ARCHIVES OF METALLURGY AND MATERIALS
Volume 51 2006 Issue 4

M. LEWANDOWSKA*

STABILITY OF SECOND PHASE PARTICLES DURING PROCESSING BY HYDROSTATIC EXTRUSION

STABILNOŚĆ CZĄSTEK DRUGIEJ FAZY PODCZAS PROCESÓW WYCISKANIA HYDROSTATYCZNEGO

In the present work, the stability of second phase particles in an aluminium alloy during the processing by hydrostatic extrusion and their effect on the process of grain refinement have been investigated. Two types of intermetallics: primary particles and secondary precipitates have been taken into account. The results have shown that the processing by hydrostatic extrusion affects both types of them. The secondary plate-like precipitates transform into spherical particles. Primary particles decrease slightly their size, change their shape into more regular and become more uniformly distributed in the matrix. Intermetallics of both types affect also the process of grain refinement which is favoured by the presence of large primary intermetallics whereas delayed by small precipitates.

Keywords: aluminium alloys, hydrostatic extrusion, grain refinement, precipitates, intermetallic particles

W niniejszej pracy analizowano stabilność cząstek drugiej fazy w stopach aluminium podczas kształtowania metodą wyciskania hydrostatycznego. Analizę przeprowadzono dla dwóch rodzajów cząstek: (1) pierwotnych cząstek faz międzymetalicznych oraz (2) wtórnych wydzieleń faz międzymetalicznych. Uzyskane wyniki pozwoliły ustalić, że proces wyciskania hydrostatycznego powoduje rozdrobnienie i zmianę kształtu wydzieleń z płytkowego na sferoidalny. Wykazano także, że pierwotne cząstki faz międzymetalicznych podczas procesów wyciskania hydrostatycznego nieznacznie zmniejszają swoje rozmiary, ich kształt staje się bardziej regularny, a rozmieszczenie bardziej jednorodne. Określono także wpływ tych cząstek na proces rozdrobnienia ziarna. Stwierdzono, że duże cząstki pierwotnych faz międzymetalicznych sprzyjają procesowi rozdrobnienia ziarna, natomiast wtórne wydzielenia opóźniają ten proces.

1. Introduction

Stability and transformations of phases are well recognized for conventional materials of the grain in the micro-meters range. Nowadays, these phenomena attract new attention in the context of nanocrystalline materials in which the large fraction of atoms are located in interfaces area and as a result phase stability might be profoundly different [1].

Nanomaterials, in particular nanometals, are often produced by the severe plastic deformation (SPD). One of the methods which allow to obtain sufficiently large plastic strains is hydrostatic extrusion. In this method, the billet is located in the container and surrounded with a pressure transmitting medium. The piston compresses the medium until the billet starts to extrude through the die. The negligibly small friction between the billet and the die allows the use of small die angles that ensure high deformation homogeneity. Previous studies have shown that hydrostatic extrusion is an efficient way to produce nanocrystalline materials, e.g. aluminium alloys or titanium [2-4]. The unique features of HE are three-axial compressive stresses within the billet and high strain rates. The high strain rates imply the adiabatic heating which may influence both microstructure evolution and phase stability. On the other hand, high strain rate may provide a new opportunity for suppressing recovery and possible grain growth in heavily deformed metals, in particular when the product will be cooled at the die exit. SPD processing results also in a large amount of defects, primarily dislocations, being accumulated in the material processed.

It has been already shown that SPD strongly affects the stability of phases, for example carbides in Fe alloys [5]. One should expect that it also affects the stability of particles in other multiphase materials, e.g. age-hardenable aluminium alloys in which second phase

---

* FACULTY OF MATERIALS SCIENCE AND ENGINEERING, WARSAW UNIVERSITY OF TECHNOLOGY, 02-507 WARSAW, 141 WOŁOSKA STR., POLAND

particles are crucial to the mechanical strength. The aim of the present work is to study the stability of second phase particles in an aluminium alloy during the processing by hydrostatic extrusion in the context of their effect on the process of grain refinement. The second phase particles in this case are: primary (particles) and secondary (precipitates) intermetallics.

2. Materials and experimental procedure

The material used in the present study was an age-hardenable 2017 aluminium alloy of chemical composition presented in Table 1. The material was solution heat treated, water quenched and aged at 200°C for 3 hours. Next, it was hydrostatically extruded in three consecutive passes. The initial diameter of samples was 20 mm, whereas the final 3 mm which corresponds to a true strain of 3,8. The process of hydrostatic extrusion was conducted at the Institute of High Pressure Physics in Warsaw. The details of the deformation procedure were described elsewhere [2].

TABLE 1

Chemical composition of 2017 aluminium alloy

alloy	Chemical composition in weight %								
	Cu	Mg	Mn	Si	Fe	Zn	Ti	Pb	Al
2017	4.6	0.53	1.00	0.31	0.30	0.05	0.01	–	balance

In order to describe the evolution of microstructure electron microscopy has been used in TEM (precipitates) and SEM (particles) modes. The sample for TEM observations were cut out perpendicularly or parallel to the extrusion direction. The foils were examined in a Jeol-1200 electron microscope operated at 120 kV. The process of grain refinement was characterized in terms of the grain size and grain boundary misorientations. The latter measurements were performed using an automatic system of Kikuchi line analysis developed at the Institute of Metallurgy and Materials Science PAS in Cracow [6].

3. Stability of precipitates during processing

Precipitates are one of the most important elements of aluminium alloys microstructure. They form during aging of solution heat treated and water quenched alloy. Age hardened aluminium alloys exhibit the highest mechanical strength among all aluminium alloys. The role of precipitates during conventional plastic deformation is quite well known. Small and coherent precipitates are sheared by moving dislocations. This may lead to the localization of plastic deformation in the form of shear bands [7] and to changes in their shape and eventually dissolution [8]. Thus, one can expect that similar processes will take place during SPD processing.

The ageing process in the alloy studied here brings about the formation of plate-like Θ′ precipitates (Fig. 1a) of approximately 200 nm in length and 20 nm in width, lying on the {100} planes. Hydrostatic extrusion to a true strain of 1,4 results in the fragmentation of precipitates into smaller parts as a result of shearing by moving dislocations. Subsequent deformation (up to a true strain of 3,8) causes that the precipitates changed their shape into small spherical particles which diameter can be estimated at 10 nm (Fig. 1b). This is an indication of precipitates being strongly refined during the process of plastic deformation. It should be noted that this refinement takes place uniformly in the whole volume of a material, with no shear banding.

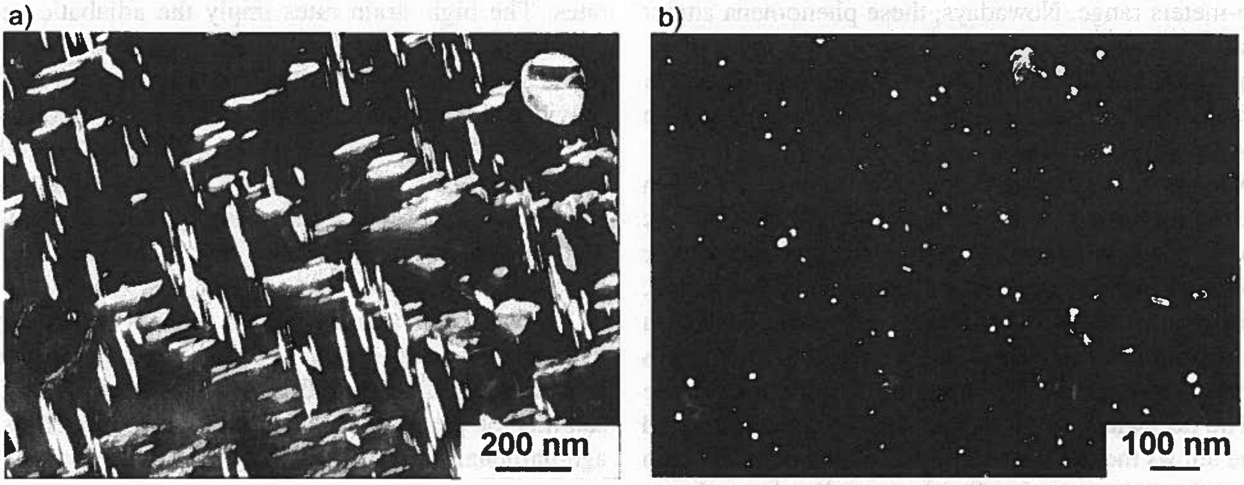

Fig. 1. TEM micrographs of precipitates in 2017 aluminium alloy aged at 200°C for 3 hours: (a) before and (b) after hydrostatic extrusion

The mechanism of the above described transformation in size and shape of precipitates is not still known. It seems that the first stage of transformation (fragmentation) is purely a result of interactions between precipitates and dislocation whereas the second (shape change) requires diffusion and thus needs thermal activation. When the precipitates become significantly fragmented, there is a strong need to decrease the surface area of precipitate-matrix interfaces. This is achieved by the change in precipitate shape. The necessary thermal activation of this process is provided by adiabatic heating which occurs during plastic deformation with very high strain rates (e.g. hydrostatic extrusion). In the case of aluminium alloys, the temperature rise can be estimated to some 200°C which is sufficient to initiate diffusional transformation.

4. Stability of intermetallic particles during processing

In commercial aluminium alloys, there are always some content of impurities such as Fe and Si which form primary intermetallic particles during solidification. These particles are relatively large (several microns in diameter) and incoherent with the matrix. They are thermodynamically stable in the wide range of temperatures and thus considered stable or changing negligibly during the SPD processing. However, the observations of microstructure evolution occurring in cast alloys (which contain a large amount of intermetallic particles) indicate that during SPD processing, such particles undergo significant transformation in terms of their size and distribution [9-11]. This suggests that similar changes can take place in alloys containing lower amount of intermetallics, but to date this was not examined in detail.

In the as-received state, the 2017 aluminium alloy contains relatively large primary intermetallic particles (Fig. 2a) with chemical composition corresponding to $(FeCuMn)_3Si_2Al_{15}$ phase. The average equivalent diameter of these particles is 1,4 μm and their spatial distribution is inhomogeneous with small ones forming clusters. These clusters correspond to the eutectic structure as the intermetallics crystallize usually as a part of eutectic. On the longitudinal sections, the characteristic arrangement of particles induced by the extrusion is observed.

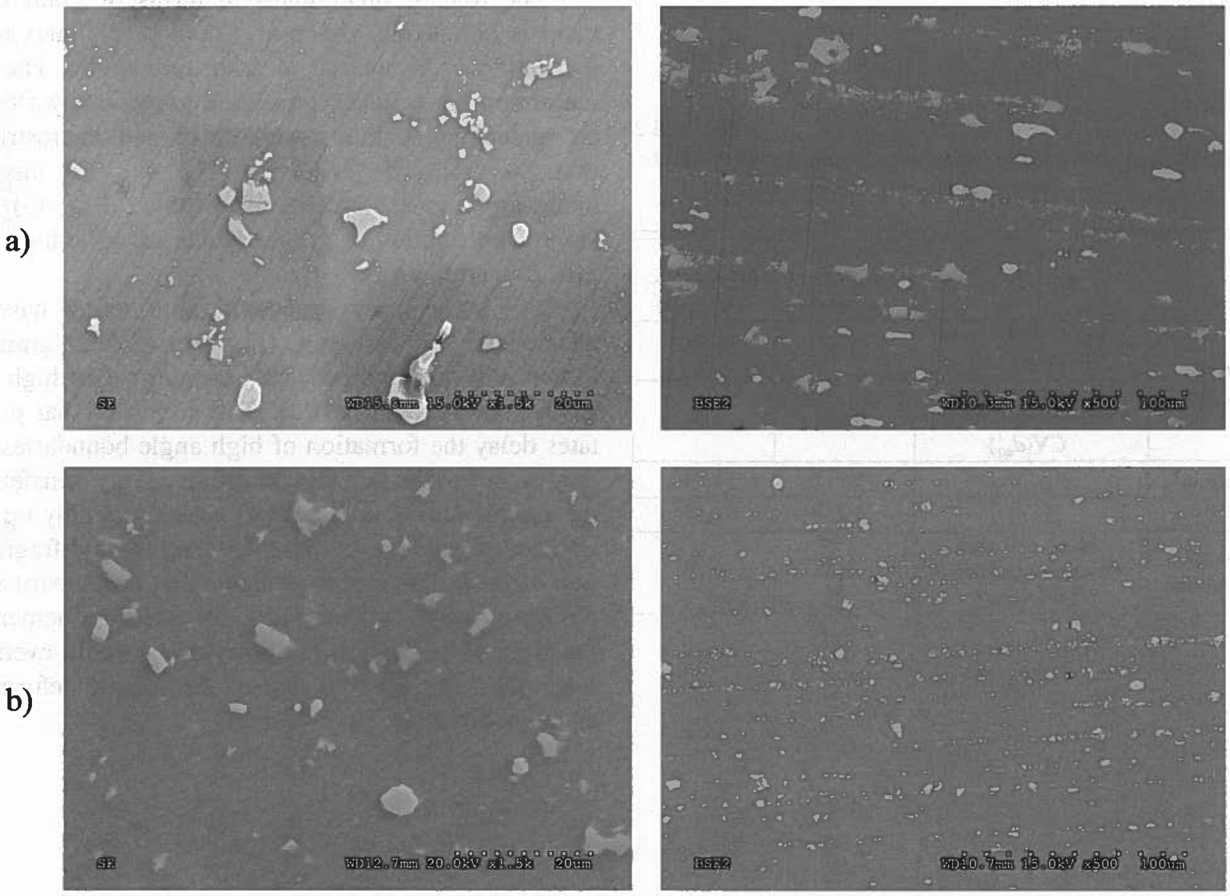

Fig. 2. SEM images of primary intermetallic particles: (a) before and (b) after hydrostatic extrusion

Hydrostatic extrusion results in significant changes in both geometrical parameters of intermetallic particles and their spatial distribution (Fig. 2b). These changes have been quantified by measuring their size (equivalent diameter, d_{eq}) and shape (shape parameters: d_{max}/d_{eq} and $p/\pi d_{eq}$ describing particle elongation and interface boundary development, respectively). The spatial distribution was determined using the method of SKIZ tessellation in which the value of variation coefficient of the area of cells surrounding each particle describes the homogeneity of spatial distribution [12]. The values of all these parameters, before and after processing by hydrostatic extrusion, are summarized in Table 1. These data clearly show that hydrostatic extrusion slightly reduces the size of primary intermetallic particles but it remains at the level of micrometers. More significant changes occur in the particle shape, which becomes more regular (the value of both shape parameters is substantially lower after hydrostatic extrusion), and spatial distribution, which becomes more homogenous. It should be noted that such changes in particle geometrical parameters will influence the mechanical properties of a material. In particular, one can expect the improvement of fracture toughness of hydrostatically extruded aluminium alloys.

TABLE 2

Stereological parameters of primary intermetallic particles before and after hydrostatic extrusion

	parameter	before hydrostatic extrusion	after hydrostatic extrusion
particle size	mean equivalent diameter, d_{eq}	1.4	1.24
	equivalent diameter variation coefficient, CV(d_{eq})	0.82	0.64
particle shape	d_{max}/d_{eq}	1.41	1.34
	$p/\pi d_{eq}$	1.23	1.18
spatial distribution	SKIZ cell area variation coefficient, CV(A_{SKIZ})	1.14	0.75

5. Influence of second phase particles on grain refinement

One of the most important aspects of the presence of second phase particles during SPD processing is their influence on the process of grain refinement. This problem has been analysed separately for primary intermetallic particles and secondary precipitates.

TEM observations have revealed that the presence of relatively large primary intermetallic particles changes the microstructure evolution during processing by hydrostatic extrusion (Fig. 3). Away from the particles, the microstructure consists of elongated grains enveloped by boundaries nearly parallel to the extrusion direction. In the vicinity of an intermetallic particle, the equiaxial and very fine grains are visible. It means that the relatively large particles favour the process of grain refinement. It should be, however, noted that their influence is limited to the neighbouring zone of thickness comparable to the particle size. These observations are in good agreement with those obtained for an aluminium alloy containing large amount of intermetallic particles [13] for which the process of grain refinement is much faster than in the case of single phase alloy.

The role of precipitates in terms of grain refinement is completely different. Small precipitates reduce the ability of a material to grain refinement. The 2017 aluminium alloy, when processed immediately after water quenching, exhibits well developed microstructure with the grain size of 90 nm (Fig. 4a). The misorientation angle measurements have shown (Fig. 4b) that a significant fraction of grain boundaries have high value misorientation angle.

The same alloy processed after aging have less developed grain structure (Fig. 4c), greater grain size (about 150 nm) and only limited number of high angle grain boundaries (Fig. 4d). This indicates that precipitates delay the formation of high angle boundaries. This may be due to the fact that the strain energy transferred to the sample during deformation is used not only for grain refinement process, but also for fracture and fragmentation of precipitates. It is probable that if the extrusion of the aged sample is continued, the grain refinement and the fraction of high angle boundaries would eventually reach the same level as that of the sample deformed in as-quenched state.

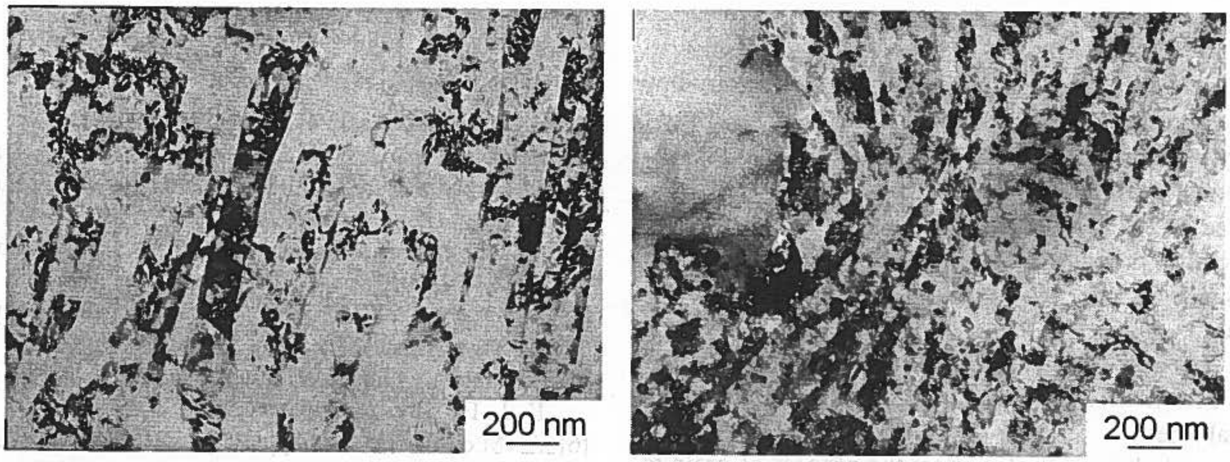

Fig. 3. The microstructure of hydrostatically extruded 2017 aluminium alloy: (a) in the region without particles and (b) in the vicinity of a particle

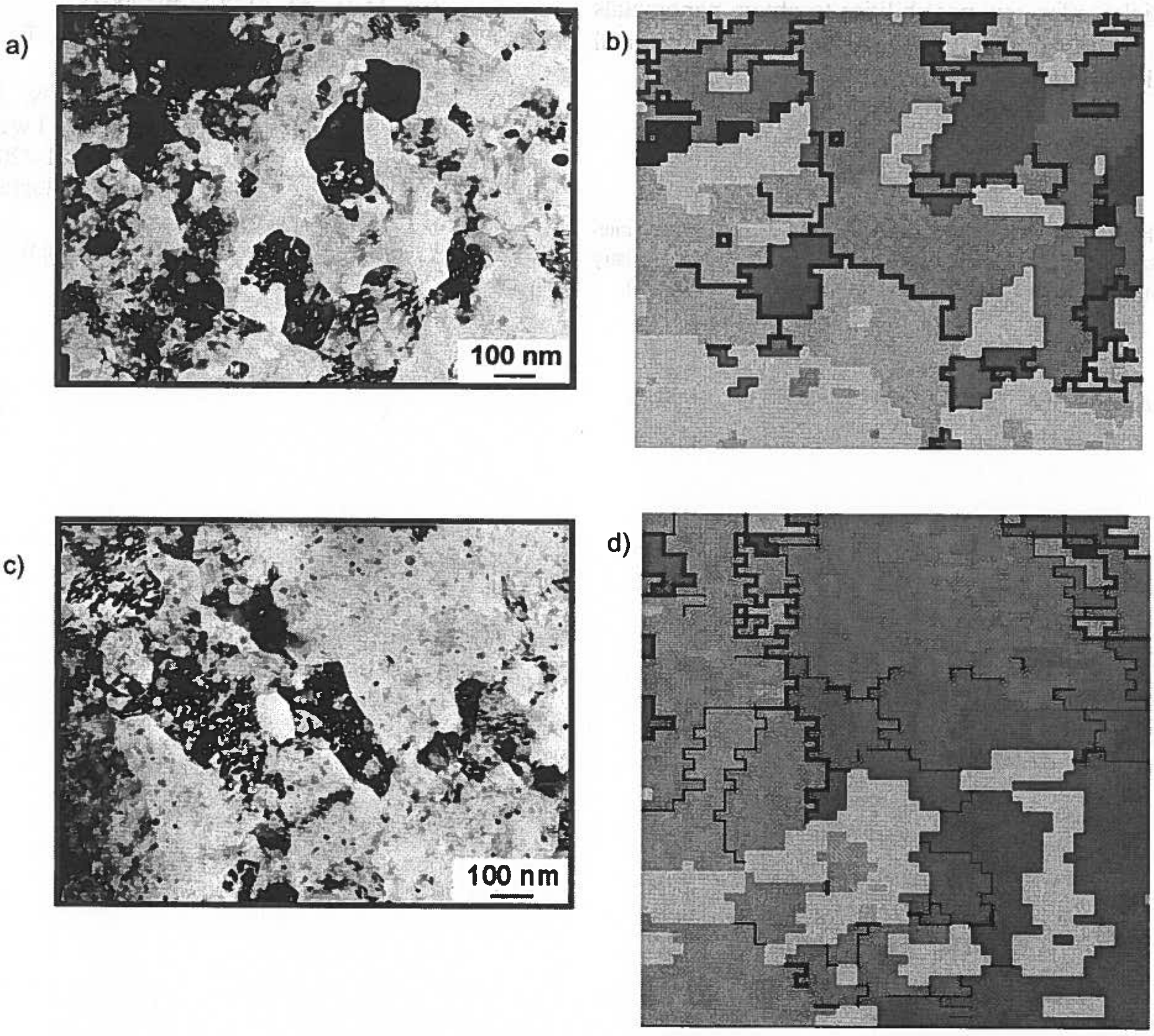

Fig. 4. The microstructures (a,c) and orientation maps (b, d) of 2017 aluminium alloy processed immediately after water quenching (a, b) and after aging at 200°C for 3 hours (c, d)

6. Conclusions

The results presented in this communication allow to point out the following conclusions:

- Hydrostatic extrusion is an effective way to produce nanocrystalline materials;
- Size, shape and spatial distribution of primary intermetallic particles in the aluminium alloy are significantly affected by the process of hydrostatic extrusion;
- Processing by hydrostatic extrusion leads to the transformation of plate-like Θ′ precipitate into spherical one;
- Second phase particles affect the process of grain refinement which is favoured by the presence on large primary intermetallics whereas delayed by small precipitates.

The described changes in precipitates and intermetallic particles offer new possibilities to obtain nanometals having improved properties (e.g. fracture toughness and thermal stability).

Acknowledgements

Helpful comments from Professor K. J. Kurzydlowski are gratefully acknowledged. The work was supported by the Polish Ministry for Science and Higher Education (grant number 3T08A 06430).

Received: 20 September 2006.

REFERENCES

[1] E. Ma, Mat. Trans. **47**, 1269-1274 (2006).

[2] M. Lewandowska, H. Garbacz, W. Pachla, A. Mazur, K. J. Kurzydłowski, Materials Science – Poland **23**, 279-286 (2005).

[3] M. Lewandowska, Solid State Phenomena **114**, 109-114 (2006).

[4] M. Lewandowska, Journal of Microscopy **226** (2006) in press.

[5] Y. Ivanisenko, W. Łojkowski, R. Z. Valiev, H.-J. Fecht, Acta mater. **51**, 5555-5570 (2003).

[6] A. Morawiec, J. Appl. Cryst. **32**, 788-798 (1999).

[7] A. W. Zhu, Acta Mater. **46**, 3211-3220 (1998).

[8] Y. Brechet, F. Louchet, C. Marchionni, J.-L. Verger-Gaugry, Phil. Mag. **38**, 353-360 (1987).

[9] C. Xu, M. Furukawa, Z. Horita, T. G. Langdon, Acta Mater. **51**, 6139-6149 (2003).

[10] Y. Nishida, H. Arima, J.-C. Kim, T. Ando, Scripta Mater. **45**, 261-266 (2001).

[11] A. Ma, K. Suzuki, Y. Nishida, N. Saito, I. Shigematsu, M. Takagi, H. Iwata, A. Watazu, T. Amura, Acta Mater. **53**, 211-220 (2005).

[12] W. A. Spitzig, J. F. Kelly, Metallography **18**, 235-261 (1985).

[13] P. J. Apps, J. R. Bowen, P. B. Prangnell, Acta Mater. **51**, 2811-2922 (2003).