Microstructure and texture development in a polycrystal and different aluminium single crystals subjected to hydrostatic extrusion

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Abstract. A hydrostatic extrusion (HE) process was applied to commercial pure polycrystalline aluminium (99.9%) and two aluminium single crystals $\langle 111 \rangle$ and $\langle 110 \rangle$. On comparison, the results obtained from single crystals and polycrystalline aggregates are unique. Microstructure and crystallographic texture investigations were performed by transmission electron microscopy, electron backscatter diffraction and X-ray diffraction (XRD). Significant differences in grain refinement and texture formation were noticed depending on the starting orientation. The deformed single crystal with $\langle 110 \rangle$ starting orientation features an average grain size value of 150% higher than the second investigated single crystal (0.5 μm for the $\langle 111 \rangle$ single crystal and 1.3 μm for the second crystal). In turn, the average grain size obtained for polycrystalline aluminium is 0.9 μm . The deformation process causes a difference in the grain sizes, while a fraction of the high angle grain boundaries have a comparable volume percentage in all the deformed microstructures—reached about 35%. The qualitative and quantitative XRD texture results proved that the HE process leads to the formation of a characteristic fibrous texture.

Keywords. Deformation texture; crystallographic orientation; single crystal; electron backscattering diffraction; aluminium.

1. Introduction

Various techniques based on severe plastic deformation (SPD) have been used to achieve grain refinement in metals and their alloys [1–3]. Most of the scientific research studies are concerned with the deformation of polycrystalline materials using the processes of equal-channel angular pressing (ECAP) [3–6], high pressure torsion [7] and accumulative roll bonding [8]. SPD methods have also been used to examine the texture and microstructure evolution of single crystals after deformation [9–12]. Their application in these studies may be easily explained. Firstly, the influence of the initial grain structure on the deformation results is excluded. Secondly, it is easy to determine the initial texture/orientation of the material. Experiments on single crystals processed by SPD have been reported latterly. Most often the single crystals of Al [10,13–15], Ni [16,17] and Cu [9] are deformed by ECAP or plain strain compression in channel die [12,18,19]. The hydrostatic extrusion (HE) process has only been reported on single crystals of nickel [20,21] and Cu-8.5 at% Al alloys [22]. The authors of the present paper have not found any analyses of single and polycrystals deformed under the same conditions in any publications.

The main aim of the present paper is to investigate the influence of initial orientation on the microstructure and the

texture formation of aluminium single crystals with different initial crystallographic orientations. Furthermore, the authors want to compare the differences in the texture and microstructure formation in hydrostatically extruded aluminium single crystals and polycrystalline aggregates.

2. Materials and methods

The experiments were conducted using single crystals with two different orientations $\langle 111 \rangle$ and $\langle 110 \rangle$ of high-purity (99.99%) aluminium single crystals grown by the modified Bridgman method. The single crystal with $\langle 110 \rangle$ orientation in the as-received state is shown in figure 1a. The polycrystalline material used in the study had the same purity and the average grain size was about 50 μm (figure 1b), measured on a cross-section perpendicular to the extrusion direction (ED).

The HE experiment was conducted at room temperature in two consecutive passes from the initial diameter of 9 to 7 mm and 5 mm, respectively. In this deformation process the billet is forced through a die by means of the pressure of a liquid. As it is possible to attain a large strain, this method is suitable for the refinement of a material's microstructure, and the HE scheme is shown in figure 2. The main advantages of

Figure 1. Samples before the deformation (a) single crystal and (b) microstructure of polycrystalline aluminium (observed in a light microscope using a polarized light beam).

Figure 2. The scheme of HE facility.

this process are the prevention of cracking and the possibility of producing relatively large products compared to the other methods of SPD [23].

The total true strain [21] after the last step of deformation was 1.2. The $\langle 111 \rangle$ and $\langle 110 \rangle$ orientations of the single crystals were perpendicular to the ED.

For detailed microstructure investigations, an focused ion beam and a transmission electron microscope (TEM) were used. The observations of the HE-treated microstructures were analysed on a JEOL 1200 TEM with an accelerating voltage of 120 kV. The TEM samples were mechanically ground to a thickness of $\sim 20 \mu\text{m}$ followed by twin-jet electropolishing at 20 V DC in a Struers A2 electrolyte [24]. The TEM micrographs were used to calculate the grain size for all HE-treated samples. It was determined by calculating the average equivalent diameter (d_2), which indicates the diameter of a circle with an area equal to the investigated grain size [25]. Two hundred randomly selected grains were used for grain parameter calculation.

EBSD analyses were carried out on a Hitachi SU70 scanning electron microscope (SEM), and backscattered electrons at 20 kV were used to reveal the crystallographic contrast. The SEM was equipped with facilities for EBSD. The pattern

acquisition and solution were achieved with HKL Channel 5 software. In all the cases examined, a step size of $0.1 \mu\text{m}$ was applied. While processing the EBSD data the orientation distribution maps, inverse pole figures (IPFs) and grain boundary (GB) misorientation distributions were performed. To eliminate spurious boundaries caused by orientation noise, a lower-limit boundary misorientation cut-off of 2° was applied. High angle grain boundaries (HAGBs) are boundaries possessing large crystal misorientation ($\geq 15^\circ$), the reverse is true in the case of low angle grain boundaries (LAGBs) ($2\text{--}15^\circ$) [26].

The global texture measurements after deformation were performed using a Bruker D8 X-ray diffractometer set-up with a $K\alpha$ radiation filter for the Co anode. The measurements were recorded within a $5^\circ \times 5^\circ$ mesh and a counting time 5 s per step. It should be pointed out that a tolerance angle of 20° was used when the volume fractions were calculated. All these experiments were conducted on a plane perpendicular to the ED.

3. Results and discussion

3.1 Microstructure

TEM observations of the HE-treated materials revealed that in all samples an ultrafine-grained structure was achieved. Figure 3 depicts the channelling contrast by using electrons generated by ions with the corresponding TEM images of the samples after deformation. It can be easily seen that the same deformation caused differences in the microstructure of all the specimens examined. In the HE-treated samples, the grains were regular in shape with a low tendency to elongation. The single crystal with $\langle 110 \rangle$ starting orientation features an average grain size value of 150% higher than the second single crystal investigated. Relatively large grains (about $1 \mu\text{m}$ in equivalent diameter) with dislocation cells were observed only in the deformed $\langle 110 \rangle$ crystal. Inside the grains, in all the HE-treated samples, some dislocations were noted. Moreover, it consisted of dense dislocation tangles showing a strong disorder and a large number of defects in the microstructure. Moreover, a ribbon-like structure was only found in this

Figure 3. Representative microstructures together with histograms of grain sizes (d_2) of HE-treated aluminium (a, b) <111>, (c, d) <110> single crystals and (e, f) polycrystal. Arrows indicate the ribbon-like structure.

HE-treated crystal (indicated by arrows). Elongated, ribbon-like dislocation cells or subgrain structures form at smaller equivalent strains during SPD at low homologous temperatures leading to highly refined, equiaxed structures at larger strains. This kind of structure is apparently developed by recovery-dominated processes in the absence of high-angle boundary migration, and their evolution is also strain-path dependent. A ribbon-like microstructure was also found in Al single crystals as a result of another deformation

mechanism—shear—implemented by the activation of primary slip planes during ECAP [15,27].

The generation and migration of dislocation loops, and the subsequent interaction, storage and recovery of the dislocations are the fundamentals of the evolution of the deformation structure. Aluminium is a face-centred cubic (Al) material with high stacking-fault energy, which makes the dissociation into partial dislocations difficult. Therefore, cross-slip and consequently also dynamic recovery, are promoted.

In cold forming of aluminium alloys it is a common assumption that all slips occur on the $\{111\}\langle 110 \rangle$ slip systems and that dislocation climb is limited.

The formation of the dislocation structure in fcc single crystals during plastic deformation demonstrates a strong dependence on the crystal orientation. Deformation along $[111]$ activates six slip systems, which are primary, coplanar, codirectional (oriented for difficult cross-slip) and conjugate to another active system. Simultaneously, deformation along $[110]$ causes the activation of only four slip systems—primary and critical [28]. The plastic deformation starts in the slip system, in which the Schmid factor has the highest value. In the case of the $\langle 111 \rangle$ single crystal, the determined Schmid factor is 0.27, while for the $\langle 110 \rangle$ crystal it is higher and amounts to 0.41. This indicates that slip is easier, at the same level of stress, for a single crystal with starting orientation $\langle 110 \rangle$. This orientation corresponds to ‘soft’ orientations, while the $\langle 111 \rangle$ is ‘hard’ orientation [29]. When the dislocations are blocked, further deformation is made possible by an increase in the force which causes exceedance of the critical stress for slip or other systems by rotation of the crystal lattice. This phenomenon aids the formation of grain boundaries and their greater fragmentation. In the $\langle 110 \rangle$ single crystal the longer range of easy slip will be activated, and simultaneously the hardening coefficient has a lower value. This is related to easier dislocation movements and the creation of the cell structure [28]. The results of the microstructure presented in this work indicate that crystallographic relationships are not the deciding factor of the structure evolution, because of the activation of heating processes during deformation, having an impact on the microstructure evolution. The influence of the heating processes on the examined materials was confirmed by the obtained texture results of the HE-treated materials through this study. The qualitative and quantitative texture results revealed the presence of the local maxima on $\langle 100 \rangle$ fibre which may indicate the initial stages of the recrystallization process [30].

One of the characteristic features of the as-deformed microstructure is the presence of small equiaxial grains and high density of dislocations. The grain size distributions in the HE-treated aluminium $\langle 111 \rangle$ and $\langle 110 \rangle$ single crystals and the polycrystal are shown in figure 3b, d and f. HE in the $\langle 111 \rangle$ single crystal results in a significant increase of the formation of small grains (80% of d_2 below $0.8 \mu\text{m}$) in comparison with the $\langle 110 \rangle$ crystal. These differences in the distribution of the grain size suggest that the evolution of grains during the processing by HE involves the conversion of large grains into smaller ones depending on the initial orientation of the single crystal.

In order to describe the changes in the microstructure of the investigated single crystals after deformation, EBSD studies were carried out. Figure 4 depicts the EBSD maps with the corresponding IPFs and grain (subgrain) boundary (GB) misorientation distributions, where HAGBs and LAGBs are depicted as yellow and black lines, respectively. The GB misorientation angles above 15° reach more than 30%

in deformed poly- and single crystals. This shows that the deformation process causes differences in the grain (subgrain) refinement while GB misorientation angles have a comparable volume percentage in the microstructure. The presented orientation data were obtained from the EBSD measurements on the plane perpendicular to the ED of the deformed single crystals. The maps were generated by plotting the crystal orientation onto a colour scale and shading each point on the grid according to some aspects of the crystal orientation. The colours presented in the IPFs directly correspond to the grain orientations on the map. The small green points represent the unidentified orientations.

The microstructure of the single crystals after HE consists of relatively large regions with the same orientation divided into smaller parts by LAGBs or less frequently by HAGBs. In the crystal with the greatest grain fragmentation (starting orientation $\langle 111 \rangle$), the subgrains arranged themselves in such a way that the direction $\langle 111 \rangle$ is perpendicular to the ED (according to the IPFs from figure 4a). What is more, deviation from the $\langle 111 \rangle$ direction of the components is large and reaches over 20° . In the second single crystal, the occurrence of the $\langle 001 \rangle$ texture component can be seen. The material after deformation features two dominant orientations: $\langle 001 \rangle$ and $\langle 111 \rangle$ parallel to the ED (figure 4c). These are the main components of the texture formed in the extrusion processes. Additionally, the fraction of the boundaries between the $\langle 001 \rangle$ and $\langle 111 \rangle$ oriented grains also increases. This means that dynamic recrystallization also occur during deformation. In the case of the polycrystal after deformation, the same two dominant orientations are present as in the single crystal $\langle 110 \rangle$, namely $\langle 001 \rangle$ and $\langle 111 \rangle$ (figure 4e). IPFs indicate that additional orientations extending around these orientations are also present. However, their content is significantly smaller than the two main components. The appearance of additional orientations is related to the processes of recovery and local recrystallization, activated during deformation.

By summarizing our own research studies and literature reviews, it can be stated that the grain orientation has a major impact on the microstructural evolution since it determines the activation of various slip systems. In order to activate a slip system, the value of the shearing component needs to exceed the critical resolved shear stress for the considered system. Winther and Huang [28] described the influence of crystallographic grain orientation on the formation of different dislocation structures. The obtained microstructures were connected with their primary orientation against the material flow and active slip systems. The authors presented different types of microstructures in relation to the grain orientation by using the standard stereographic triangle. Three general types of dislocation structures can be distinguished. The first type is a cell-block structure with straight and parallel sets of cell-block boundaries aligned with $\{111\}$ planes and with deviation from the slip plane up to 10° . The grains demonstrating this type of structure are located in the middle part of the triangle. The second type of microstructure is an equiaxed dislocation cell and the grains forming this type of structure

Figure 4. The EBSD maps and the corresponding IPFs and GB misorientation distributions of the HE-treated aluminium (a, b) $\langle 111 \rangle$, (c, d) $\langle 110 \rangle$ single crystals and (e, f) polycrystals.

are near the $\langle 100 \rangle$ pole. The last, the third type of structure, is a cell-block structure deviating from $\{111\}$ by more than 10° . Boundaries contain short and long segments but the majority of them are aligned with the deformation direction, they are close to the $\langle 111 \rangle$ pole. The three types of dislocation structures can be related to slip classes which are sets of mutually active slip systems. In the $\langle 111 \rangle$ single crystal, the deformation microstructure was characterized by high dislocation density, cell walls with a complicated morphology and very fine grains (subgrains) whose average size does not exceed $0.5 \mu\text{m}$. In the case of the $\langle 110 \rangle$ aluminium single crystal, the deformation microstructure was composed of cells with similar orientations but different sizes. The average grain (subgrain) size is almost two times higher than that of the second investigated single crystal.

3.2 Texture

Three pole figures, namely $\langle 111 \rangle$, $\langle 200 \rangle$ and $\langle 220 \rangle$, were used to calculate the orientation distribution functions, which fully describe the texture of individual samples. In order to determine the influence of initial texture on the formation of the deformation texture, it was necessary to analyse the texture of the polycrystalline aluminium before the HE process. Figure 5 depicts the recalculated pole figures (RPFs) $\langle 111 \rangle$ and $\langle 200 \rangle$ for all examined samples.

Other studies on polycrystalline alloys after HE revealed the formation of double fibre texture [23,31,32] with different volume fractions of the $\langle 111 \rangle$ and $\langle 001 \rangle$ components. In order to perform a more detailed analysis, quantitative calculations of texture components were conducted (figure 6).

Figure 5. The RPF (111) and (200) of the polycrystalline aluminium (a) before deformation and HE-treated materials: (b) polycrystal, (c) $\langle 111 \rangle$ single crystal and (d) $\langle 110 \rangle$ single crystal.

In polycrystalline aluminium before the HE the main texture component is $\langle 001 \rangle$ fibre. As a result of the deformation, weakening of the volume percentage of this fibre occurs, and additionally a $\langle 111 \rangle$ component is formed. The $\langle 111 \rangle$ and $\langle 100 \rangle$ double fibre texture is characteristic for drawing, swaging or extrusion of fcc materials [33]. Suwas and Ray [33] reported that the volume fraction of $\langle 111 \rangle$ and $\langle 100 \rangle$ components is interrelated to the stacking fault energy (SFE). Metals with high SFE, like aluminium, exhibit principal $\langle 111 \rangle$ fibre. The fraction of $\langle 100 \rangle$ fibre is higher than that of $\langle 111 \rangle$

fibre for low SFE materials. Thus, they concluded that in extrusion texture distribution, $\langle 111 \rangle$ and $\langle 100 \rangle$ components vary with the extrusion speed and temperature, and the amount of $\langle 001 \rangle$ oriented grains increases linearly with the temperature at a given speed. The results demonstrated in this study revealed the major influence of the initial texture on its formation after deformation and, what seems to be the most important, undermine the theory of double fibre formation in the texture after extrusion. The fibre components are present in the texture of the HE-treated materials.

Figure 6. Volume fraction of the principal texture components in the investigated materials.

In the $\langle 110 \rangle$ single crystal, the HE process caused perpetuation of the initial orientation in 20% volume fraction and small fraction of the $\langle 111 \rangle$ component. In the second single crystal, there was a total loss of the original texture after HE and low level texture was formed (background 78 vol.%). The $\langle 001 \rangle$ fibre, which was predominant in the $\langle 110 \rangle$ single crystal (figure 6) has undergone a significant broadening and weakening in the second investigated single crystal. The high level of the background (in both examined single crystals) denotes the formation of randomly oriented components in significant volume fraction. The texture results of the HE-treated single crystals show the presence of local maxima on $\langle 001 \rangle$ fibre which can be typical for the recrystallization process in fcc materials [34,35] and indicates that during deformation, the thermal effects have taken place.

The lowest level of texture was recorded in single crystals after deformation irrespective of their initial orientation (background above 75% vol). The HE process also caused a weakening of the texture in the polycrystalline materials and the volume fraction of the random orientation increased to 52%. It should be emphasized that the presented results do not reflect the characteristic orientations obtained by the extrusion process.

The texture results are in good coincidence with the microstructure analysis. In consequence of the HE process of the $\langle 110 \rangle$ crystal, there was a significant increase in the component of $\langle 100 \rangle$ in the volume fraction, the presence of which is assigned to overlap the processes of recovery and recrystallization. This result is confirmed by the grain size, wherein the deformed single crystal of $\langle 110 \rangle$ orientation grains is more than twice the grain size in the $\langle 111 \rangle$ single crystal.

4. Conclusions

This study was devoted to the investigation of a microstructure and texture evolution in commercial pure aluminium single crystals and polycrystals after the HE process. From the results obtained, the following major conclusions can be drawn:

1. The initial orientation of the investigated materials has a major influence on the deformation texture formation.
2. The significant effect of texture/orientation before deformation on the microstructure was proved.
3. As a result of deformation of single crystals, randomly oriented grains form above 75% of the volume fraction and the microstructure becomes more inhomogeneous.
4. The heating processes had an influence on the microstructure and texture formation.
5. The crystallography relationships of slip systems cannot be the only factor indicating the microstructural evolution in fcc materials after uniaxial deformation. The influence of heating processes is very important on the material properties.

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References

- [1] Higuera-Cobos O F, Berríos-Ortiz J A and Cabrera J M 2014 *Mater. Sci. Eng. A* **609** 273
- [2] Zhilyaev A P, Kim B-K, Szpunar J A, Baró M D and Langdon T G 2005 *Mater. Sci. Eng. A* **391** 377
- [3] Bazarnik P, Romelczyk B, Kulczyk M and Lewandowska M 2013 *Mater. Sci. Forum* **765** 423
- [4] Beyerlein I J and Tóth L S 2009 *Prog. Mater. Sci.* **54** 427
- [5] Lipińska M, Olejnik L, Pietras A, Rosochowski A, Bazarnik P, Goliński J *et al* 2015 *Mater. Design* **88** 22
- [6] Ciemiorek M, Chromiński W, Olejnik L and Lewandowska M 2017 *Mater. Design* **130** 392
- [7] Vorhauer A and Pippin R 2004 *Scr. Mater.* **51** 921
- [8] Jamaati R and Toroghinejad M R 2014 *Mater. Sci. Eng. A* **598** 263
- [9] Miyamoto H, Fushimi J, Mimaki T, Vinogradov A and Hashimoto S 2005 *Mater. Sci. Eng. A* **405** 221
- [10] Furukawa M, Kawasaki Y, Miyahara Y, Horita Z and Langdon T G 2005 *Mater. Sci. Eng. A* **410–411** 194
- [11] Pardis N, Chen C, Ebrahimi R, Tóth L S, Gu C F, Beausir B *et al* 2015 *Mater. Sci. Eng. A* **628** 423
- [12] Miszczyk M, Paul H, Driver J H and Maurice C 2015 *Acta Mater.* **83** 120
- [13] Fukuda Y, Oh-Ishi K, Furukawa M, Horita Z and Langdon T G 2006 *Mater. Sci. Eng. A* **420** 79
- [14] Fukuda Y, Oh-Ishi K, Furukawa M, Horita Z and Langdon T G 2004 *Acta Mater.* **52** 1387
- [15] Han W Z, Zhang Z F, Wu S D and Li S X 2007 *Acta Mater.* **55** 5889
- [16] Grosdidier T, Fundenberger J-J, Goran D, Bouzy E, Suwas S, Skrotzki W *et al* 2008 *Scripta Mater.* **59** 1087
- [17] Skrotzki W, Tóth L S, Klöden B, Brokmeier H-G and Arruffat-Massion R 2008 *Acta Mater.* **56** 3439
- [18] Stanford N, Dunne D and Ferry M 2003 *Mater. Sci. Eng. A* **348** 154
- [19] Dymek S and Blicharski M 1996 *Acta Metall.* **35** 417
- [20] Zdunek J, Mizera J, Wąsik K and Kurzydłowski K J 2013 *Mater. Sci. Eng. A* **569** 92
- [21] Jakubowska D, Zdunek J, Kulczyk M, Mizera J and Kurzydłowski K J 2015 *Adv. Mater. Sci. Eng.* **2015** ID 613839
- [22] Jakubowska D, Zdunek J, Kulczyk M, Mizera J and Kurzydłowski K J 2016 *Arch. Metall. Mater.* **61** 933
- [23] Chrominski W and Lewandowska M 2016 *Acta Mater.* **103** 547
- [24] <http://struers.com/>
- [25] Wejrzanowski T, Pielaszek R, Opalińska A, Matysiak H, Łojkowski W and Kurzydłowski K J 2006 *Appl. Surf. Sci.* **253** 204
- [26] Suwas S and Gurao N P 2008 *J. Indian Inst. Sci.* **88** 151
- [27] Swaminathan S, Ravi Shankar M, Lee S, Hwang J, King A H, Kezar R F *et al* 2005 *Mater. Sci. Eng. A* **410–411** 358
- [28] Huang X and Winther G 2007 *Phil. Mag.* **87** 5189
- [29] Winther G and Huang X 2007 *Phil. Mag.* **87** 5215
- [30] Samajdar I and Doherty R D 1998 *Acta Mater.* **46** 3145
- [31] Bohlen J, Yi S B, Swiostek J, Letzig D, Brokmeier H G and Kainer K U 2005 *Scripta Mater.* **53** 259
- [32] Majda T, Moszczyńska D, Zdunek J, Kulczyk M, Mizera J and Kurzydłowski K J 2017 *Mater. Sci. Tech.* **836** 1
- [33] Suwas S and Ray R K 2014 *J. Indian Inst. Sci.* **88** 151
- [34] Doherty R D, Hughes D, Humphreys F J, Jonas J J, Juul Jensen D, Kassner M E *et al* 1998 *Mater. Sci. Eng. A* **1** 14
- [35] Kashyap K T and George R 2006 *Bull. Mater. Sci.* **29** 197