cells [Helmig 2008]. With increasing number of passes the width of the cell walls decreases and the fraction of subgrain and grain boundaries increases [Dalla Torre 2004]. At first the disorientation between adjacent grains is only a few degrees, but with the integration of an increasing number of dislocations into the boundaries the disorientation increases, resulting in a growing fraction of HAGBs with the number of passes.

Cells gradually transform into subgrains, which are partially confined by clear and relaxed grain boundaries [Dalla Torre 2006]. The size of the grains is already determined in the first pass: it is the breadth of the elongated cells [Langdon 2007].

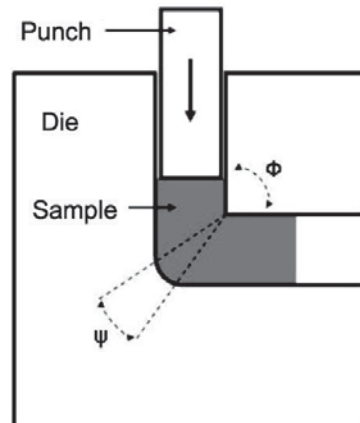### Equal Channel Angular Pressing

Figure II. 4: Schematic view of ECAP die [Berghammer 2011].

In ECAP a sample of square or circular cross section is pressed through a die consisting of two intersecting channels. The channels meet under an angle Φ, which usually lies between 90° to 120°. The transition of the channels often shows a curvature ψ, to facilitate the flow of the material. In the intersection of the channels the sample is deformed by shear [Suwas 2005]. As the sample geometry is kept constant, the billet can be reinserted and another ECAP-pass can be carried out. Between passes the sample can be rotated around its longitudinal direction. Depending on the nature of the rotation, different routes can be distinguished: route A – without rotation, B_A and B_C – rotation about 90° back and forth and in the same direction, respectively and C – rotation about 180°. As these routes activate different sets of shear planes, the selection of the route has a strong impact on the resulting microstructure and texture [Furukawa 1998]. Route B_C has the highest potential to generate an equiaxed structure [Molodova 2008; Valiev 2006b; Furukawa 1998; Furukawa 2002].

The imposed van Mieses stress can be calculated as