Grain boundary assembles developed in an austenitic stainless steel during large strain warm working

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ABSTRACT

The evolution of strain-induced grain boundaries in a 304-type austenitic stainless steel during multiple warm forging was studied. The developed grain boundary distributions significantly depended on the processing temperature. The misorientation distribution of deformation boundaries that evolved after total strain of 4 at 500 °C is characterised by a sharp maximum against small angles below 10° and a flat-type distribution with almost the same fractions for different misorientations for high-angle boundaries. On the other hand, two sharp peaks on the boundary misorientation distribution are developed after processing at 800 °C. One of them corresponds to low-angle subboundaries and the other results from twin boundaries with misorientations of 60°. The difference in the characters of strain-induced grain boundaries is associated with different mechanisms of dynamic recrystallization operating at 500 and 800 °C.

1. Introduction

Properties of metallic materials depend significantly on their microstructures. One of the most important structural parameters affecting mechanical behaviour of various metals and alloys is the grain size. A beneficial combination of mechanical properties can be attained by grain refinement. The ultrafine grained materials with a grain size below about a micrometer have been shown to possess high strength, enhanced toughness, and sufficient ductility [1–4]. The ultrafine grained microstructures in metallic semi-products can be obtained by plastic deformations, which are accompanied by the development of dynamic recrystallization [5–7]. Since the dynamic grain size sensitively depends on processing temperature, the ultrafine grained microstructures can be developed under warm deformation conditions, i.e. during plastic working at relatively low temperatures of about 0.5 of melting point (0.5Tm) [8–10].

Another important structural parameter affecting the properties of metallic materials is a grain boundary character. The grain boundary engineering focusing on the development of optimal grain boundary character distributions in structural metallic materials has attracted a great interest among materials scientists around the world [11–14]. The grain boundary engineering is mainly aimed to control the fraction of so-called special boundaries, i.e. those having dense coincident site lattice (CSL), which is characterised by the reciprocal density of coinciding sites, Σ. In face centered cubic metals and alloys with low stacking fault energy (SFE), most of the special boundaries developed by grain boundary engineering are $\Sigma 3$ or $\Sigma 3^n$ CSL boundaries related to twin boundaries. In spite of great efforts in the development of ultrafine grained materials by warm working [4,7,15], however, the effect of processing conditions on the character of strain-induced grain boundaries in these materials has not been clarified.

Generally, the dynamic recrystallization processes in metallic materials with low SFE can be categorized as discontinuous and continuous ones [6–8]. During the discontinuous dynamic recrystallization, the new grains nucleate due to local bulging of grain boundaries and then the recrystallizing nuclei grow at expense of work hardened grains [16]. The recrystallized

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microstructures, therefore, consist of two structural components, i.e. recrystallized and work hardened ones. As a result, the final grain boundary assembly is characterised by the presence of strain-induced and recrystallized grain boundaries. The fractions of recrystallized and strain-induced boundaries depend on kinetics of dynamic recrystallization and can be controlled by variation of processing conditions. In contrast to discontinuous dynamic recrystallization, continuous dynamic process does not involve the extended growth of recrystallized grains [8]. The grain boundary characters in such microstructures should be the same with those developed in deformation microstructures. Therefore, the utilization of different recrystallization mechanisms is a powerful tool for development of desired microstructures with various grain boundary characteristics. The formation of annealing twins, which are inherent in discontinuous dynamic recrystallization, should lead to a large fraction of special grain boundaries. On the other hand, continuous dynamic recrystallization can be used to retain the deformation origin of grain boundary distribution.

The aim of the present study is to clarify the effect of processing conditions on the grain boundary distributions, which develop in dynamically recrystallized ultrafine grained structures. A 304-type austenitic stainless steel was selected as a typical representative of materials with low SFE, exhibiting discontinuous dynamic recrystallization during hot working. The study is focused on the relationship between the operating mechanisms of dynamic recrystallization and the developed grain boundary assembles.

2. Experimental

An S304H austenitic stainless steel, 0.10%C–18.2%Cr–7.85%Ni–2.24%Cu–0.50%Nb–0.008%B–0.12%N–0.95%Mn–0.10%Si and balance Fe (all in weight%), was used as the starting material. The original hot rolled steel bar was annealed at 1100 °C for 30 min. Such treatment resulted in the development of uniform microstructure with an average grain size of about 10 μm (Fig. 1a) containing homogeneous distribution of dispersed carbonitrides with an average size ~50 nm [17]. The annealed microstructure exhibited a special grain boundary distribution, which was composed by high-angle boundaries including a large fraction (approximately 0.5) of Σ3 twin boundaries with misorientations around 60° (Fig. 1b). The fraction of low-angle boundaries with misorientations below 15° did not exceed 0.03.

The warm deformation was carried out by means of isothermal multi-pass forging (compression) tests with a change of the loading direction in 90° in order of three orthogonal axes as schematically shown in Fig. 2 [18,19] by using rectangular samples with initial dimension of 10 mm × 12 mm × 15 mm. The samples were compressed at 500 and 800 °C under a strain rate of 10^{-3} s^{-1} to a strain of about 0.4 in each pass followed by water quenching. The samples were ground to rectangular shape before each subsequent test and then reheated to deformation temperature with a heating rate of ~35°/min. The structural investigations were carried out on the sample sections parallel to the compression axis in the last pass by using a Quanta 600F scanning electron microscope equipped with an electron back scattering diffraction (EBSD) analyser incorporating an orientation imaging microscopy (OIM) system. The specimens were prepared by electro-polishing using a solution of 10% perchloric acid in glacial acetic acid. The following step sizes were used in the EBSP analyses: 100 nm for the specimens subjected to 1–3 forging passes at 500 and 800 °C, 30 nm for specimens processed by 5–10 forging passes at 500 °C, and 60 nm for specimens processed by 5–10 forging passes at 800 °C. The OIM images were

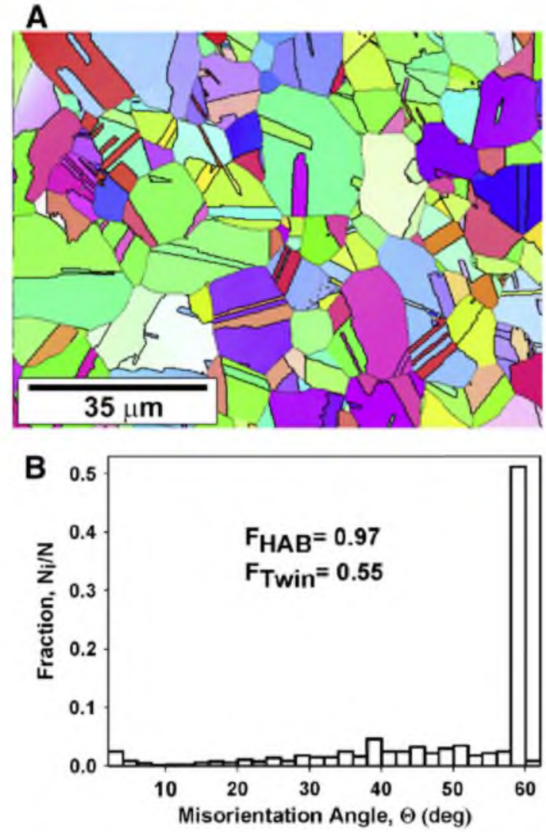

Fig. 1 – Typical microstructure (a) and boundary misorientation distribution (b) developed in an austenitic stainless steel after annealing at 1100 °C. Twin boundaries are indicated by the thick black lines in the micrograph.

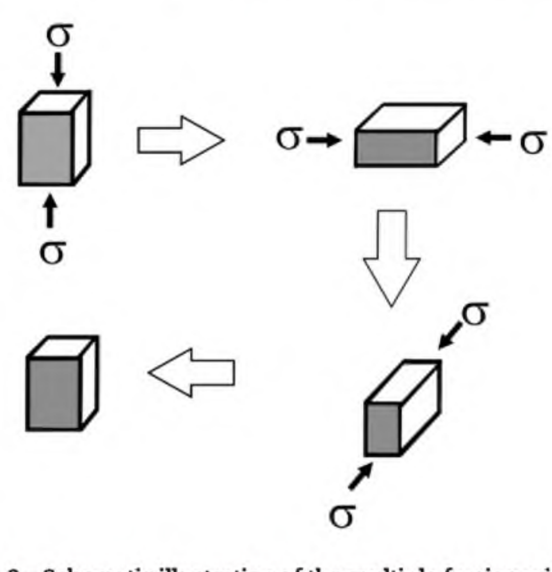

Fig. 2 – Schematic illustration of the multiple forging with sequential change of the loading direction.

subjected to cleanup procedure, setting a minimal confidence index of 0.1. The grain size was measured as a distance between high-angle boundaries along the last pass compression axis by using TSL software (version 5.2).

3. Results

3.1. *Deformation Behaviour*

Depending on the deformation temperature two kinds of deformation behaviour are observed in a 304-type austenitic stainless steel subjected to multiple forging (Fig. 3). Dynamic recovery type behaviour [5] takes place during deformation at relatively low temperature of 500 °C (about 0.45Tm). The flow stress drastically increases during the first forging pass. Remarkable strain hardening is also observed during the second forging pass, while further processing is accompanied by an apparent strain softening. However, the flow stress envelope curve plotted over 10 forging passes by dotted line in Fig. 3 does not demonstrate any strain softening. The flow curve envelope indicates a rapid strengthening in the strain range of $0<\varepsilon<0.5$ followed by a gradual strengthening during the straining to about 3, when the flow stress envelope approaches saturation level of about 800 MPa. Such flow behaviour is associated with a progressive strengthening by multiple forging. The yield stress in each subsequent pass is either almost the same or just above the flow stress in the preceding pass just before unloading.

On the other hand, dynamic recrystallization type deformation behaviour [5,16] takes place during multiple forging at elevated temperature of 800 °C (about 0.6Tm). Significant strain strengthening is clearly seen in the first forging pass, whereas subsequent passes are characterised by a steady state flow behaviour, when the flow stress approaches its saturation right after reloading. It should be noted that after the forth forging pass the flow stresses after reloading are somewhat lower than the flow stresses before unloading. Therefore, the envelope flow curve shows single peak type behaviour. Namely, the flow stress increases to its maximum with straining to about 0.5, then the strain softening takes place followed by a steady state flow at large strains of above 2.5.

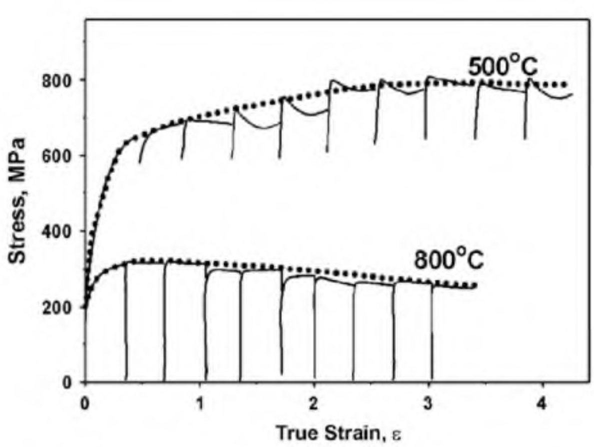

Fig. 3 – True stress–strain curves for an austenitic stainless steel under multiple forging.

3.2. *Deformation Microstructures during Multiple Forging at 500 °C*

Typical microstructures evolved after the second, third and tenth forging passes at 500 °C are shown in Fig. 4. The multiple forging brings about the development of spatial net of strain-induced boundaries including low-angle subboundaries and high-angle grain boundaries. It is clearly seen in Fig. 4a and b that the strain-induced grain boundaries subdivide the original

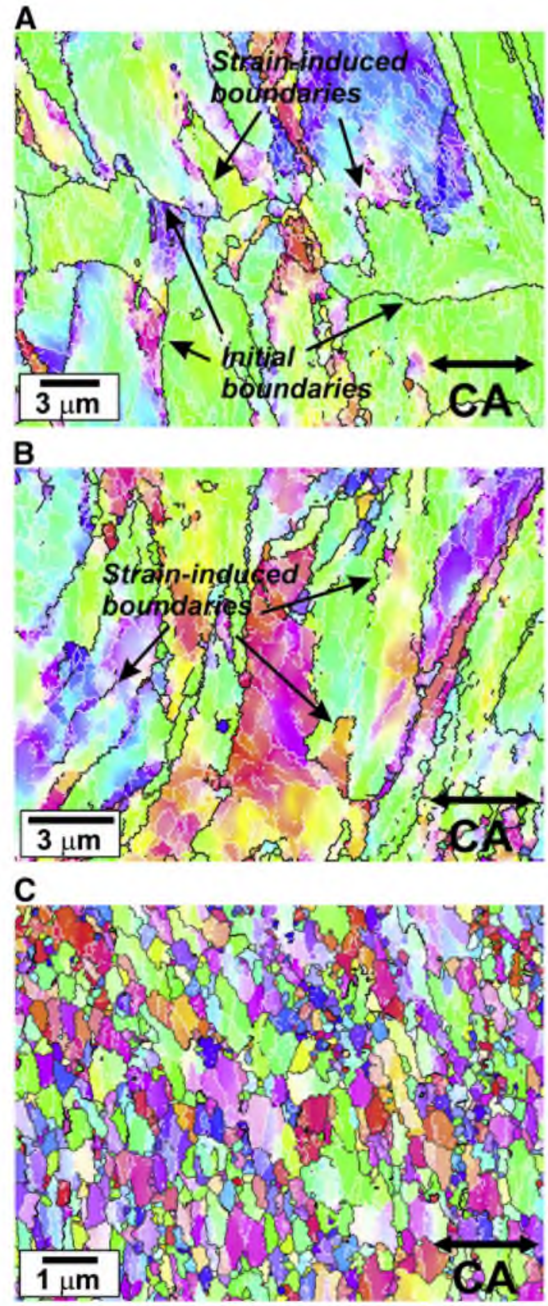

Fig. 4 – Typical microstructures evolved in an austenitic stainless steel after (a) 2, (b) 3 and (c) 10 forging passes at 500 °C. Low- and high-angle boundaries are indicated by white and black lines, respectively. Occasionally evolved twin boundaries are indicated by the thick black lines in (c). The direction of the last pass compression axis is indicated by CA.

grains into small fragments, leading to the development of new fine grains. The strain-induced grain boundaries are predominantly involved in vicinities of original grain boundaries, although certain number of them is clearly seen in original grain interiors (Fig. 4a). The misorientation of some strain-induced boundaries varies from large angles (near original grain boundaries) to small angles (in interiors of original grains). A number of the strain-induced grain boundaries increases with straining. After three forging passes, original grain boundaries can hardly be distinguished from strain-induced ones. This is typical of continuous dynamic recrystallization [6,8,10,18]. In such a case the evolution of the strain-induced boundaries with progressively increasing misorientations leads to remarkable grain refinement during plastic working, which is characterised by a recovery type stress–strain behaviour. As a result, the ultrafine grained microstructure with a grain size of 0.22 μm develops at 500 °C after sufficiently large strains (Fig. 4c).

The evolution of boundary misorientations during multiple forging at 500 °C is shown in Fig. 5. A number of low-angle subboundaries are involved by the first forging pass. The originally large fraction of high-angle boundaries decreases to 0.34. The boundary misorientation distribution is characterised by a sharp peak corresponding to low-angle subboundaries and a small diffuse peak against high-angle boundaries with misorientations around 60°. The latter one is evidently associated with a remnant of twin boundaries in the original microstructure. The second forging pass makes the angular distribution of high-angle boundaries flat with almost the same fractions of grain boundaries with various misorientations; the small peak associated with original twin boundaries disappears. The fraction of low-angle boundaries increases up to 0.8, although the low-angle peak decreases and spreads out towards large angles. Upon further multiple forging the fractions of high-angle boundaries increases; they comprise more than 0.4 and 0.5 after the fifth and tenth forging passes, respectively. It is worth noting that the fraction of high-angle boundaries increases homogeneously during processing at 500 °C. Almost flat misorientation distribution with the same fractions of about 0.03 for different misorientations develops for high-angle boundaries.

3.3. *Deformation Microstructures during Multiple Forging at 800 °C*

The structural changes during multiple forging at 800 °C are illustrated by Figs. 6 and 7, which show representative deformation microstructures evolved after the second and tenth passes, respectively. The second forging pass is characterised by a maximum at the stress–strain curve envelope, and the tenth pass corresponds to steady state deformation behaviour. The single peak stress–strain curves were frequently observed during hot working accompanied by discontinuous dynamic recrystallization, when the new grains grew out consuming work hardened surroundings [5,6,10,16]. Therefore, Fig. 6 corresponds to the beginning stage of discontinuous dynamic recrystallization. The bulging of frequently serrated original grain boundaries is clearly seen in the enlarged portion of micrograph. Assisted by strain-induced boundaries such bulges result in the development of recrystallizing nuclei, which are able to grow during the following deformation. The repeated formation of a number of recrystallized grains during multiple forging leads to the development of uniform ultrafine grained microstructure with a grain size of 0.69 μm after 10 forging passes at 800 °C (Fig. 7). The growth of recrystallization nuclei is accompanied by the appearance of annealing twins as indicated by thick black lines in Fig. 7.

The boundary misorientation distribution evolved after one forging pass at 800 °C is identical to that developed at 500 °C (Fig. 8). The same peaks for low- and high-angle boundaries are developed after the first forging pass irrespective of different processing temperatures (cf. Figs. 5 and 8). Similar to multiple

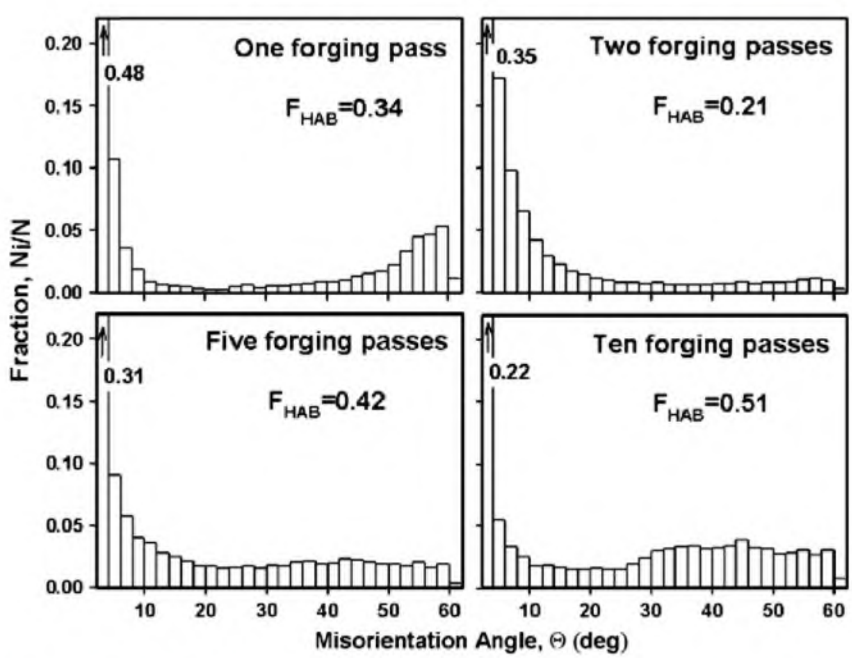

Fig. 5 – Misorientation distribution for boundaries developed in an austenitic stainless steel during multiple forging at 500 °C.

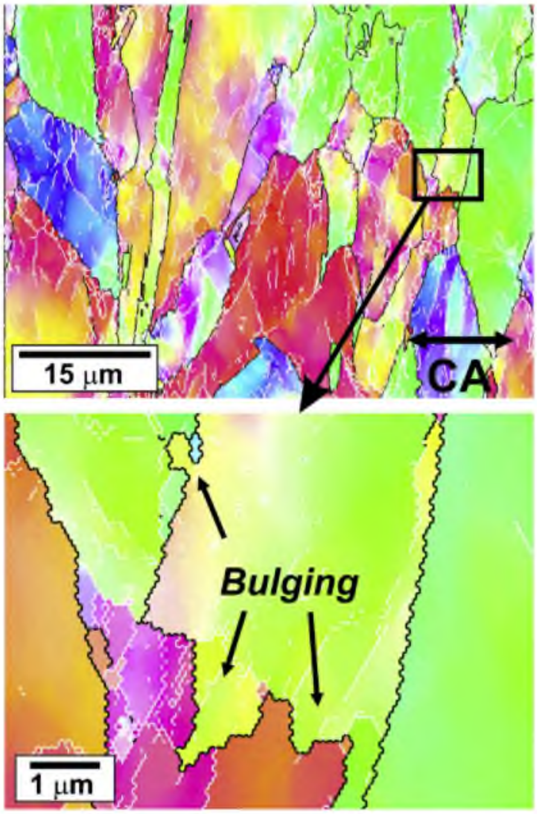

Fig. 6 – Representative OIM micrograph for deformation microstructure evolved in an austenitic stainless steel after two forging passes at 800 °C. Low- and high-angle boundaries are indicated by white and black lines, respectively. The direction of the last pass compression axis is indicated by CA.

forging at 500 °C, the subsequent working at 800 °C results in gradual decrease of the fraction of high-angle boundaries, especially those related to original twin boundaries with misorientations around 60°, and forms almost flat misorientation

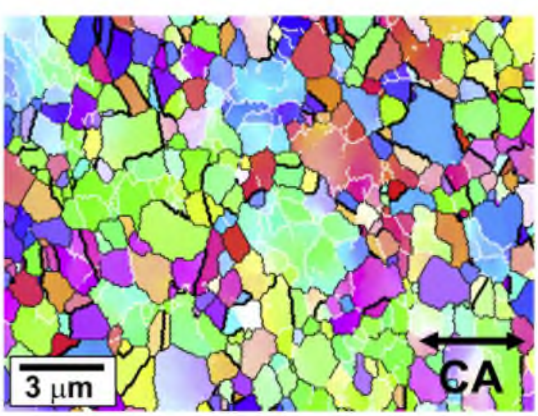

Fig. 7 – Typical microstructure evolved in an austenitic stainless steel after 10 forging passes at 800 °C. Low- and high-angle boundaries are indicated by white and black lines, respectively. The twin boundaries are shown by thick black lines. The direction of the last pass compression axis is indicated by CA.

distribution for high-angle boundaries after five forging passes. Further processing at 800 °C is accompanied by increase in the fraction of high-angle boundaries to 0.55. However, the misorientation distribution evolved after 10 forging passes at 800 °C differs from that at 500 °C by the appearance of a sharp peak against 60° misorientations. This is associated with an increasing number of annealing twins, which result from the operation of discontinuous dynamic recrystallization at relatively high temperature of 800 °C. The fraction of Σ3 twin boundaries comprises 0.12 after the final forging pass. It is worth noting that percentage of Σ9 and Σ27 boundaries is negligibly small, it does not exceed 1%. Therefore, the bimodal misorientation distribution with two peaks corresponding to low-angle boundaries and Σ3 twin boundaries develops after multiple forging to large total strains at 800 °C.

4. Discussion

The present results suggest that ultrafine grained microstructures can be successfully developed in a 304-type austenitic stainless steel by multiple forging to large total strains of about 4 at temperatures of 0.45 and 0.6Tm. The grain refinement is assisted by the presence of second phase particles, which stimulate the development of local strain gradients upon straining [20]. Moreover, the second phase particles slow down the grain coarsening kinetics [15], leading to the submicrocrystalline structure even at relatively high deformation temperature of 800 °C. The developed ultrafine grained structures are characterised by a rather large fraction of low-angle boundaries of about 0.5 irrespective of processing temperature within the studied temperature range. This is a common feature of ultrafine grained structures developed by large strain deformation because of dislocation cell substructure inherent in work hardened grains [6,8,16,18]. However, the fraction of special boundaries depends remarkably on the processing conditions (Fig. 9). The change in the forging temperature from 500 to 800 °C results in more than threefold increase in the fraction of twin boundaries finally developed in the ultrafine grained structures. The difference in the boundary distributions is associated with different mechanisms of dynamic recrystallization operating at 500 and 800 °C.

The sequences of structural changes in the studied material during multiple forging at 500 and 800 °C are schematically shown in Fig. 10. Each forging pass at 500 °C brings about the formation of strain-induced boundaries, which subdivide original grains into small misoriented fragments. The number of strain-induced boundaries and their misorientation increases upon multiple deformations, finally leading to the development of submicrocrystalline structure, which is composed by low to high-angle boundaries. Relatively low deformation temperature of 500 °C inhibits any diffusion-controlled grain boundary motion irrespective of large total strain and, therefore, high driving force for boundary migration [21]. Crystallographic parameters of the developed boundaries are controlled by misfit dislocations, which are accumulated at subboundaries during deformation [22]. Since the multiaxial deformation is not accompanied by a development of any special texture [8], almost random grain boundary distribution develops during the multiple forging. Some special boundaries can be occasionally involved; however, their number is quite small (Fig. 9).

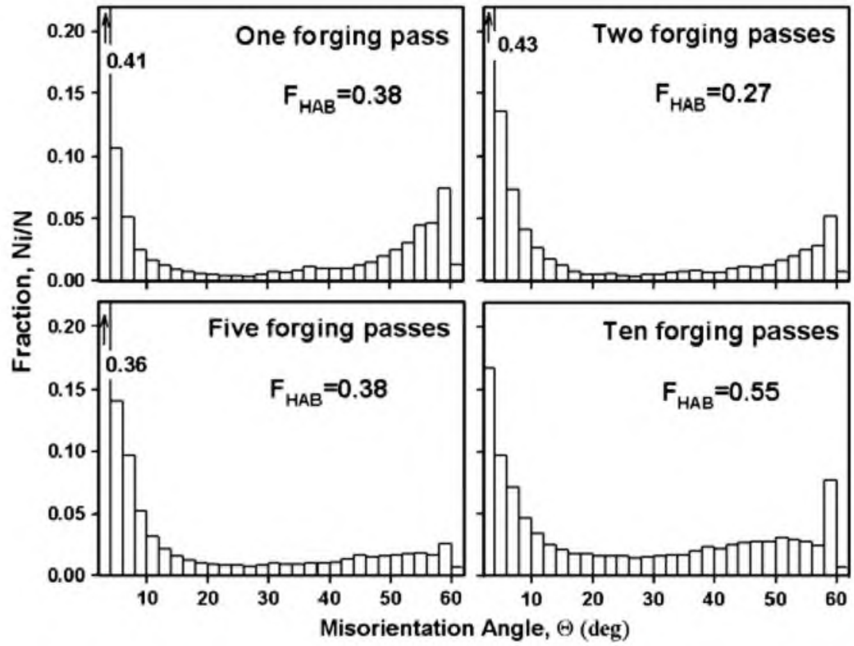

Fig. 8 – Misorientation distribution for boundaries developed in an austenitic stainless steel during multiple forging at 800 °C.

On the other hand, the development of new fine grains at 800 °C is assisted by grain boundary migration (Fig. 10), which is the essence for the bulging mechanism of discontinuous recrystallization [10]. This deformation temperature is not high enough for rapid grain boundary migration over large distances, so only local boundary bulging can occur. Each forging pass leads to the formation of fine bulges at grain boundaries. These bulges are separated from the parent grains by strain-induced boundaries upon further deformation and, thus, become new grains. The deformation subboundaries with increasing misorientations also develop during processing at 800 °C. Some of such strain-induced subboundaries may evolve in grain boundaries leading to grain refinement concurrently with the operation of bulging mechanism. An increase of strain stimulates further bulging of grain boundaries. The number of new fine grains increases during deformation, finally leading to homogeneous ultrafine grained microstructure. The grain boundary migration is accompanied by appearance of annealing twins. It should be noted that the twin boundaries developed during multiple deformations can be transformed to ordinary grain boundaries

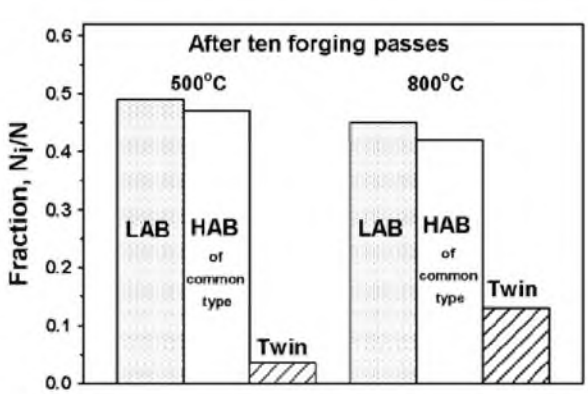

Fig. 9 – Fractions of different boundaries, which developed in an austenitic stainless steel after 10 forging passes at 500 and 800 °C.

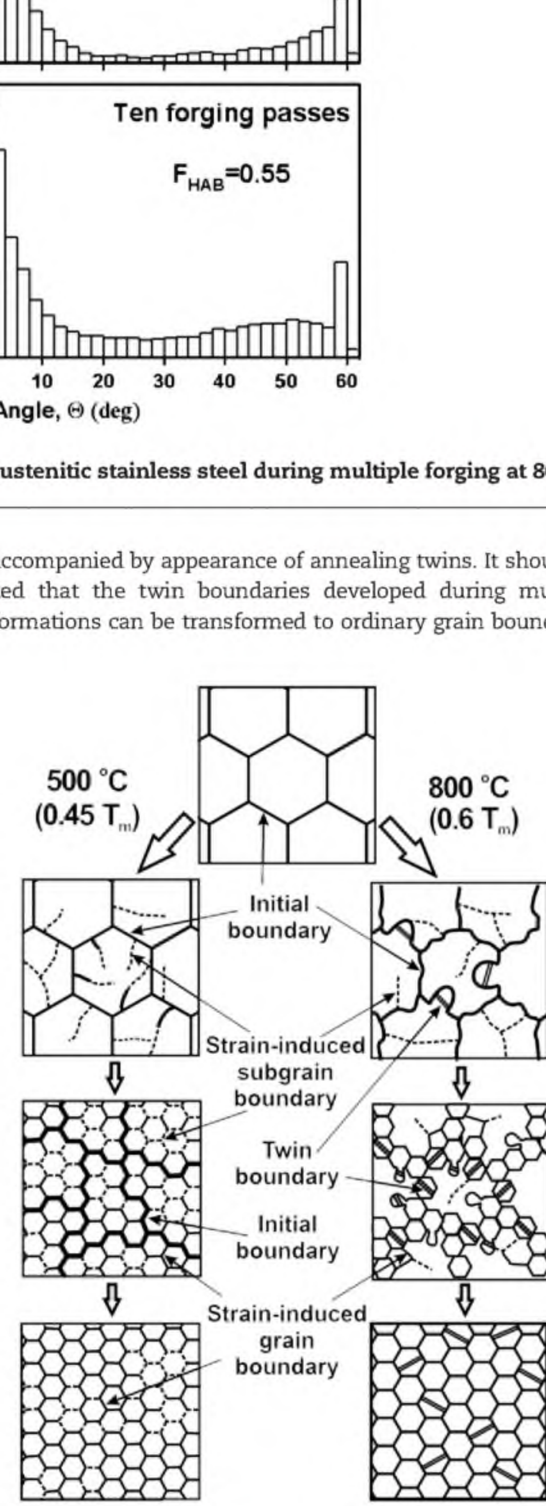

Fig. 10 – Schematic representation of the structural changes in an austenitic stainless steel during multiple forging at 500 and 800 °C.

by subsequent deformation as it happened with original microstructure after the first forging pass. Therefore, the special boundaries in the ultrafine grained microstructure after multiple forging at 800 °C do correspond to the first generation twins, which are evolved mostly by the final forging pass. The fraction of special boundaries in the ultrafine grained microstructure developed by multiple forging at 800 °C comprises 0.12, which is much smaller than that of 0.5 in the original annealed microstructure, but it is significantly larger than that of 0.04 evolved by the processing at 500 °C.

5. Conclusions

The misorientation distribution of boundaries evolved in ultrafine grained microstructures developed in an austenitic stainless steel by multiple forging at temperatures of 500 and 800 °C was studied. The main results can be summarised as follows.

1. The multiple forging at 500 °C resulted in the development of ultrafine grained microstructure with an average grain size of 0.22 μm. The developed microstructure was characterised by almost the same fractions of low- and high-angle boundaries with negligibly small fraction of special boundaries.
2. The ultrafine grained microstructure with an average size of 0.69 μm was developed by multiple forging at 800 °C. The fractions of low- and high-angle boundaries were 0.45 and 0.55, respectively. The latter ones included relatively large fraction of 0.12 of special boundaries.
3. The difference in the boundary characters was associated with different mechanisms of dynamic recrystallization operating at 500 and 800 °C. Continuous dynamic recrystallization at 500 °C resulted in the development of strain-induced boundaries of common type, while discontinuous recrystallization mechanism during deformation at 800 °C involved grain boundary migration leading to the formation of annealing twins.

Acknowledgements

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REFERENCES

[1] Morris DG. Strength and ductility of nanocrystalline materials: what do we really understand? In: Dinesen AR, Eldrup M, Jensen D, Linderoth S, Pedersen TB, Pryds NH, et al, editors. Science of Metastable and Nanocrystalline Alloys. Roskilde, Denmark: Riso National Laboratory; 2001. p. 89–104.

[2] Wang Y, Chen M, Zhou F, Ma E. High tensile ductility in a nanostructured metal. Nature 2002;419:912–4.

[3] Kimura Y, Inoue T, Yin F, Tsuzaki K. Inverse temperature dependence of toughness in an ultrafine grain-structure steel. Science 2008;320:1057–60.

[4] Valiev RZ, Langdon TG. The art and science of tailoring materials by nanostructuring for advanced propertoes using SPD techniques. Adv Eng Mater 2010;12:677–91.

[5] McQueen HJ, Jonas JJ. Recovery and recrystallization during high temperature deformation. In: Arsenault RJ, editor. Treatise on Materials Science and Technology, vol. 6. New York: Academic Press; 1975. p. 394–490.

[6] Wusatowska-Sarnek AM. The new grain formation during warm and hot deformation of copper. J Eng Mater Technol 2005;127:295–300.

[7] Belyakov A. Changes in the grain structure of metallic materials upon plastic treatment. Phys Met Metall 2009;108: 390–400.

[8] Belyakov A, Gao W, Miura H, Sakai T. Strain induced grain evolution in polycrystalline copper during warm deformation. Metall Mater Trans A 1998;29A:2957–65.

[9] Belyakov A, Sakai T, Miura H, Kaibyshev R. Grain refinement under multiple warm deformation in 304 type austenitic stainless steel. Iron Steel Inst Jpn Int 1999;39:592–9.

[10] Dudova N, Belyakov A, Sakai T, Kaibyshev R. Dynamic recrystallization mechanisms operating in a Ni-20%Cr alloy under hot-to-warm working. Acta Mater 2010;58:3624–32.

[11] Watanabe T. An approach to grain boundary design for strong and ductile polycrystals. Res Mech 1984;11:47–84.

[12] Randle V. Twinning-related grain boundary engineering. Acta Mater 2004;52:4067–81.

[13] Wang WG, Yin F, Guo H, Li H, Zhou B. Effecs of recovery treatment after strain on the grain boundary character distributions of subsequently cold rolled and annealed Pb-Ca-Sn-Al alloy. Mater Sci Eng 2008;A491:199–206.

[14] Fang X, Wang W, Cai Z, Qin C, Zhou B. The evolution of cluster of grains with $\Sigma 3^n$ relationship in austenitic stainless steel. Mater Sci Eng 2010;A527:1571–6.

[15] Humphreys FJ, Prangnell PB, Bowen JR, Gholinia A, Harris C. Developing stable fine-grain microstructures by large strain deformation. Phil Trans R Soc Lond 1999;357:1663–81.

[16] Sakai T, Jonas JJ. Dynamic recrystallization—mechanical and microstructural considerations. Acta Metall 1984;32:189–209.

[17] Yanushkevich Z, Mogucheva A, Tikhonova M, Belyakov A, Kaibyshev R. Structural strengthening of an austenitic stainless steel subjected to warm-to-hot working. Mater Character 2011;62:432–7.

[18] Belyakov A, Sakai T, Miura H, Tsuzaki K. Grain refinement in copper under large strain deformation. Philos Mag 2001;A81: 2629–43.

[19] Nakao Y, Miura H. Nano-grain evolution in austenitic stainless steel during multi-directional fordging. Mater Sci Eng 2010;528:1310–7.

[20] Belyakov A, Murayama M, Sakai Y, Tsuzaki K, Okubo M, Eto M, et al. Development of a high-strength high-conductivity Cu–Ni–P alloy. Part II: processing by severe deformation. J Electron Mater 2006;35:2000–8.

[21] Belyakov A, Sakai T, Miura H. Fine-grained structure formation in austenitic stainless steel under multiple deformation at 0.5 Tm. Mater Trans JIM 2000;41:476–84.

[22] Argon AS, Haasen P. A new mechanism of work hardening in the late stages of large strain plastic flow in F.C.C. and diamond cubic crystals. Acta Metall Mater 1993;41:3289–306.