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# EVOLUTION OF COPPER MICROSTRUCTURE SUBJECTED TO EQUAL CHANNEL ANGULAR PRESSING

**Carmela GURAU, Gheorghe GURAU**
"Dunarea de Jos" University of Galati
email: ggurau@ugal.ro

## ABSTRACT

*This paper aims to study bulk severe plastic deformation processes capable to produce ultrafine grain and also nanostructured 3D materials, interesting for processing in current industry. Samples of copper alloy were solution treated and then were subjected to repetitive Equal Channel Angular Pressing at room temperature in 1 to 8 passes, using route C. Severely deformed specimens were studied after each deformation pass. Their microstructural evolution and mechanical properties were investigated. Optical microscopy progression is evaluated on each separate ECAP pass. It is well known that although copper is a deficient resource it is used to the same extent as aluminum, a rich resource on the earth's crust. This research signs up in the category of new technologies for obtaining bulk metallic nanostructures that allow more judicious use of copper alloys by the substantial improvement of properties of use.*

KEYWORDS: SPD, ECAP, UFG, in bulk nanomaterials, coopper, microstructural evolution

## 1 Introduction

Ultrafine grain materials and much interesting materials with nanoscale dimensions nowadays are a challenge for researchers and also an issue up to date. Special attention is given now to study how to obtain massive nano materials. In bulk nanostructures have all three dimensions at the nanometric scale (3-D). Numerous methods have been developed for the manufacturing of nanomaterials. Severe plastic deformation is remarkable way for metallic massive material. In this approach it can be obtained crystalline grain size varies from less then 100 nm or up to 200 nm until 500 nm for ultra-fine grained in bulk metallic materials [1-3].

Since the 80s when Ruslan Valiev developed equal channel angular extrusion, this type of top-down methods, investigations using this method have exploded. Since then has tried producing massive material with three dimensions at the nanoscale by ECAP. Equal channel angular pressing or extrusion (ECAP or ECAE) have been defined that a process used to applied severe plastic deformations (SPD) to processed materials with the target of improving their mechanical properties by reducing the grain size. ECAP uses a die where there are cut two channels, equal in cross-section, intersecting at an inner angle denoted in literature with Φ, which is generally close to 90$^0$. The sample is pressed around a sharp corner. ECAP introduce large plastic strains in metals and their alloys to reduce grain size, using repetitive pressing. The final ECAP products have nanometric grains or ultrafine grains, but keep the cross section and shape like initial sample. Therefore the ECAE process allows the achievement of very refined grains as a consequence of the shear deformation that that takes place in the billet when crossing the intersection between channels. ECAP is based on increasing the free energy of polycrystalline alloys initial coarse grain, thru introduction high density of defects, especially dislocations [4].

The advanced materials science has got in central topics getting an optimal combination of properties of hardness and plasticity simultaneously. The interest increasingly higher in all used or potential severe plastic deformation methods of grain refinement is related to the fact that homogeneous pore-free polycrystals with ultrafine grain can be achieved currently. In this way ECAP is far one of the most promising SPD methods due to its potential for industrial exceeding the limits [5].

The properties of bulk nanostructured or UFG alloys differ very much from those polycrystalline with the same average chemical composition, massive with micrometer scale. In case of ECAP, the grain size of the final product can be varied by controlling: die designed angles (Φ and ψ the outer angle), pressure, number of successive passes, processing route. Two of the most important contribution are processing route and die angle because of slip process during ECAP. They are mostly reflected in the contributions of newly activated slip systems and reversed slip systems at the pass-to-pass processing, [6] different for each route applied.

In the present work we studied technical pure copper, which especially occur the grain size and influence of deformation, often deviates from simple shear, due to the existence of friction, strain hardening of material, or die design.

ECAP are feasible process for bulk nanostructure or ultrafine grain materials for copper alloys, whose performance increased spectacular by severe deformation. That allows more judicious use of copper alloys in the industries.

## 2. Experimental

This study was carried out on a technical pure copper, supplied as rolled billets 10 mm x10 mm and cut at 10mmx10mm x50mm. The billets were pressed in ECAP die with channel section 10mm x10 mm. The specific die angles, inner Φ $90^0$ and outer Y $13^0$ determine effective strain 1.07 in one pass [7].

Specimens were pressed via route C at room temperature. After first pressing the ECAP die was rotated and the specimen was pressed back in the vertical channel. Then specimen was pushes without extraction 8 passes, only with die rotates. The constant pressing was employed using a 20 tf press. Before pressing the samples were lubricated with a suspension of graphite in mineral oil to reduce friction. The speed of pushing plunger was 17.3 mm/s. Similar ECAP process was conducted to obtain specimens with 1, 2, 3, and 8 passes, equivalent strain: 1.07, 2.14, 3.21, respectively 8.56.

Following the ECAP procedure were obtained eight samples, with different equivalent strain. The samples were used for investigations after severe plastic deformation. The force variation in both processes, ECAP respectively direct extrusion was monitored with Hottinger Spider 8 system and force-stroke data was recorded.

Micro-Vickers hardness measurements were made on center and margin ECAP samples in longitudinal section using a digital instrument 400DAT2 NAMICON, fitted with of accuracy optical and electrical systems. $HV_{0.05}$ measurements were made using 50gf pressing force, pressing for 30 seconds, five determination for every sample on center and 2-3μm on margins.

The optical microscopy study has been carried out using a microscope Olimpus BX45 with digital image resolution at higher magnifications Metallographic samples were processed in longitudinal section for ECAP deformed samples and attacked to specific reagent, ferric chloride.

## 3 Result and discussion

### *3.1. Variation of force in ECAP process*

Variation of the extrusion force in equal channels angular pressing specifics looks in concordance with similar observation on copper ECAP severe deformation.

When the first layer of material leave the plan shear zone of die, force increases until it reaches a maximum. From the moment force tends to reduce until a value that maintain during deformation appearance a plateau, after which suffers slump in the finish zone of deformation. In figure 1 is presented force versus time of deformation plot for effective plastic strain 8.56 at eight pass.

Maximum force is between 93.646kN and 46.823kN. The maximum value was obtained on sample after first ECAP step. In this case of copper force trend is decreasing pass to pass.

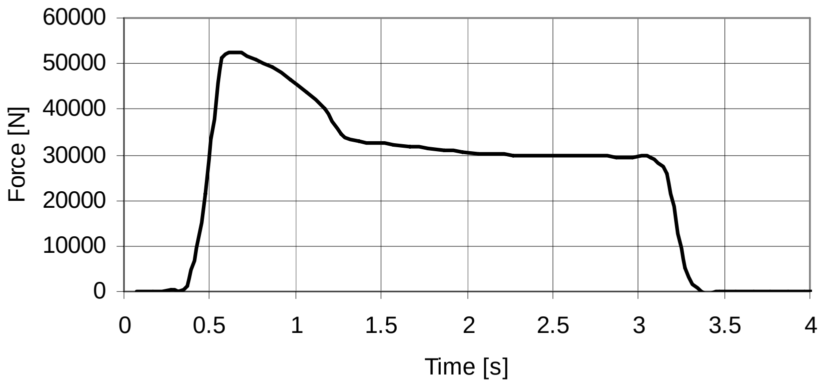


***Fig. 1.*** *Variation of force in the eighth pass of ECAP process - effective plastic strain 8.56*

Variation of force is consistent in the sense that after reach its peak force tends to form a plateau and after all sample cover decrease. In ECAP process force increased continuously until the maximum is achieved then take places a small decreasing variation. To the end of ECAP process force became almost constant. After all billet lives zone of deformation, force decreases dramatically to zero. Entire process takes place in about 25 until 30 seconds.

### *3.2. Microhardness*

Micro-Vickers hardness measurements values are presented in Figure 2. Were taken into account the values of severely plastically deformed ECAP samples with different degrees of deformation and measured values in center and at 2 3µm from margin. Note that all samples ECAP increase value of microhardness with each repeated deformation. Hardness value increased by 1.66 times from 78MPa for sample in initial state to 130MPa for sample after eight ECAP passes, in center billets. Same increase took place at the edge samples.

At low temperatures, the increasing of grains boundaries behave as slip barriers, their area being far increased than in micrometric grains bulk alloys. In case of nanomaterials are specific a high density of imperfections and dislocations with different settlement at grain boundaries.

That affects significantly the mechanical properties. Due to their reduced grain size, increase numbers of atoms on surface, nanomaterials are expected to become more ductile at the same temperature then coarse grained polycrystalline alloys with the same composition. More of that, ultrafine grained materials (100–500 nm) exhibit increased yield strength along with good ductility in comparison to nanograined materials. It has been correlated changes in strain hardening behavior with change of grain sizes [2, 3].

Severe plastic deformation leads to an advanced finishing structure. As a consequence hardness increases with increasing deformation. Note that the true deformation degree threshold value 8.56 above which plastic deformation by ECAP have still effect on mechanical properties.

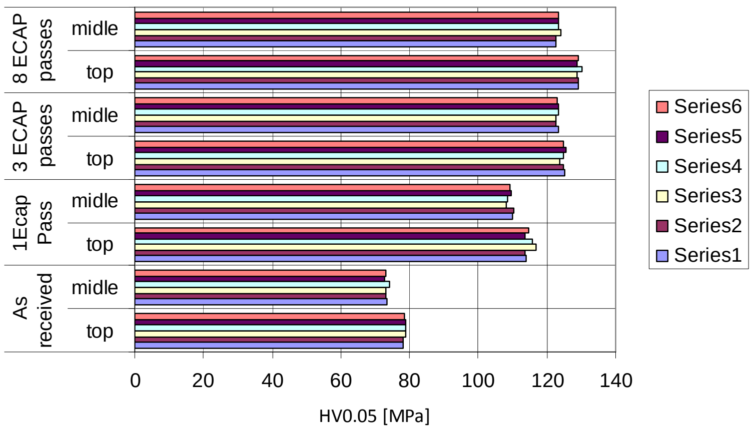


***Fig. 2.*** *Micro hardness versus number of ECAP passes (middle and top of X plane)*

### *3.3. Study microstructure of technically pure copper severely plastically deformed samples by ECAP*

In the investigation were studied by light microscopy copper specimens.

The specimens were observed in initial state and also after severe plastic deformation by ECAP process, with 1, 3 respective 8 passes by route C. Metallographic samples were processed in longitudinal section.

In the initial state in the sample are highlighted undistorted structure consists of a coarse-grained α solid solution and rarely fine intermetallic compounds precipitated [Fig.3].

The specimens in initial state were solubilized prior to severe plastic deformation.

After solubilization the microstructure of specimens present only equiaxed grains with dimensional inhomogeneity, polyhedral shaped, with straight edges and numerous twins homogenously distributed specific to fcc crystallographic cell. Grain size is about 35µm but is observed also finer grain as 10µm. Grain uniformity is quite obvious. On grains boundaries are evident preferential nucleation of fine precipitates black colored.

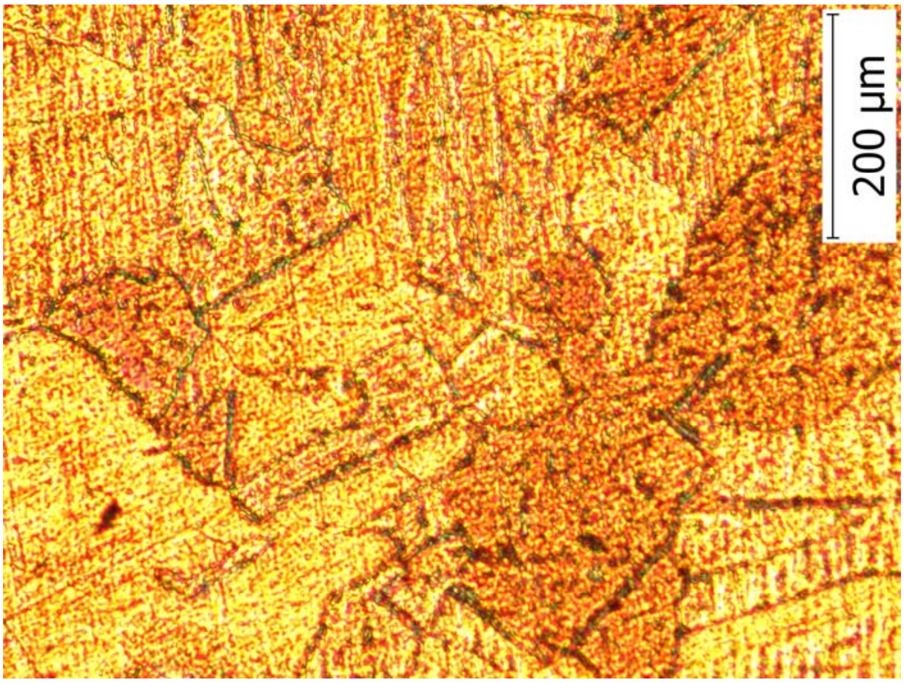


***Fig. 3.*** *Optical micrography of copper as received*

Microstructural aspect of copper after plastic deformation by ECAP method for each successive passages 1, 3, 8 of processing using route C are given in Figures 4-8.

On the outer surface of the sample which is in contact with the mold, especially in entrance area mold took place texturing in longitudinal section of solid solution α. The phenomenon of distorted appeared even the material is not yet in deformation. Crystalline grains are slightly deformed. Intermetallic compounds are observed such as dark particles. Texture is only because friction with the mold walls.

Prior to severe deformation, all rectangular samples were lubricated all over longitudinal contact with the mold.

One possible reason is the effect of friction texture appearance that can not be completely excluded. We strongly believe that at least at beginning of severe deformation process friction is an important factor to be considered.

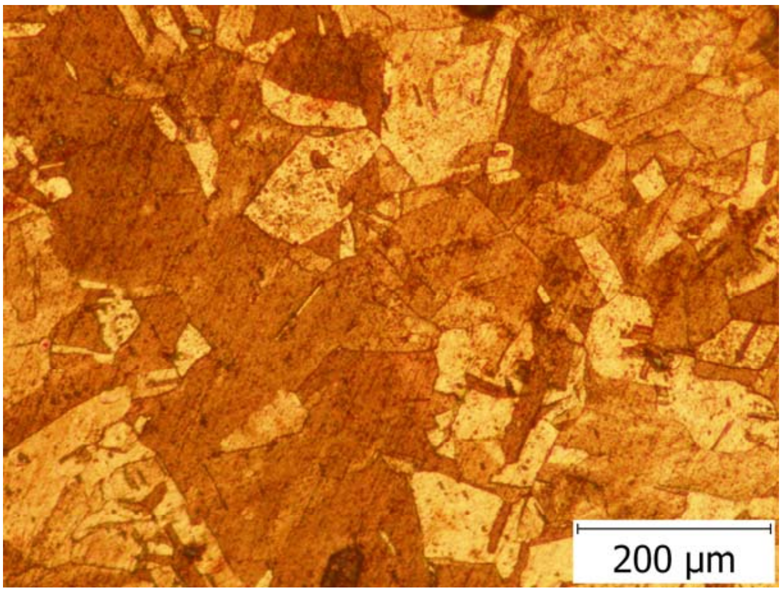


***Fig. 4.*** *Optical micrography Copper in entry area of inner channel*

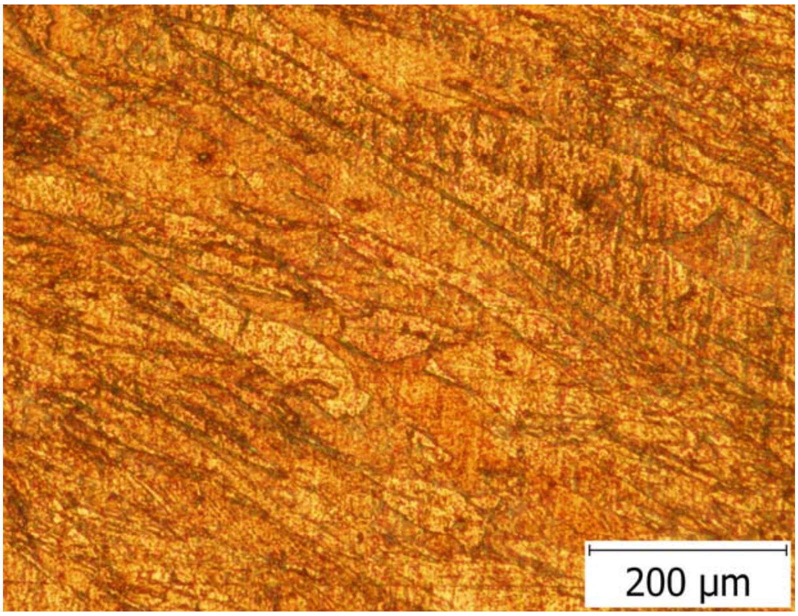


***Fig. 5.*** *Optical micrography Copper after one pass ECAP*

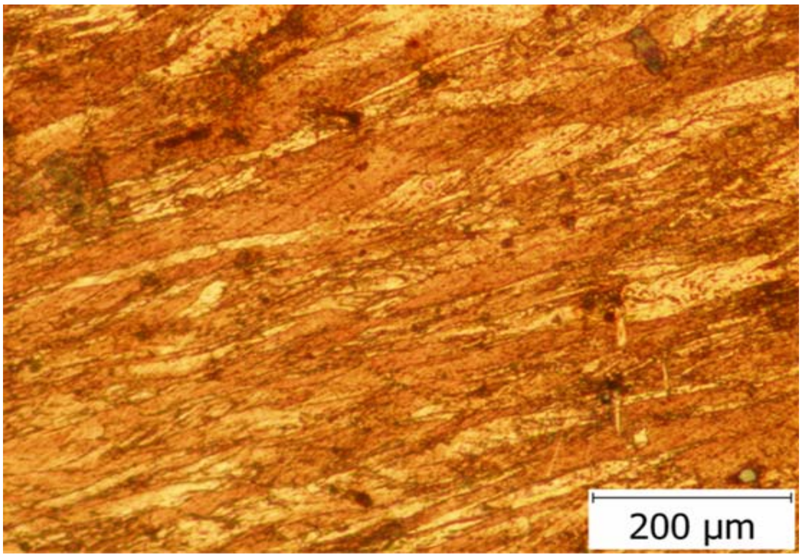


***Fig. 6.*** *Optical micrography Copper after three passes ECAP at the output of channel*

Copper micrographs after one pass ECAP disclose a significantly refinement of grains size and also modification of shape. The new structure is excessive elongated. The grains boundaries have lamellar aspect with parallel strings. The refinement and elongation is due to increasing density of dislocations. The ECAP process is based, like all severe plastic deformations, on high density of dislocations, Fig. 6.

The shape of grains is more and more elongated as the number of passes increased. The grains size is finer and finer. The aspect of grains is corrugated. Waving of the grains is a phenomenon that accompanies severe plastic deformation figures 6 and 7. In the microstructures of samples after eight passes ECAP it can be observed only flow lines. The grains boundaries no longer distinguished by OM it can be seen fiber of deformation, Figs. 8 a, b, c, d.

The plastic deformation took place almost monotonically in share plan. The share plan is at the intersection between inner channel and outer channel. The solid solution grains begin to refined and corrugated from firs pass of deformation. The deformation is produced by successive shear of grains that are already refined in previous passes. The structure of SPD samples present aligned bands more refiner which highlighted only flow lines in the eighth passes.

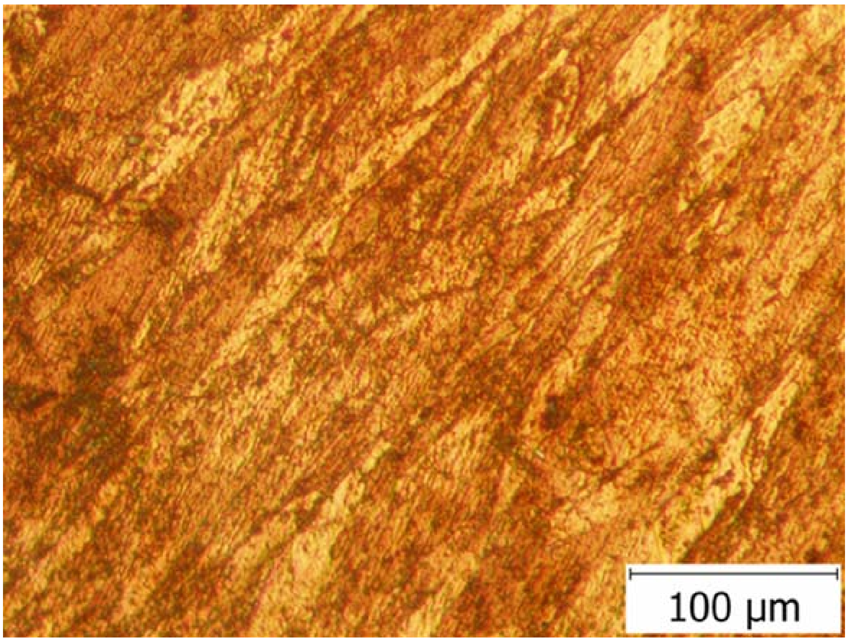


***Fig. 7.*** *Optical micrography Copper after three passes ECAP central area*

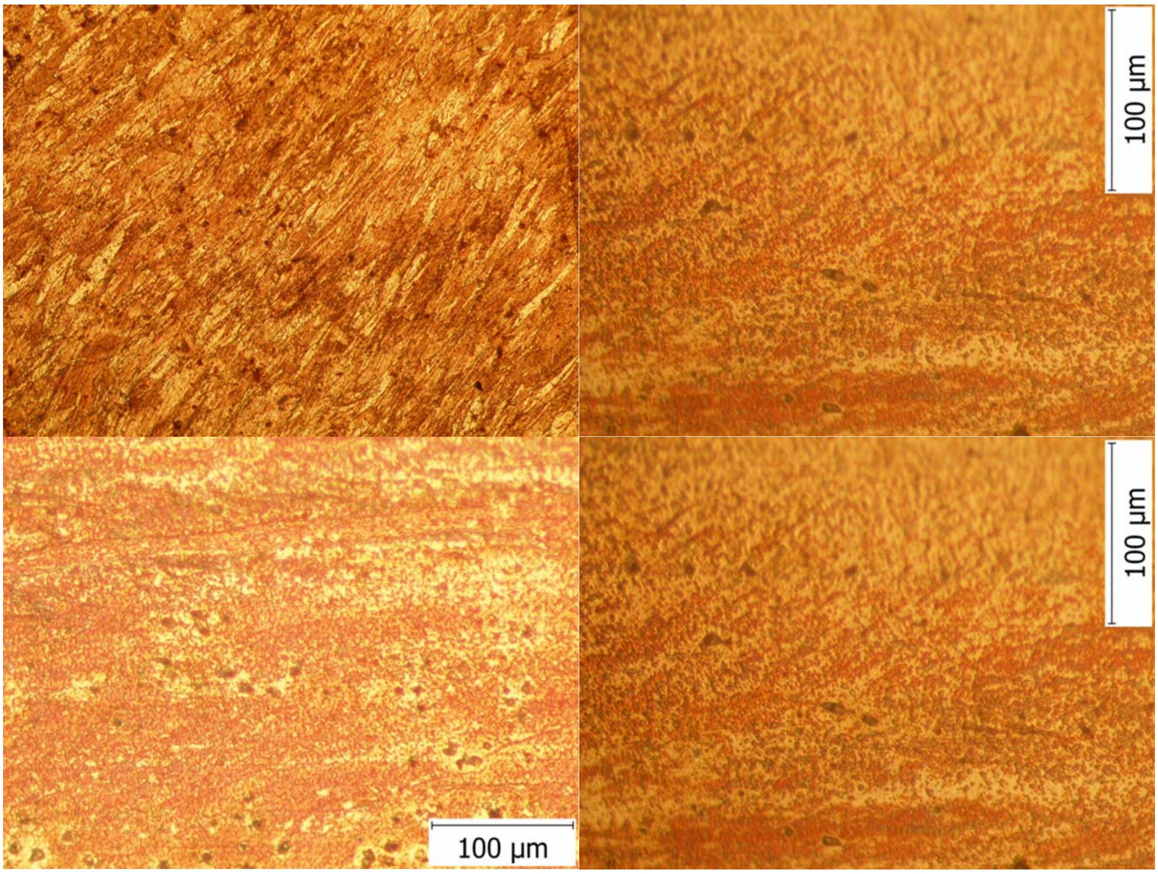


***Fig. 8.*** *OM Copper after eight passes ECAP. Longitudinal section of sample: a) left side, b) central area at superior edge, c) central area, d) right side*

As the degree of deformation increases in the microstructure of samples, it can be observed significant differences, regarding to the internal structure. In the first step microstructure are elongate, after each pass the structure became finer and afterwards ultrafine. The grains boundaries become increasingly less visible, only the fiber flow appears evident. The fiber structure is compressed in transversal direction and is rotated in extruded direction. Central area of samples from picture 9 c, d and e highlighted parallel fiber lines which make an angle of 65$^0$ to the vertical direction of extrusion. The aspect of microstucture is uniform and ultrafine. In OM can be seen only alginate bands to shear plan. Inside solid solution it can be observed the same refining of insoluble particles dark colored. Same authors consider that part of these particles can be dissolved in solid solution because of thermal effect in severe deformation by ECAP.

The microstructure aspect of samples after eight passes route C is one very dense with flow lines at 65$^0$ in all volume.

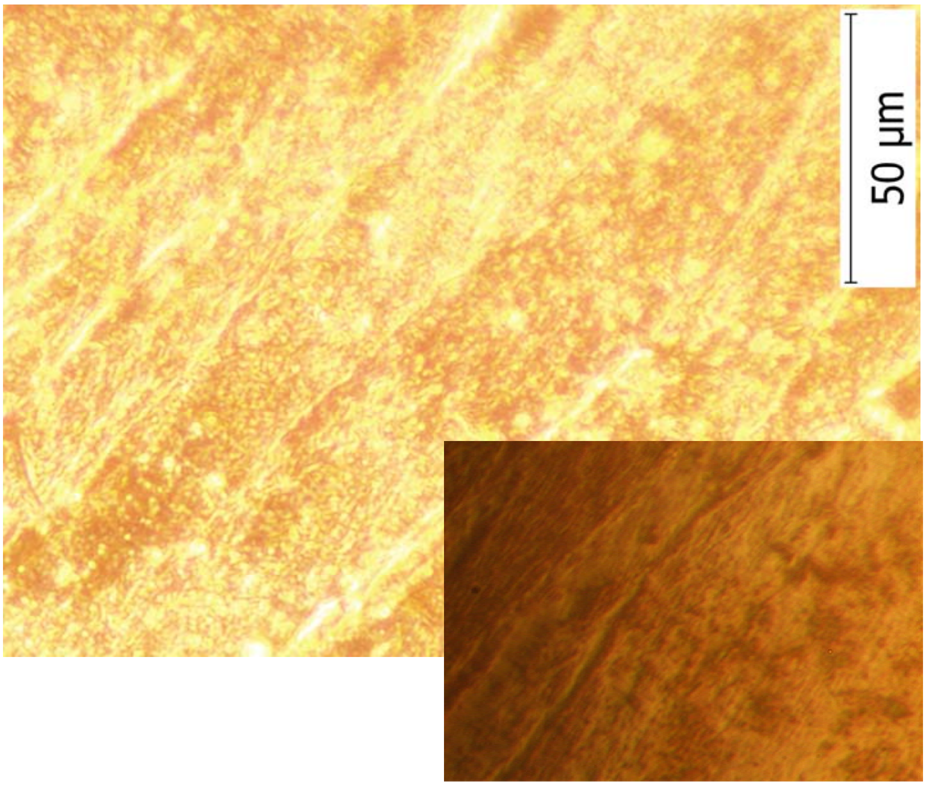


***Fig. 9.*** *OM Copper after eight passes ECAP in central area 1000X and detail 2000X*

The grain size finishing and also fibrous texture was determined by increasing deformation degree by ECAP process. Extruded samples after eight passes ECAP deformations by route C, have only fiber lines (Fig. 9).

## 4. Conclusions

Severe Plastic Deformation processes leads to ultrafine grained microstructure even nanostructure. These processes works at high pressure (GPa) and high deformation degrees.

Characteristic for SPD are an intense share process in transverse section of sample and unchanged shape of metal volume (ECAP particular case). The initial grains are divided in finest subgrains by slipping and rotation on various systems of sharing bands.

The equivalent strain is different for different materials and deformation conditions. Generally the equivalent strain is achieved in successive passes generally more than one.

For example in aluminum case are necessary 5 passes on route C as opposed to copper which need more than 8 passes. True strain starts from 1.07 after firs pass and goes to 5.35 after five passes.

That determines a change of grains shape.

The grains became elongated sometimes corrugated then are reduced gradually until gets simply flow lines. After eight passes for copper we can see only flow lines with fibrous aspect and dense surface.

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