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Mechanism for Single Crystal Refinement in High Purity Niobium During Equal-Channel Angular Pressing

Lembit KOMMEL*, **Natalya LAEV**

Department of Materials Engineering, Tallinn University of Technology, Ehitajate tee 5, 19086 Tallinn, Estonia

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High-purity (99.99 wt.%) niobium (Nb) single crystal (SC) specimens as test material were received by electron-beam melting (EBM) technology and used as start material for equal-channel angular pressing (ECAP) of severe plastic deformation (SPD). First pressing was conducted at room temperature. The single crystal refinement via banded texture forming in the shear region of ECAP die during first pass and after fourth passes was studied. The dislocation ribbons and band morphology revealed in single crystal on intersection plane at first pressing. Primary grain boundaries initiation starts simultaneously. After four passes of ECAP the microstructure of intersecting bands in metal was formed and it was verified, that formed grain measures are 2 micrometers in mean.

The supplement microstructure and properties improving of Nb was conducted under hard cyclic viscoplastic (HCV) deformation. Results show that hardening rate during very first cycles was as maximal and decreased at next cycles. The viscoplastic behavior of Nb depends on tension-compression amplitude and microstructure, which was formed previously during ECA pressing. During HCV deformation Nb show: a) fully elastic behavior up to ±0.2 %, b) hardening – at compression and softening – at tension from ±0.5 % up to ±1 %, c) stable viscoplastic behavior at strain amplitude from ±1 % up to ±2 % and d) softening occurs at strain over ±2 % of cyclic straining amplitude. As a result of HCV deformation the UFG microstructure with low angle grain boundaries was preformed to high angle grain boundaries with lowering of dislocation density and metal has improved toughness and uniform elongation under tension loading.

Keywords: niobium, single crystal, severe plastic deformation, softening-hardening.

1. INTRODUCTION

Severe plastic deformation (SPD) techniques used in large amount of scientific investigation in the field of modern materials science for the production of bulk ultrafine-grained (UFG) materials [1]. These investigations include experiments using polycrystalline metals as start material [2]. Usually, before equal-channel angular pressing (ECAP) these metals can be heat treated for obtaining of soft and plastic material for processing [3, 4]. It is well known that during ECAP under shear stresses the deformation process affected by grain boundaries and by any interactions between adjacent grains [5, 6]. Usually, for the test results theoretical analyses the shearing model [7, 8] can be used. In practice, it is difficult to use these methods to obtain precise and detailed information about the deformation process in single crystal or in specimen, which contain two or three crystals only. Well known, that electron-beam melted (EBM) high purity niobium (Nb) usually has crystals in centimeter dimensions [9]. These crystals are oriented in direction of cast solidification and have length up to 250 mm and width up to 30 mm – 40 mm. Furthermore, it is impossible to select any initial crystals orientation and boundaries between crystals in specimens which were cut off from cast ingot. These EBM cast ingots have diameter of 180 mm – 220 mm. By this under ECAP usually can be used the specimens with diameter of 10 mm – 30 mm. Up to the present the single crystal (SC) have been used in few investigations of ECAP conducted with copper (Cu) [10, 11], aluminum (Al) [12 – 14] and niobium (Nb) [15, 16]. In which connection up to present there is a little information currently available on the mechanism of SC refinement depending on initial crystallographic orientation during the first pass [10 – 16] of ECAP. By this, for electronic, telecommunication, semiconductors and aerospace applications are needed not only the high purity but Nb with ultrafine-grained microstructure [17]. In connection with this in the present study, the main features of the pure Nb SC refinement via banded texture and UFG microstructure forming in shear region of ECAP die during first- and after fourth passes as well as under HCV deformation are presented and discussed in view of viscoplastic behavior depending on collected equivalent strain.

2. EXPERIMENTAL

In the present study, the main features of the experiments were conducted using billets which were manufactured from EMB cast ingot with diameter of 180 mm. The specimens were cut off from cast ingot in direction of solidification. A high-purity multiple EBM cast ingot was obtained from AS Silmet metallurgical plant. According to Quality Certificate this high purity grade niobium metal ingots has in chemical composition the following elements: N<30, O<72, H<10, C<20, Ta<160, Si<20, P<15, Mo<10 and W+Mo<20 in wt. ppm. In the cast ingot the columnar grains with grain boundaries are lying nearly parallel to the longitudinal direction. These grains are in average of 10 mm – 40 mm in wide and of 150 mm – 350 mm in long. Before the specimens machining the hardness was measured according to Standard ISO 6506-1 and ISO 6508 in cross-section and in longitudinal section in extent of

*Corresponding author. Tel.: + 372-620-3356; fax.: + 372-620-3196.
E-mail address: *lembit.kommel@ttu.ee* (L. Kommel)

1200 mm of cast ingot. The stationary hardness tester BRIN 200A and portative hardness tester ERNEST COMPUTES M3087 for this were used, correspondingly. The microindentation testing was conducted according to ISO 14577-1:2002 on Zwick Z2.5/TS1S universal testing machine.

From this cast the ingot was cut off specimens with dimensions of (16×16×135) mm and turned to diameter of 15.5 mm in accompanying to columnar crystal growth direction during EBM. The specimens were preliminary subjected to the heat treatment in vacuum furnace at temperature of 1090 °C for 2 hours. The heat treated cylindrical bars have decreased hardness and were covered with a copper thin film and lubricated with graphite-oil slush. These procedures were conducted to decrease of the flow stress under pressure, lowering of friction and to eliminate the friction welding under hydrostatic pressure between the sample and walls of intersected channels during ECA pressing. The samples form Nb SCs were oriented in one direction of entrance channel of the ECAP die. Between two channels of the die the angle was 90°. The specimens (SC and MC – multi crystals) were pressed partially through the ECAP die in a single pass at room temperature and the pressing was terminated when the samples were at the mid-point of the die. In addition, the HCV deformation [22] on materials testing installation INSTRON-8516 of ECA pressed FG Nb samples was conducted for additional microstructure evolution and viscoplastic properties study. For HCV deformation the tensile test specimens with reduced section of 8 mm in diameter and of 15 mm in length (from these before ECAP processed samples) were manufactured. Under HCV deformation the tension-compression cycling was conducted with frequency of 0.5 Hz at strain amplitudes in interval from ±0.05 % to 2 % for 20 cycles during one series and at six different strain amplitudes, but constant inside of each test series. The shear bands and followed UFG microstructure forming during SPD was studied in shear region by optical microscope (OM) and field emission scanning electron microscope (FE SEM).

3. RESULTS AND DISCUSSIONS

The micrograph of single crystal (SC) refining starts at shear region and first pass of ECAP (Fig. 1). Up to intersection plane (IP) [16 – 18] or up to shear plane [7] of the shear region in pressure direction (PD) the test Nb has SC structure. For theoretical investigations of Han and Tóth the IP have angle of 45° as the channels in ECAP die have identical cross-sections. In Fig. 2 this angle (near inside the corner of ECAP die) is shown as equal to 42°. Investigations show that in practice the IP have spherical surface. The dislocations ribbons (DR) take's forming on IP at first.

Afterward of IP the shear bands (SB) and grain boundaries (GB) of very large grains forming starts. The bands after shear region (under shear stresses) in extrusion direction (ED) the banded macrostructure can be formed. The axis and plains of ECAP is shown in plane witch is perpendicular to the transverse direction (TD) in Fig. 2.

The microstructure in shear region at first pass in transverse plain (TP) with dislocation banded macrostructure is shown in Fig. 3 in diametric (a) and on half radius (of specimen) sections (b). As is shown (Fig. 3, a) the GB in banded structure can be formed at first in diametric section. The bands were formed under shear stresses and have dislocation structure. The shear bands direction (SBD) is not identical with IP direction (see Fig. 2). The shear ribbons direction (RD) is oriented under angle of ~26° to ED. This result is accompanied with that's of Tóth [18] and Han [17] latest investigations. The shear region is shown in Fig. 2 at fourth passes by route B_c of ECAP. On the upper side (before IP) the intersecting shear bands are shown. As is shown in Fig. 2 the shear bands were reoriented under pressure stresses in transverse direction (TD) and they are perpendicular to PD. The optical microscopy picture (Fig. 3, a) shows that at first pass the

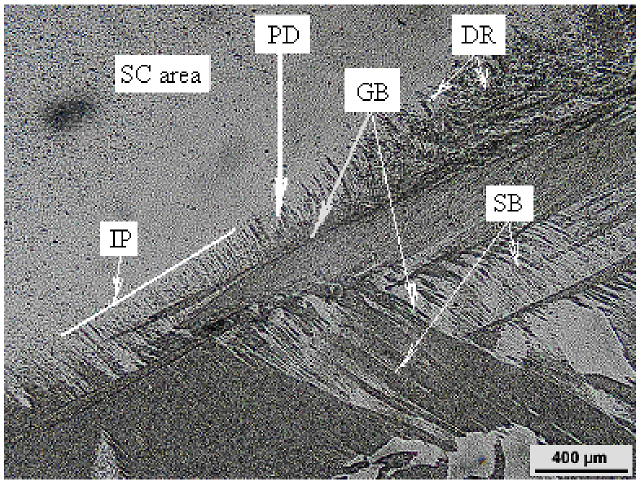

Fig. 1. Optical image of specimen (Nb) in a shear region of ECAP after first pass is shown. Designations: SC – single crystal area, PD – pressure direction, IP – intersection plane, GB – grain boundary, DR – dislocation ribbons, SB – shear bands

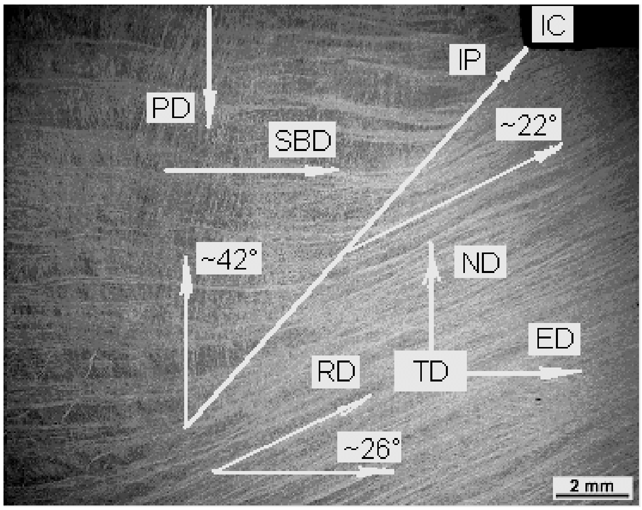

Fig. 2. Schematic drawing of ECAP in a shear region (near intrinsic corner (IC) of ECAP die) during next passes by B_c route is shown. The pressure direction are shown by arrows (PD), normal direction (ND), extrusion direction (ED), transverse direction (TD), shear bands direction (SBD) before intersection plane (IP) and ribbons direction (RD) after IP in SPD Nb. The angles between these directions are: 42° – between PD and IP, 22° – between IP and RD, 26° – between RD and ED

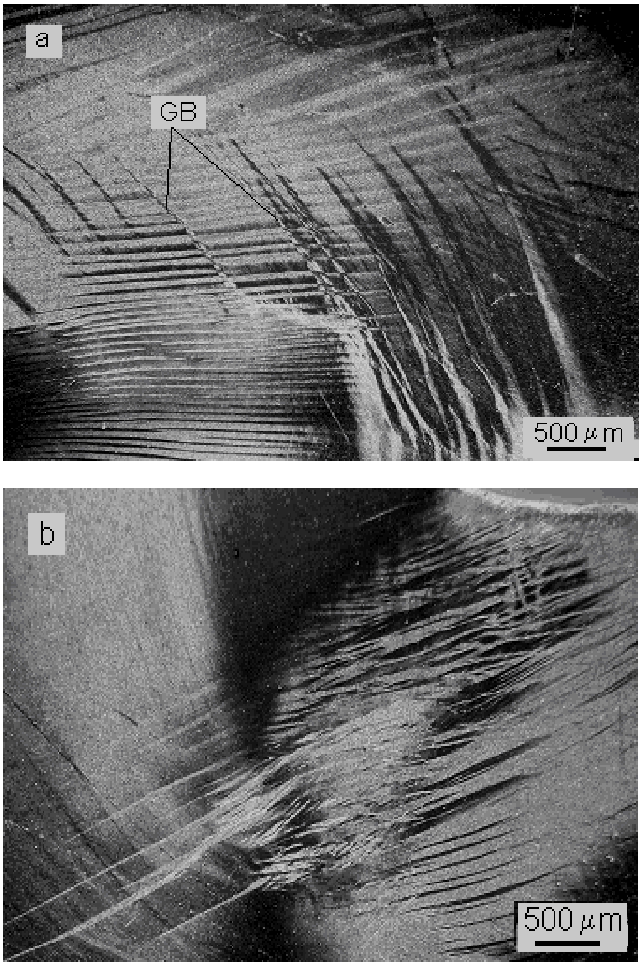

Fig. 3. Band morphology revealed in SC Nb by longitudinal sections (TD plane) in shear region of diametric (a) and 0.5 radius (b) sections at ECAP. The GB forming initiation is shown in (a) by arrows

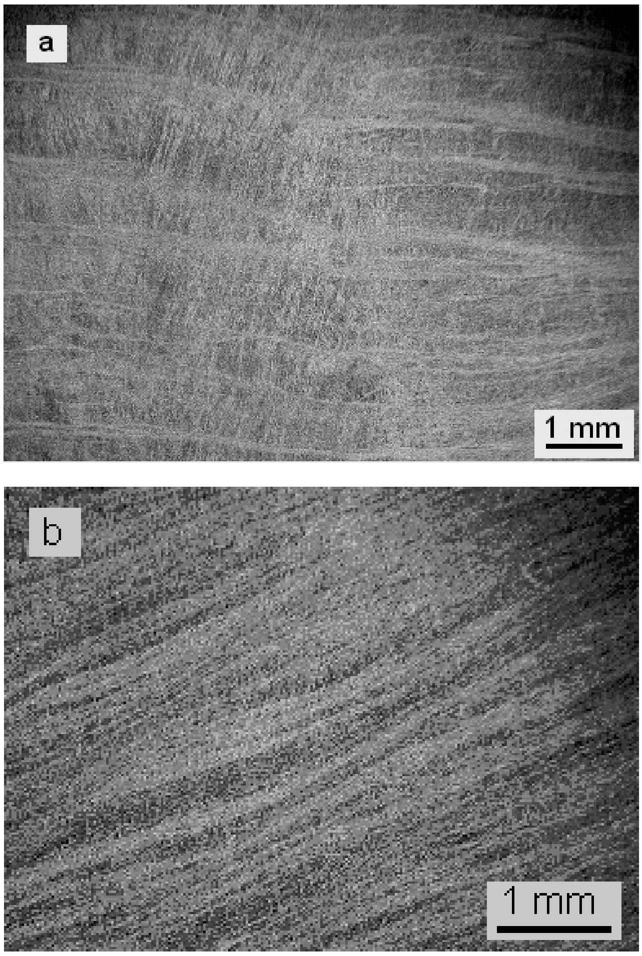

Fig. 4. The optical images of shear band's structure after three B_c passes of ECAP nearby shear zone (a) and in pressed Nb sample (b)

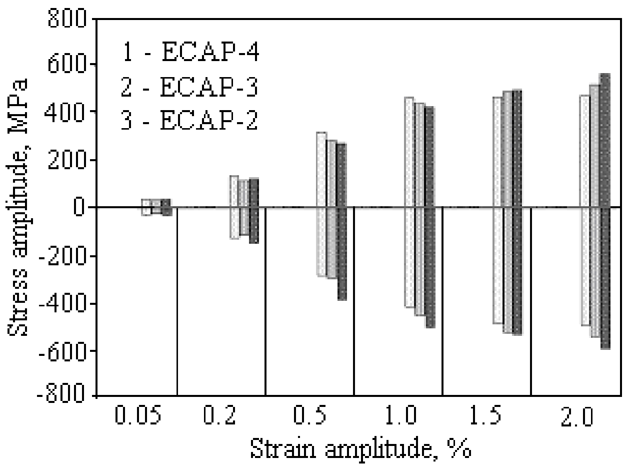

Fig. 5. The strain- and stress amplitudes diagram of samples for first cycle under HCV deformation. Designations: 1 – 4 passes of ECAP, 2 – 3 passes and 3 – 2 passes

grain boundaries (GB) forming was started. After three-four ECA pressings there are lightly warped intersecting bands in OM micrographs (Fig. 4, a).

The detailed view of Nb macrostructure (texture) can be conducted from Fig. 2. Large number of slip system were activated, resulting in the formation of deformation shear bands as observed in Fig. 4, a and b, in transfers plane (before and after IP) (see Fig. 2).

The HCV deformation, as one of SPD method was used for additional microstructure evolution study. By this method the viscoplastic behavior was studied also. These test results for samples with different collected strain at ECAP and strain amplitudes at HCV deformation is presented in Fig. 5. The HCV deformation test results for first cycle are shown. The three samples were tested under identical test regimes. These samples were preliminarily processed by ECAP at four, three and two B_c routes (1 – ECAP-4, 2 – ECAP-3, 3 – ECAP-2), respectively. For the strain amplitudes of ±0.05 %, ±0.2 %, ±0.5 % at 20 cycles these values were increased slightly for all samples with compare to first cycle values. It means that the low stress hardening was conducted at low strain amplitudes. At this stage of processing the metal shows fully elastic behavior.

Results show that up 1 % of true strain the sample 1 – ECAP-4 show at first cycle asymmetric stresses. The highest tensile and lowest compression stresses are found for ECAP-3 and ECAP-2 samples. At strain amplitudes of 1.5 % and 2 % to the contrary at first cycle highest but symmetrical stresses show sample ECAP-2. Under HCV deformation in Nb sample the double-banded microstructure was formed (Figs. 6, 7). It contains initial shear bands (ISB) formed during ECAP and secondary shear bands (SSB), which were formed during HCV deformation. We speculate that the dislocation density was increased too.

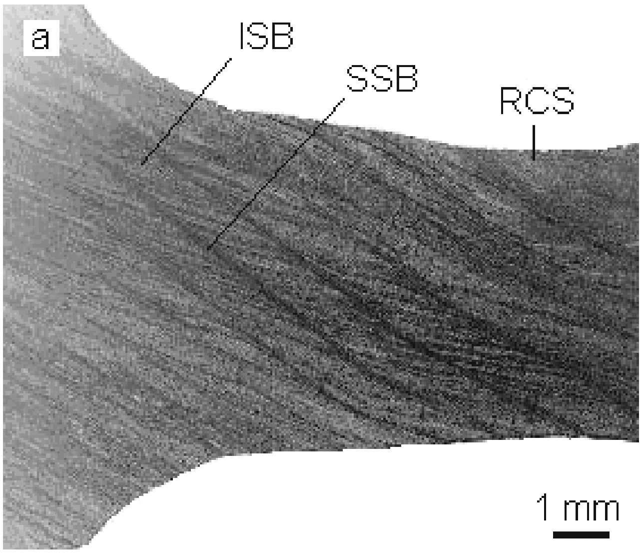

Fig. 6. The double-banded microstructure of pure Nb after SPD under ECAP and followed HCV deformation. Designations: ISB – initial shear bands, SSB – secondary shear bands, RCS – reduced cross section

Unfortunately, in this investigation for dislocation density study the TEM and XRD investigations were not conducted.

It is well known, that the dislocation density increase leads to increase of tensile strength and hardness but decrease in plastic properties of UFG metals, processed by SPD methods [1, 2].

Work-hardening behavior (or work hardening rate) [2] of niobium, tantalum, copper and aluminum processed by ECAP using the B_c route is maximal in shear plane during first's passes and increase up to fourth passes only. But comparing the first pass in this region and fourth passes the microhardness increases slowly and has uniform level of values, as the GS was decreased down to 2 μm in mean (Fig. 8).

The results show that the pure Nb with dislocation-banded microstructure has stable viscoplastic behavior during cyclic straining. The strain hardening/softening behavior was minimal. The HCV deformed microstructure (Fig. 7) is different as compared to ECAP processed (Fig. 4) microstructure. The HCV deformed bulk Nb has double-banded microstructure. The received results show that Nb with such double-banded microstructure show best elasticity behavior at hard cycling and at tensile straining. Usually during HCV deformation the reduced test part dimensions don't change. The tension-compression cycling starts at "0" and end at "0" elongation as was shown by extensometer. The test part diameter was reduced (see Fig. 6, RCS) at true strain of 20 % during subsequent tension for Young's modulus measure. The high purity Nb after HCV deformation has Young's modulus of 107.7 GPa.

The universal hardness as well micro hardness was increased significantly during ECAP but this increase was very small at HCV deformation. The test results show that the hardness change under HCV deformation depends on UFG metals microstructure state before cycling. The EBM cast ingot of pure Nb with length of 1200 mm has four maximal peaks with hardness of $103^{\pm 10}$ HB_{30} and four minimum peaks with hardness of $82^{\pm 7}$ HB_{30} with the steps between peaks of 300 mm – 350 mm. In cross-section the hardness distribution was identical. It means that the crystallites and boundaries have different hardness. It compare the ingots hardness, measured before processing the UFG Nb has very uniform hardness properties but different in different directions of samples.

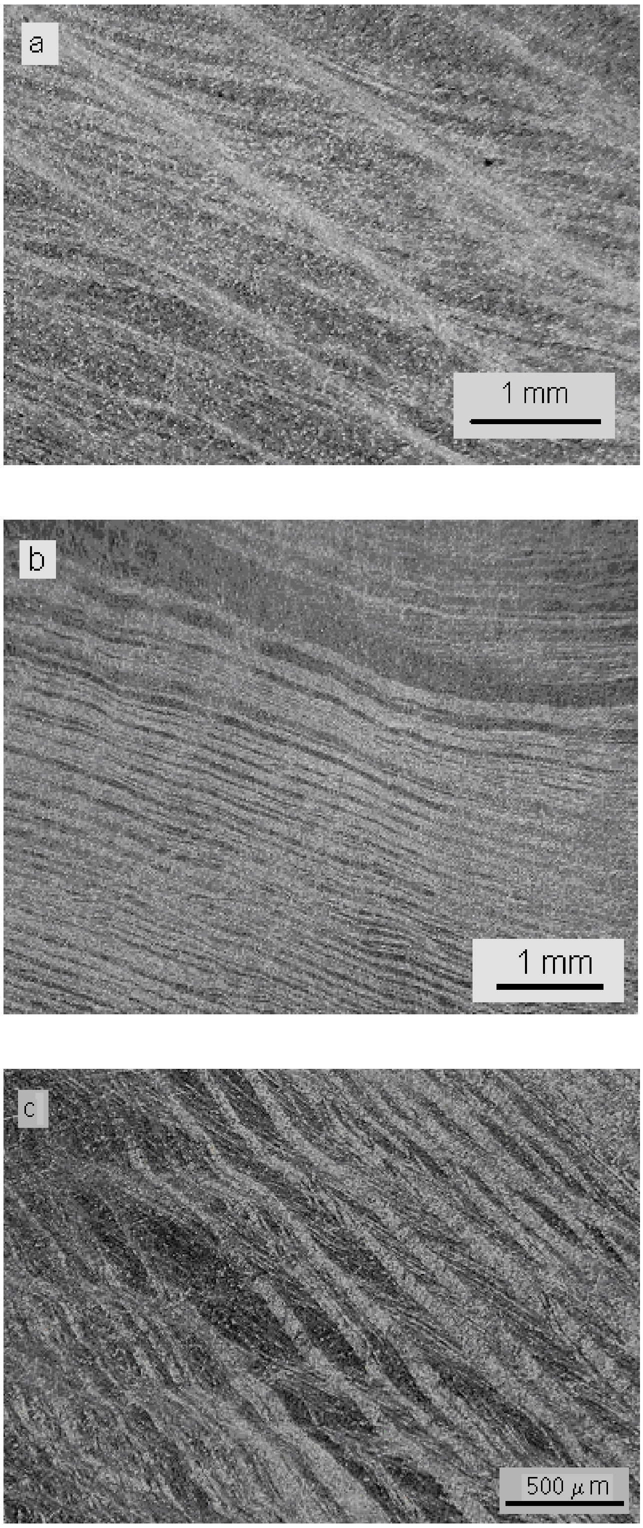

Fig. 7. The double-banded 3D-microstructures of pure Nb in longitudinal: a – ED in TP, b – ED in NP and in cross section: c – in EP (after shear region)

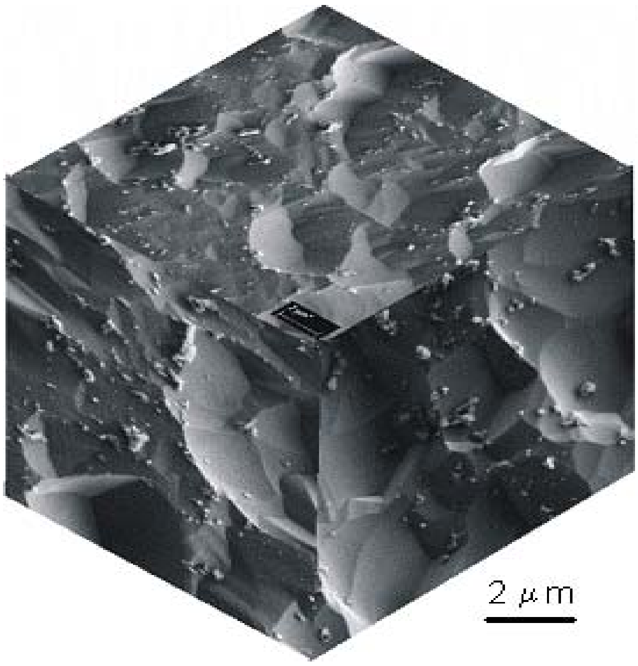

Fig. 8. The FE SEM 3D picture of pure Nb after four passes by B_c route of ECAP

4. CONCLUSIONS

During ECAP of the Nb SC the microstructure refining in intersection plane of shear region take place through dislocation ribbons, shear bands and grain boundaries forming.

At very first pass of ECAP the initial grain boundaries forming starts in single crystal.

After three-four passes in Nb the microstructure of intersecting bands was formed.

Under HCV deformation the double-banded microstructure was formed.

High purity Nb show low strain hardening rate during ECAP as well during HCV deformation.

The pure Nb show under HCV deformation stabile viscoplastic behavior and at tension high toughness.

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