J. ZRNÍK, I. MAMUZIĆ, S. V. DOBATKIN, Z. STEJSKAL, L. KRAUS

ISSN 0543-5846
METABK 46 (1) 21-27 (2007)
UDC - UDK 669.15–194–135–174:620.17 =111

LOW CARBON STEEL PROCESSED BY EQUAL CHANNEL ANGULAR WARM PRESSING

Received - Primljeno: 2005-10-20
Accepted - Prihvaćeno: 2006-08-10
Original Scientific Paper - Izvorni znanstveni rad

Low carbon steel AISI 10 was subjected to a severe plastic deformation technique called Equal Angular Channel Pressing (ECAP) at different increased temperatures. The steel was subjected to ECAP with channel's angle ϕ = 90°, at different temperature in range of 150 - 300 °C. The number of passes at each temperature was N = 3. Light, scanning electron microscopy (SEM) and transmission electron microscopy (TEM) of thin foils were used to study the formation of substructure and ultrafine grains in deformed specimens. The size of newly born polygonized grains (subgrains and/or submicrocrystalline grains) is in range of 300 - 500 μm. The formation of such of predominant submicrocrystalline structure resulted in significant increase of yield stress [R_e] and tensile strength of the steel [R_m].

Key words: *low carbon steel, warm ECAP, structure evolution, dynamic recrystallization, mechanical properties*

Toplo kutno kanalno prešanje niskougljičnog čelika. Analiziran je utjecaj velikih deformacija kod niskougljičnog čelika AISI 10 postupkom kutno kanalnog prešanja (KKP) pri raznim temperaturama. Za proces KKP rabljen je kut ϕ = 90° pri temperaturama 150 - 300 °C. Broj deformacijskih prolaza pri svakoj temperaturi je bio N = 3. Za određivanje stvaranja substruktura i ultrafinih zrna u deformiranim uzorcima rabljene su metode SEM, TEM, optička metalografija. Veličina novonastalih poligonalnih zrna (subzrna i/ili submikrokristalna zrna) su u rasponu 300 - 500 μm. Stvaranje te nove submikrokristalne strukture dovodi do značajnog porasta granice razvlačenja [R_e] i vlačne čvrstoće [R_m].

Ključne riječi: *niskougljični čelik, toplo kutno kanalno prešanje (KKP), razvoj strukture, dinamička rekristalizacija, mehanička svojstva*

INTRODUCTION

Nanomaterials are receiving increasing attention in the technical community and the public at large. Nanomaterials are defined as solids with nanoscale (typically 1 - 100 nm) structures or substructures [1]. Two approaches have been developed to synthesize nanostructured materials [2]. The first one is a "bottom up" approach; the second approach for producing nanostructural materials is a "top-down" approach. The most bottom-up approaches produce nanopowders, while the most successful top-down approach has been via severe plastic deformation (SPD) techniques [3, 4]. The average grain size of materials processed by SPD is above 100 nm and after high pressure torsion even less than 100 nm.

The fabrication of bulk materials with ultrafine grain sizes has attracted a great deal of attention over the past two decades because of their enhanced properties [5 - 8]. The term ultrafine grain structure is referring to nanostructure with grain size less than 100 nm, and submicrocrystalline structure with grains between 100 to 1000 nm. In recent years it has become a worldwide effort to develop manufacturing process to obtain ultrafine grain structures in steels. Currently, there are two main approaches for refining ferrite grains down to the ultrafine grain regime in bulk steels. While the first group comprises advanced thermomechanical processes, the approach of the second group is via employing the various sever plastic deformation techniques to refine structure.

The purpose of the advanced thermomechanical processes is to pursue the processing conditions in order to increase ferrite nucleation sites and suppress grain growth

---

J. Zrník, L. Kraus, Comtes FHT, Ltd., Plzen, Czech Republic, I. Mamuzić, Faculty of Metallurgy University of Zagreb, Sisak, Croatia, S. V. Dobatkin, A. A. Baikov Institute of Metallurgy and Materials Science, Russian Academy of Sciences, Moscow, Russia, Z. Stejskal, West Bohemian University, Plzen, Czech Republic

in a great amount for breaking through the limit of grain refinement of the conventional thermomechanical process. In order to produce ultrafine ferrite grains of 1µm or less, the following types of the processes were found effective: strain-induced ferrite transformation [9], dynamic recrystallization of the austenite during hot deformation with subsequent $\gamma \rightarrow \alpha$ transformation [10], hot rolling in the intercritical region [11], and dynamic recrystallization of ferrite after heavy warm deformation [12]. The ultrafine grain steels achieved 900 MPa strength without additional alloying and showed excellent properties like strength-elongation balance, toughness and fatigue strength [13].

The second group comprises the various sever plastic deformation techniques to refine structure. To introduced large plastic strains into bulk materials the different techniques have been used such as ECAP [5, 14], high pressure torsion (HTP) [15], accumulative roll bonding (ARB) [16, 17], constrain groove pressing (CGP) [18], and others. Especially ECAP generates interest among investigators since it is one of the advanced methods of severe plastic deformation used for metallic materials to produce massive billets with an ultrafine grained structure. Generally, materials fabricated via these ways exhibit a high strength but only limited ductility. Recently different approaches have been considered in an attempt to overcome this limitation [7, 19 - 21]. A significant increase in plastic elongation resulted when structure with bimodal distribution of grain size was fabricated.

Most works are related to the ECAP of pure metals and rather plastic alloys. Make assessment of previous contribution on SPD methods the use of ECAP for commercial steels have been poorly studied. There are the works dealing with the use of ECAP for low carbon steel available. Upon cold ECAP, low carbon steels can only be subjected to two or three passes at channel intersection of 90° without the failure of sample. The two to four passes realized currently upon cold ECAP are insufficient and achievable deformation is insufficient to produce a completely grain structure [22]. To form stable ultrafine grain structure in metals and alloys, ECAP should be carried out at temperature corresponding to the temperature of cold working [23].

The purpose of this work is to study the formation of submicrocrystalline structure commercial low carbon steel AISI 1010 subjected to the large strain warm deformation in dependence of varying temperature of ECAP pressing. The influence of the temperature on the formation of ultrafine grain microstructure and in particular on the course of recovery process was studied by use of scanning and transmission electron microscopy.

MATERIAL AND EXPERIMENTAL

Table 1. **Chemical composition of AISI 1010 steel in weight percent**
Tablica 1. **Kemijski sastav AISI 1010 čelika u težinskim postocima**

Element	C	Mn	Si	P	S	Al	N	As	Cu
wt. pct	0,1	0,42	0,08	0,029	0,05	0,002	-	0,032	0,02

In this work, for experimental the commercial low carbon steel AISI 10 was used. The chemical composition of steel is shown in Table 1. Experimental steel was received as rolled down plate. Prior ECAP pressing, the conventional austenitization of square shaped billets at temperature of 920 °C for 1 hour followed by air cooling was carried out. The deformed ferrite with cementite particles in microstructure of samples after such primary treatment is documented in Figure 1. From treated billets the cylindrical specimens, with initial diameter of 9 mm and length of 50 mm, were cut off for ECAP experiment. The ECAP pressing was performed at four different temperatures of 150, 200, 250 and 300 °C. The angle of intersection of the two channels (φ) was equal to 90°. The ECAP die used for experiment was heated to the consecutive pressing temperature and held for 30

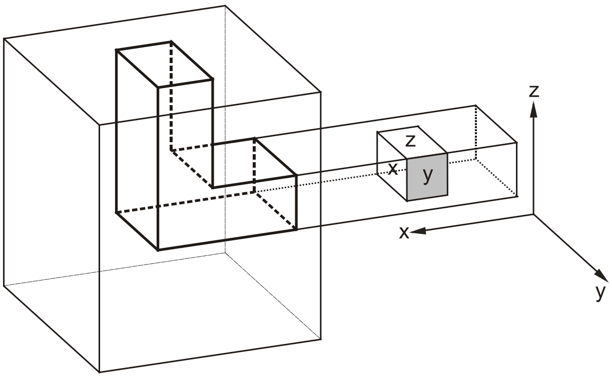

Figure 1. **Principle of ECAP processing**
Slika 1. **Princip kutno kanalnog prešanja (KKP)**

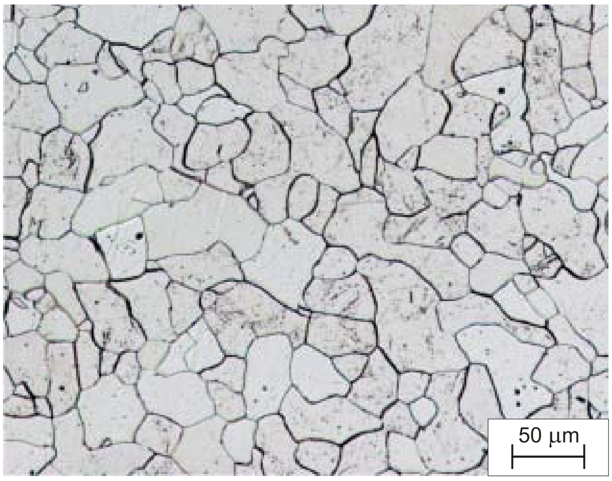

Figure 2. **Optical micrograph of initial structure in billets after thermal treatment**
Slika 2. **Početna mikrostruktura gredice poslije toplinske obradbe (optički mikroskop)**

minutes. The samples heating for 300 s prior pressing was done inside the pre-heated die until it reached the pressing temperature. A 250 tonne hydraulic press was employed and pressing rate of 16 mm/s was used. The temperature of die was controlled in range of ± 1 °C. Each billet was pressed up to a total of three passes (N) through die; the billet between the consecutive passes was rotated along the longitudinal axis by angle equal to 90° in the same sense. Three passes correspond to the total strain of $\varepsilon \sim 3$ before failure. This procedure is generally termed processing route B_c and it was selected because it leads most rapidly to a formation of a homogeneous microstructure of equiaxed grains separated by high angle grain boundaries [24, 25]. It was not expected that strain should be lowered due to static polygonization upon holding in die between passes.

The microstructure of processed samples was examined by NICON 200 light optical (OM), JEOL JSM 6380 (SEM), and JEM-2000 FX (TEM) microscopes. The samples were sliced normal to the longitudinal axis of ECAP pressed billets. The specimens for light optical microscopy were mechanically polished to a 0,05 μm finish and etched using a 3 % Nital solution. Micrographs were taken at 1/3 of from front edge of sample. Samples selection and preparation for microstructural analysis in the SEM and TEM was the same as that for the optical metallography. The TEM samples were produced by mechanical polishing to a thickness of about 50 μm, followed by electropolishing in mixture of perchloric acid (10 %) and acetic acid (90 %) at room temperature and 12 V. Observation in OM and TEM were made on transverse plane X which lies perpendicular to the longitudinal axis of the billet as shown in Figure 1. The selected area diffraction was applied to specify structure development at different temperature of ECAP.

The microhardness was measured using a Vickers device and a load of 300 g. The microhardness distribution over the cross section of sample was measured. The mechanical properties were determined using ZWICK universal testing machine equipped with Multisens extensometer. Tensile

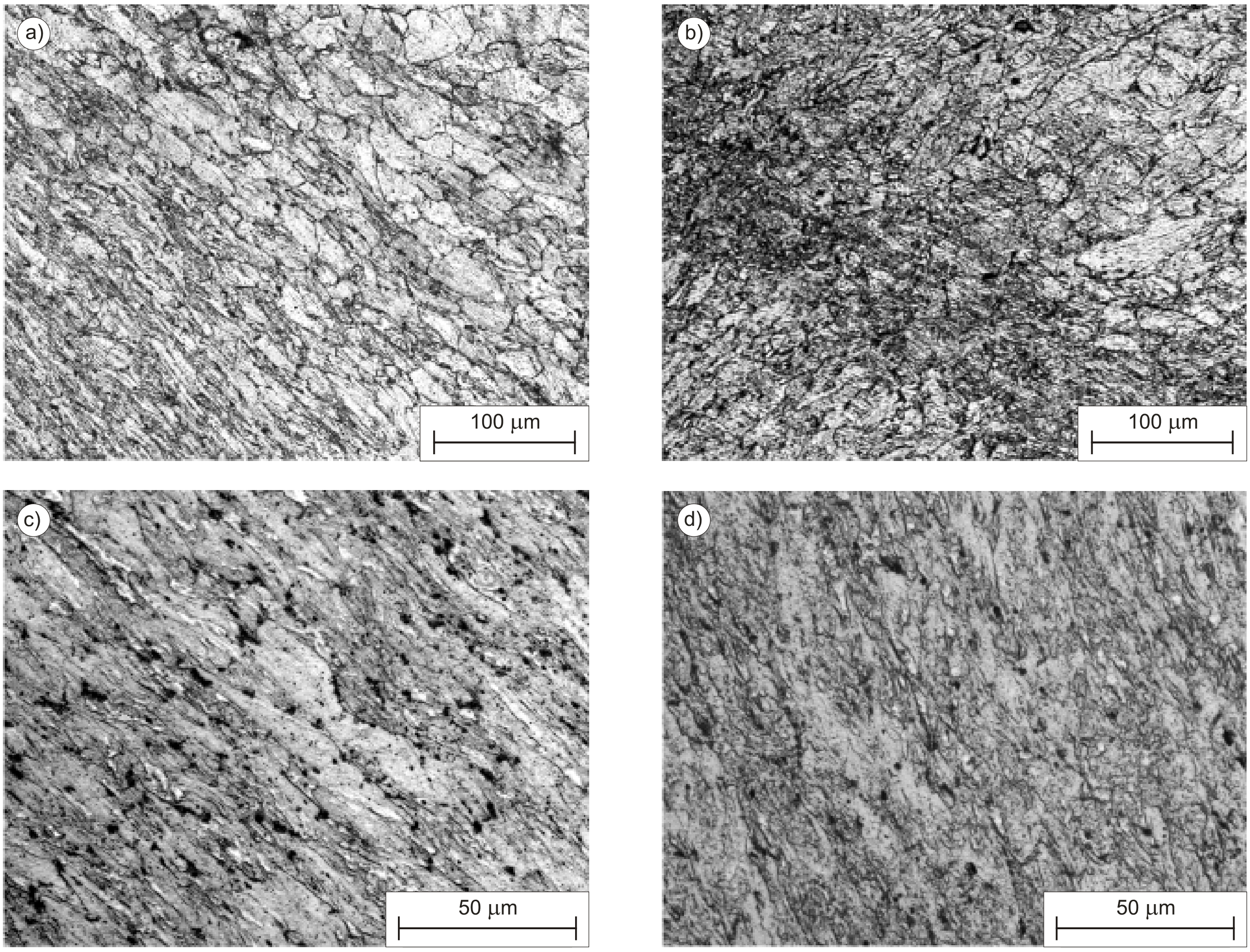

Figure 3. **Optical micrographs taken on the plane perpendicular to billet axis after three ECAP passes at temperature: a) 200 °C; b) 200 °C; c) 150 °C; d) 250 °C**

Slika 3. **Mikrostruktura uzorka okomito na os gredice poslije tri provlake na temperaturama: a) 200 °C; b) 200 °C; c) 150 °C; d) 250 °C (optički mikroskop)** specimens were cut off from the ECAP as-pressed billets with gauge length of 20 mm and the gauge portion equal to 3 mm was lying parallel to the direction of pressing. Tensile tests at a constant crosshead speed of 0,016 mm/s until failure were carried out. The engineering stress-strain curves were constructed.

EXPERIMENTAL RESULTS AND DISCUSSION

Microstructural observation after ECAP

The study of grain structure showed that the initial ferrite with scattered small cementite particles of austenitized structure is uniform over the cross section of a sample. The average grain size of ferrite D_F subjected to ECAP is ~ 50 μm. The cementite particles are precipitated along grain boundaries. Figure 2. represents the OM micrograph of initial austenitized microstructure of the steel. The microstructure is consisted of mainly ferrite grains. The mean linear intercept size of larger and smaller ferrite grains was ~ 100 μm and ~ 10 μm respectively.

The use of repetitive pressing through ECAP die provides and opportunity to develop different microstructures by rotating the samples between consecutive passes. Deformation characteristics for chosen processing route B_c have been analyzed in details in sections normal to longitudinal axis. Both, LOM and SEM micrographs of the as-pressed steel provide evidence of effective straining.

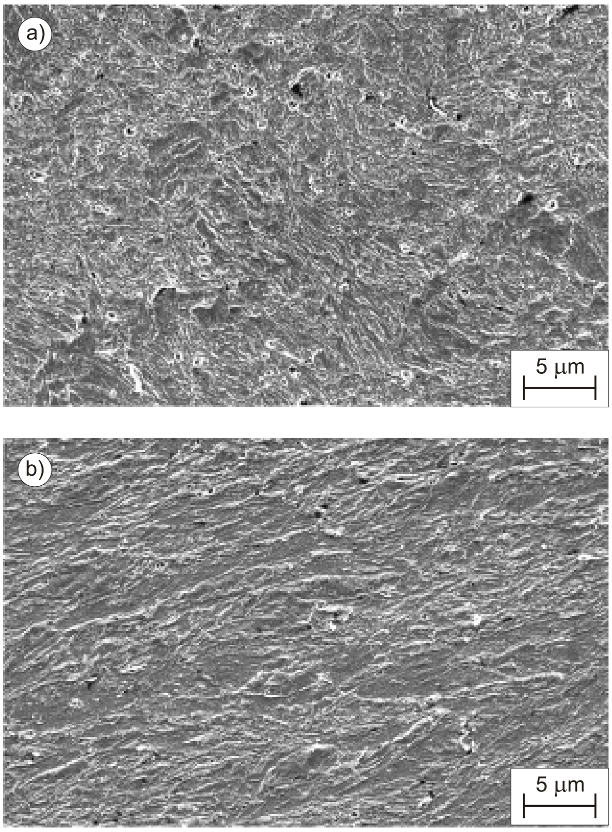

Figure 4. **SEM micrograph of the low carbon steel subjected to three ECAP passes: a) $T = 150$ °C; b) $T = 300$ °C**

Slika 4. **Mikrostruktura niskougljičnog čelika poslije tri provlake: a) $T = 150$ °C; b) $T = 300$ °C (SEM)**

Representative optical micrographs, taken on X plane are shown in Figure 3. Inspecting the effect of three fold die pressing inhomogeneity in straining across the plane X is apparent regardless the applied temperature of ECAP. Locally on this plane still some areas with equiaxed grains are still presents what can be evidence that there they did not experienced heavy strain of ~ 3 as could be expected for route B_c in contrary to results observed for low carbon steels and presented in [24, 25].

To demonstrate the effect of deformation conditions on structure development in low carbon steel during warm ECAP the SEM analysis was used as well. The tendency to flow localization for two selected temperatures of 150 and 300 °C, which are related to the lower and higher temperature of ECAP, can be noticed from Figure 4. On polished and etched surfaces the banded morphology of severely elongated grains is clearly visible (Figure 4.a). A very fine lamellar structure of elongated grains in one direction or irregularly bent lamellar structure (Figure 4.b) is observable on the surface. This fact, however, confirms that sample experienced after three passes heavy deformation but there is no doubt that in microvolumes the deviations due to the different imposed straining are present.

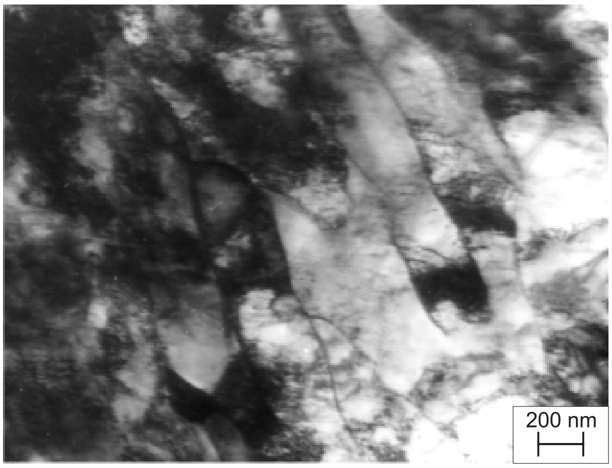

Figure 5. **TEM micrograph of elongated subgrains of ferrite after ECAP at 150 °C**

Slika 5. **Mikrostruktura deformiranih feritnih subzrna poslije KKP na 150 °C (TEM)**

As pointed out previously, multiphase ECAP produces remarkably uniform microstructure if the number of passes is higher of three and if the angle of intersection of channels is $\varphi = 90°$. But no uniformity in strain distribution appeared as it was observed by OM. The microstructure of samples subjected to warm ECAP at 300 °C was further investigated on their normal planes (Figure 1., X plane) by TEM. This analysis provided the substantial evidence that at structure formation not only structure fragmentation modified new ultra fine grain structure but in situ recovery processes contributed by good deal to development of ultra fine grain structure. TEM provided opportunity to analyze the changes in structure which run during ECAP pressing on submicron level.

Figure 5. shows the corresponding TEM image of the deformed ferrite passing ECAP die at temperature of 150 °C. The microstructure mainly consists more or less of parallel bands of elongated grains. The inhomegeneity in grain size and morphology is presented in Figure 6. There have not been observed the effect of increased temperature on onset of recovery process. Higher dislocation density and dislocation cells inside elongated grains are apparent.

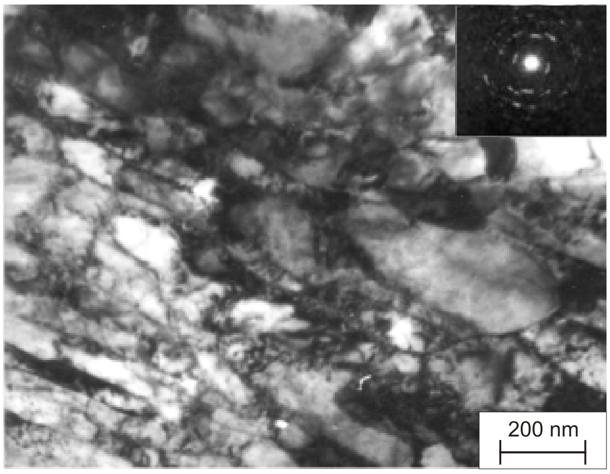

Figure 6. **Different size of fragmented ferrite subgrains produced at ECAP of 150 °C**

Slika 6. **Različite veličine fragmentiranih feritnih subzrna sa KKP na 150 °C**

Figure 7. **TEM micrograph of new subgrains formation at ECAP temperature of 200 °C**

Slika 7. **Mikrostruktura novih subzrna stvorenih KKP na temperaturi 200 °C (TEM)**

The structure characteristics were observed not to changed substantially when ECAP was performed at temperature of 200 °C. However, substructure characteristics depend on the local position. In some elongated ferrite grains dislocations activities can be related to progress in polygonization and preliminary nucleation of newly born subgrain, Figure 7. The fringe contrast along grain boundaries of elongated subgrains and small grain nuclei are strong evidence of continuous recovery, probably dynamic recovery, in time of ECAP pressing and/or onset of dynamic recrystallization. Difraction pattern from a selected area of 1 μm indicates notable change in the angular spead of the spots (streaks).

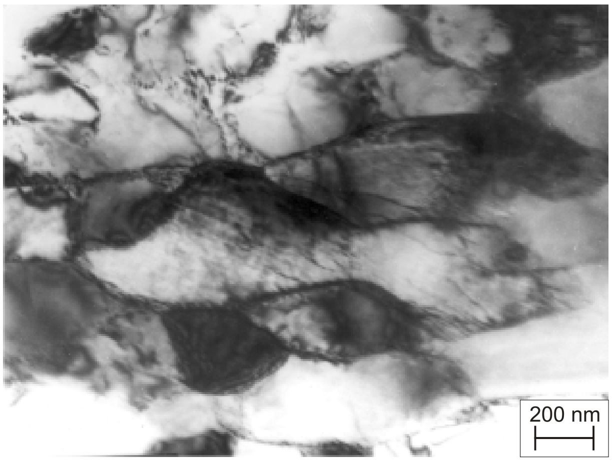

Figure 8. **TEM micrograph of new subgrains experienced dynamic recovery**

Slika 8. **Mikrostruktura novostvorenih subzrna pri dinamičkoj rekristalizaciji (TEM)**

As temperature of warm ECAP increases (T_{ECAP} = 250 °C) the tendency for development of submicrocrystalline structure became stronger, which can be attributed to in situ

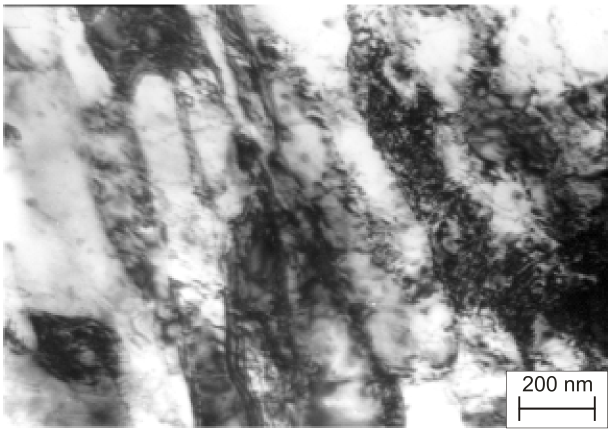

Figure 9. **TEM micrograph of newly born submicrocrystalline structure**

Slika 9. **Mikrostruktura novo stvorene submikrokristalne strukture (TEM folije)** dynamic polygonization and recrystallization. As a result of this process new subgrains generate in clusters and the discernible dislocations inside of subgrains are forming the dislocation networking, Figure 8. These subgrains can act as nuclei at formation of submicrocrystaline structure. The more advanced already equiaxed grains freer of dislocations can be seen in Figure 9. In this time the ECAP was performed at the temperature of 300 °C and the triple effect of working temperature, inserted strain and latent heat generated at severe deformation act as effective driving forces for dynamic recrystallization process, which supported in local areas the formation of polygonal recrystallized submicron grains, Figure 10. The presence of net pattern in SAED suggests the presence of a reasonable portion of boundaries having high angles of misorientation.

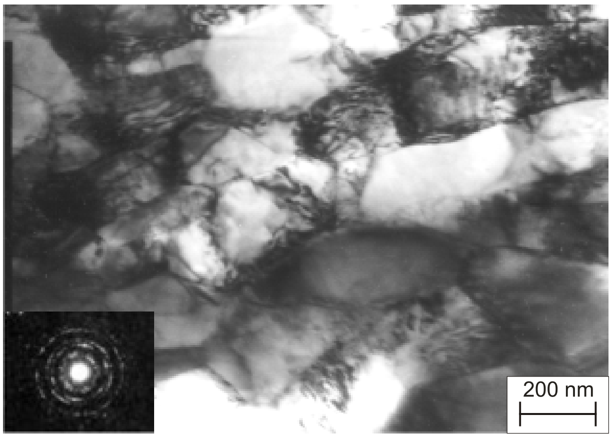

Figure 10. **TEM micrograph of well defined polygonal submicrocrystalline grains**
Slika 10. **Mikrostruktura izvijene submikrokristalnih poligonalnih zrna (TEM)**

Mechanical properties of steel after ECAP

In order to examine the effect of ECAP temperature the Vickers hardness HV1 was measured prior and after ECAP on the plane perpendicular to the pressing direction, (X area). A 1 kg load applied for 10 s was ensured for the measurement. The hardness values were taken as the average of a minimum of 3 measurements. The records are stated in Table 2. The hardness variation with increasing temperature of ECAP is seen, and a little unexpected is value at the highest temperature of ECAP 300 °C where effect of dynamic recrystallization on submicrocrystalline structure formation was observed to be the most advanced.

Table 2. **Microhardness (HV1) of initial and ECAP samples**
Tablica 2. **Mikrotvrdoća (HV1) početnih i kutno kanalno prešanih uzoraka**

$\mathrm{ECAP_{temp.}}$	annealed	150 °C	200 °C	250 °C	300 °C
$\mathrm{HV1_{edge}}$	87,0	239,5	233,6	235,7	242
$\mathrm{HV1_{center}}$	86,5	238	233	228	240

The results of tensile testing at room temperature using an initial crosshead rate of 5 mm/min for samples are shown in Figure 11., for fully annealed condition, and in Figure 12. for ECAP specimens. In case of fully annealed condition, there is an extensive period of strain hardening and a high elongation to failure. The deformation behaviour of ECAP specimens is very similar for all specimens where tensile strength is decreasing as ECAP temperature increases. However, a little different behaviour is observed concerning the specimen 3, which does not exhibit any work hardening following yielding. After reaching a maximum strength at a small strain a continuous drop in stress-strain curve is clear. The amount of uniform deformation is, therefore, very small and the stress-strain curve is similar to that anticipated in a work hardening. (It is noticeable that the reduction area value is, however, of similar one as for other samples, Figure 12.).

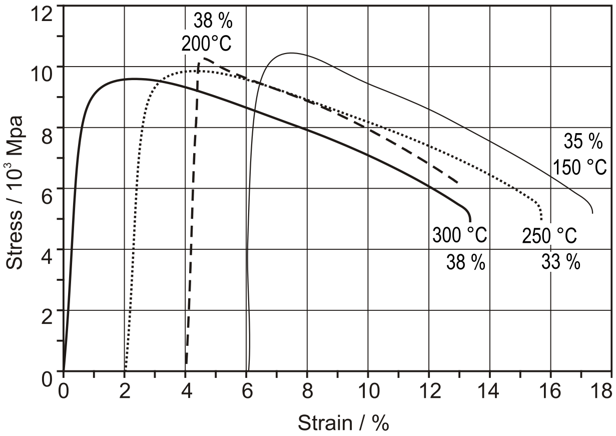

Figure 11. **Engineering stress - strain curves of annealed steel**
Slika 11. **Krivulje naprezanje - deformacija odžarenih uzoraka**

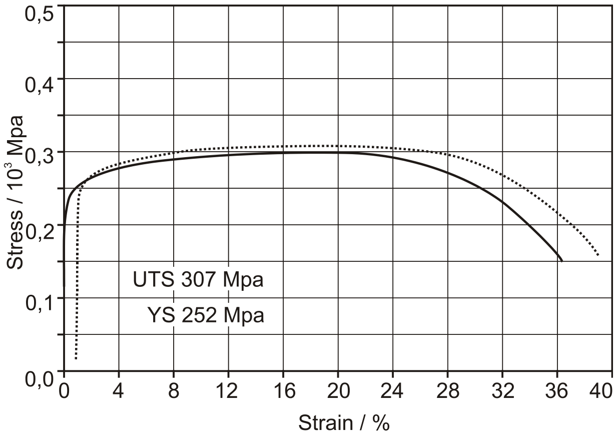

Figure 12. **Stress - strain curves of ECAP samples**
Slika 12. **Krivulje "naprezanje - deformacija" KKP uzoraka**

Generally, the obtained results confirm considerable increase of tensile strength compared to that of annealed steel. The yield stress is more than twice higher, reaching the maximum value of 680 MPa at T_{ECAP} = 250 °C. The region of strain hardening prior the softening is visible and the amount of uniform elongation is increasing with increasing temperature of ECAP. It can be attributed to the effective polygonization and to the dynamic recrystallization process resulting in forming of submicrocrystalline microstructure.

CONCLUSION

Microstructural evolution during warm ECAP pressing was studied in low carbon steel with ~ 0,1 wt % C steel. The major results can be summarized as follow:

1. The ECAP processing route B_c was performed at four different increased temperatures and billet experienced three passes. The intense and still nonuniform deformation as obtained throughout the billets, excluding end regions, as observed by light microscopy.
2. Formation of heavily deform substructure was apparent in sample investigated by TEM analysis. In elongated ferrite grains the substructure consisting of dislocation cells and subgrains was found.
3. The ECAP conducted at increased temperature was apparent to support and accelerate the process of polygonization of deformed structure, which process was observed already effective at structure recovery at the ECAP lowest temperature of 150 °C.
4. At the highest ECAP temperature of 300 °C the process of dynamic recrystallization effectively transformed the elongated structure of ferrite and contributed to formation of the stable submicrocrystaline structure, which influenced mechanical behaviour of steel.

REFERENCES

[1] H. Gleiter, Deformation of Polycrystals: Mechanisms and Microstructures, ed. N. Hansen et al. (Roslilde, Denmark: Risø national Laboratory, 1981), p.15.
[2] Y. T. Zhu, X. Z. Liao, Nature Materials 3 (2004), 351.
[3] R. Z. Valiev, Nature Materials 3 (2004), 511.
[4] Y. T. Zhu, T. G. Langdon, JOM 56 (2004) 10, 58.
[5] V. M. Segal, V. I. Reznikov, A. E. Drobyshevskii, V. I. Kopylov, IAN SSSR, Met. 1 (1981), 115.
[6] V. M. Segal, Mater. Sci. Eng. A 197 (1995), 157.
[7] R. Z. Valiev, R. K. Islamgaliev, I. V. Alexandrov, Progr. Mater. Sci. 45 (2000), 103.
[8] Y. Iwanishi, Z. Horita, M. Nemoto, T. Langdon, Acta Mater. 46 (1998), 3317.
[9] P. D. Hodgson, M. R. Hickson, R. K. Gibbs, Scripta Mater. 40 (1999), 1179.
[10] R. Kaspar, J. S. Distl, O. Pawelski, Steel Research 59 (1988), 421.
[11] Y. Matsumura, H. Yada, ISIJ Int. 27 (1987), 492.
[12] A. Najafi-Zadeh, J. J. Jonas, S. Yue, Metall. Trans A 23 (1992), 2607.
[13] S. Aihara, Procs. of First Int. Conf. on Advanced Structural Steels (ICAAS 2002), Tsukuba, Japan, May 2002, 7.
[14] R. Z. Valiev, A. V. Korznikov, R. R. Mulyukov, Fiz. Met. Metalloved. 4 (1992), 70.
[15] R. Z. Valiev, R. K. Islamgaliev, I. V. Alexandov, Progr. Materials Science 45 (2000), 103.
[16] Y. Saito, H. Utsunomiya, N. Tsuji, T. Sakai, Acta Mater. 47 (1999), 579.
[17] N. Tsuji, R. Ueji, Y. Minamino, Scripta Mater. 47 (2002), 69.
[18] D. H. Shin, J. J. Park, Y. S. Kim, K. T. Park, Mater. Sci. Eng. A 375 (2002), 178.
[19] Y. Wang, M. Chen, F. Zhou, E. Ma, Letter. Natur. 419 (2002), 912.
[20] R. Z. Valiev, I. V. Alexandrov, Y. T. Zhu, T. C. Lowe, J. Mater. Res. 17 (2000), 5.
[21] Y. M. Wang, E. Ma, Acta Mater. 52 (2000), 1699.
[22] S. V. Dobatkin, P. D. Odesskii, R. Pippan, G. I. Raab, N. A. Krasilnikov, A. M. Arsenkin, Russian Metallurgy (Metally) 1 (2004), 94.
[23] S. V. Dobatkin, Severe Plastic Deformation of Steels: “Structure, Properties and Techniques”, in Investigations and Applications of Severe Plastic Deformation, Ed. by T.C. Love and R. Z. Valiev (Kluwer, Netherlands, 2000), Vol. 3/80, 13 - 22.
[24] K.Oh-ishi, Z. Horita, M. Furukawa, M. Nemotao, T. G. Langdon, Mater. Sci. Eng. A 29 (1998), 2011.
[25] Y. Fukuda, K. Oh-ishi, Z. Horita, T. G. Lagdon, Acta Materialia 50 (2002), 1359.

Acknowledgement

This work was supported by the Ministry of Education, Youth and Sports of the Czech Republic in frame of Research Proposal the contract No. MSM 2631691901.