

## **Characterization of nickel alloy 600 with ultra-fine structure processed by severe plastic deformation technique (SPD)**

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**Abstract.** High strength nickel based alloys are used in a multitude of advanced systems where lightweight, high power density mechanical power transmission systems are required. Components such as gears, bearings and shafts could be made significantly smaller and more durable if a major improvement in nickel based alloy mechanical properties could be achieved. A significant refinement in grain size (includes nanometric level) is thought to be a promising method for achieving fundamental improvements in mechanical properties. Grain size is known to have a significant effect on the mechanical behavior of materials. One of the most favorable methods of achieving extreme grain refinement is by subjecting the materials to severe plastic deformation (SPD). The principal microstructural variations in superalloys are the precipitation amount and morphology, grain size and the distribution of carbide precipitation that could reduce the mechanical properties of the alloys. This work shows optical and transmission electron microscopy analysis and also hardness data after severe plastic deformation (pure shear stress).

### **Introduction**

Despite the physical and mechanical properties of metals are determined by several factors, the average size of grain of material usually plays a very important role and is often a dominant factor in determining the properties and the final application of metals. It is noteworthy that the increase of mechanical resistance with the grain size reduction has aroused increasing interest in the manufacture of materials with extremely small grain size.

The grain sizes of commercial alloys are generally manufactured for specific applications, through predetermined thermo-mechanical attacks treatments in which the alloys are subjected to specific regimes of temperature and mechanical tests. However, these processes cannot be used to produce materials with sub-micrometric grain sizes because there is a fixed lower bound, on the order of a few microns, representing essentially the minimum grain size achieved using these procedures.

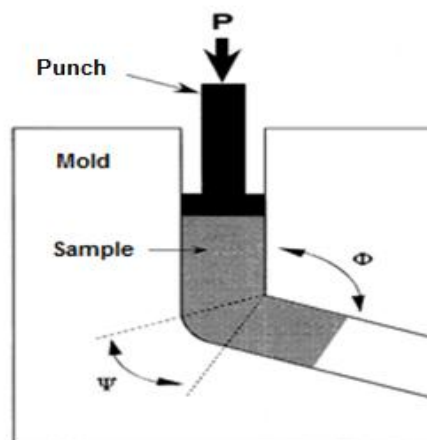
In practice, conventional methods of forming, such as lamination and extrusion, have restricted capacity to produce materials with grains ultrathin for two important reasons: first, there is a limitation on the total deformation can be imposed on the material due to the reduction in cross-sectional dimensions of samples; Second, the deformations imposed by conventional methods are insufficient to introduce ultra-fine structure due to low workability of alloys at room temperature and relatively low temperatures [1]. As a consequence of these limitations, the need arose to develop alternative processing techniques, based on the application of severe plastic deformation (SPD), where extremely high deformations are

imposed for manufacture of materials with grain size in the range submicrométrica or nanoscale [2].

The applicability of nickel alloy 600 is associated to the high melting point (1726K), adequate corrosion resistance, and ability to dissolve, so limited, number of metallic elements that serve to strengthen or improve its properties, corrosion and high temperatures [3-5]. The nickel-chromium-iron alloys have their applicability based on the presence of chromium and other alloying elements. Chromium is essential constituent for resistance to corrosion, being normally present in these alloys containing between 15 and 30%, the other elements are usually present on a smaller level, ranging from 3 to 20%, and are related to the increased resistance to deformation at high temperatures.

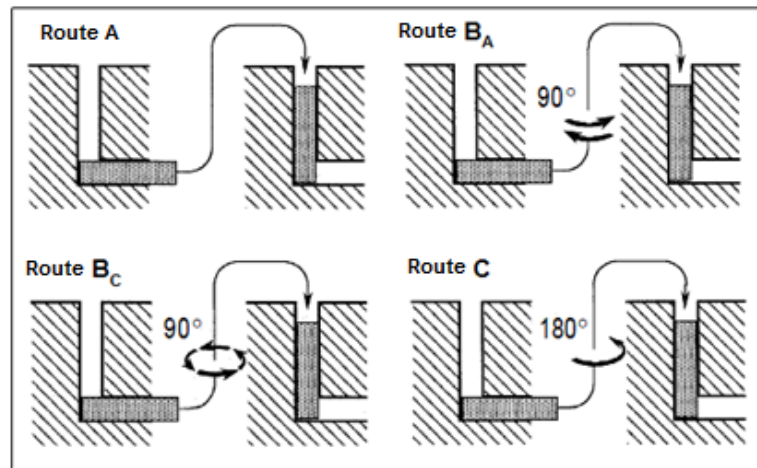
Nickel alloy 600 is an alloy austenitic of solid solution containing 76% nickel, 15% chromium, 8% iron and minor additions from other elements. Containing such high nickel content, this alloy has low solubility of carbon, so that while being exposed to temperatures between 813K and 1253K, the excessive carbon if precipitate as chromium carbides. Depending on the temperature of exposure, these carbides can appear both scattered in the array and contours of grain, as predominantly on the contours of grain. The League features 600 austenitic array, solid solution, face-centered cubic phase (and several secondary phases (carbides and metal precipitates) [5].

Severe plastic deformation (SPD) is a simple and feasible method to achieve grain refinement of up to 100nm in metallic materials. However, SPD of any high-strength material, such as Inconel 600 equiangular channels by pressing or twisting in high pressure is inherently difficult due to tooling requirements [6]. Among these methods, pressing in SPD, also called Angular channel extrusion (ACE), is one of the simplest methods of deformation, which under ideal conditions occurs by simple shear. This technique was originally developed by Segal et al., aiming to introduce severe plastic deformation in materials without changing the cross-section of sample, and it is possible, therefore, repeat the process several times to increase the level of deformation imposed [7-9]. The extrusion principle for SPD is illustrated schematically in Figure 1. The mold (die) is constructed with two sequential channels, which intersect at an angle  $\Phi$  and there is also an additional angle,  $\psi$ , which defines the arc of curvature of the junction. The sample test is inserted within the mold channel and pressed with a punch in a load  $P$  [9].



**Figure 1** – Schematic diagram showing the extrusion principle for SPD: two sequential channels, which intersect at an angle  $\Phi$  and also an additional angle,  $\psi$ , which defines the arc of curvature of the junction.

The process recurrence allows developing different microstructures by rotating the sample between consecutive passes. In Figure 2 shows the four basic processing routes (paths), where sample rotations between consecutive passes are  $0^\circ$ , to the route;  $90^\circ$ , to route B, and  $180^\circ$ , to route c. The route B is still divided into routes  $B_A$  and  $B_C$ , with  $90^\circ$  rotations in opposite directions along the same lines, or alternating after each pass, respectively. According to Langdon and collaborators  $B_C$  processing route is preferable in relation to the other to achieve a homogeneous microstructure [10].



**Figure 2** – Selections for billet rotation between consecutive passes through SPD die.

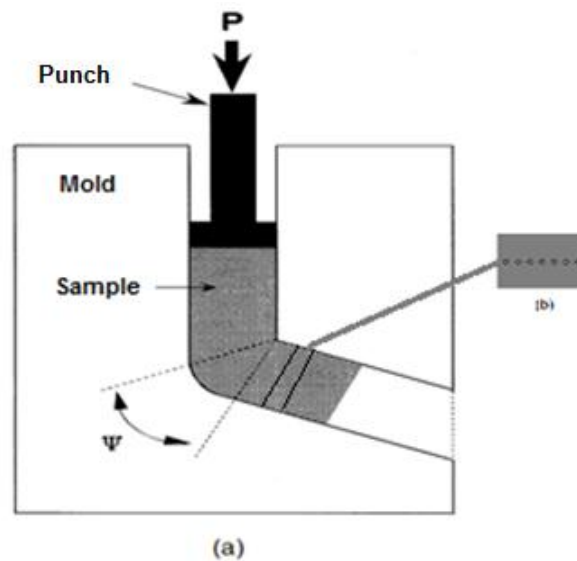
## Materials and methods

The applied material of this work is a nickel-based superalloy, nickel alloy 600, received in the form of bar with length of 1.6 m and diameter of  $7.10^{-3}$  m (Firm: Multialloy, Brazil). The chemical composition in percentage by weight is: 73,8% Ni; 16,3% Cr; 8,55% Fe; 0,33% Mn; 0,39% Si; 0,26% Ti; 0,17% Al; 0,02% Cu; 0,03% Co; 0,002% S; 0,007% P; 0,002% B; 0,007% C.

Before the SPD (severe plastic deformation) technique process the specimens were machined with dimensions close to the channel of the mold, and the material was subjected to a heat treatment at 673K during 1800s, followed by cold water quenching. For the carrying out of the trials used a bipartite mold (D2 tool steel) with two rectangular channels forming angle ( $\Phi$ ) of  $120^\circ$  to each other and with external elbow angle ( $\psi$ ) of  $60^\circ$ . The punch used in pressing operation was also manufactured in D2 tool steel to maintain a small clearance between the same and the channel. After the extrusion process, the pinhole is removed and the deformed billet is also removed from the second channel. The process can be repeatedly performed in the same tool with various alternatives and thus the intensity of total deformation will be N (number of passes) times a single shear deformation (figures 1 and 3).

The SPD technique was conducted at room temperature, positioning the array in a universal testing machine INSTRON 4400R operating at a constant speed of 5 mm/min in order to reduce the effects of friction and grip and prevent problems during processing, the channel and the specimens were lubricated with copper grease. In the course of processing a few interruptions were necessary for lubrication. Several routes were carried out via the SPD technique, in this paper is presented according to the results  $B_C$  route, i.e., each pass the

sample is rotated by 90° in relation to the previous pass. Were held up to six passes for each sample.



**Figure 3** - Schematic drawing of the: (a) Sample array during the SPD procedure ( $B_C$  path) in mold (die). (b) Cross section of the sample preferred where took place the hardness tests.

The specimens were exposed to total of 2, 4 and 6 passes using the  $B_C$  path of deformation, in which the body of evidence is turned 90° in the same direction in relation to the longitudinal axis after each pass. After SPD processing, the specimens were cut in the plane perpendicular to the longitudinal axis. For the tests of Vickers microhardness was used the Buehler Micromet microdurometer 2103 and the measurements were performed with load of  $5.0 \times 10^{-2}$  kg on the surface of the sample (with optical microscope) with the indentations sufficiently spaced out, so as not to interfere with each other. The results consist of the average of the values obtained. All of these measurements were performed at SPD plane perpendicular to the longitudinal axis of the samples over the line passing through the center, as illustrated in Figure 3.

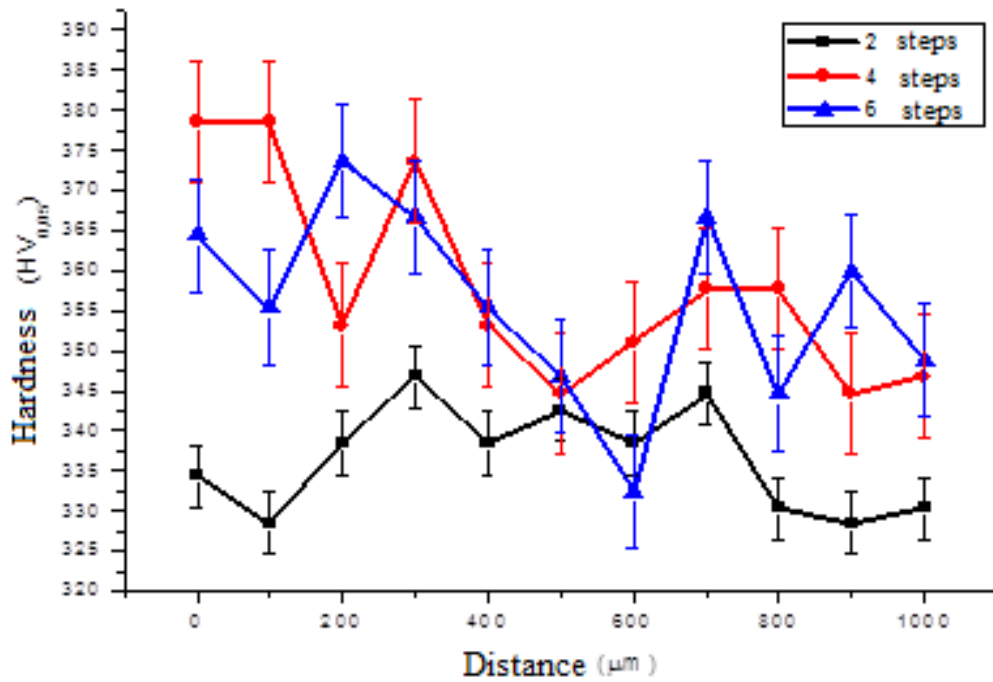
## Results and discussion

The mechanical properties values (Vickers hardness) indicated a significant increase of mechanical strength of nickel alloy 600 (figure 4). Vickers hardness tests carried out in the received material show mean value of  $(313 \pm 4) HV_{0.05}$ .

The OM observations in the samples of the as received nickel alloy 600 showed grain size variations and carbide and nitride precipitations (Table 1) identified by XRD (Figure 5) and EDS microanalysis (SEM). After heat treatment at 673K during 1800s it is observed grain size variation and presence of carbide precipitations (Figure 6). Through the presence of the deformation processes there is a decrease in grain size after an increase in the applied SPD number of passes. (Figure 7)

In some situations the particles present (carbides) have dimensions compared to the new SPD induced grain. With the obtained data from the mechanical tests and with the microstructure

during observations by OM and TEM indicate that nickel alloy 600 after SPD process exhibited higher strength while maintaining ductility.



**Figure 4** - Vickers hardness values in nickel alloy 600 (cross-section samples) after 2, 4 and 6 steps of intense deformation by SPD technique.

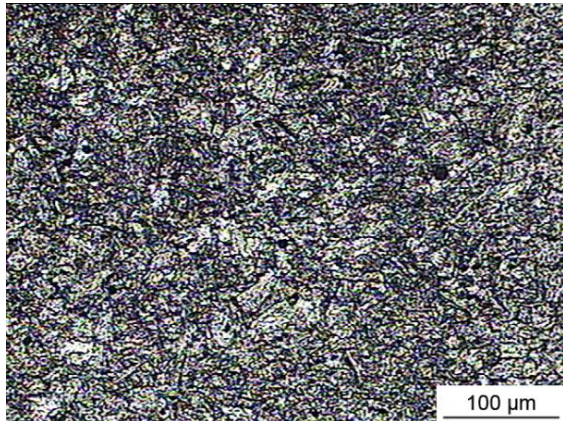
Table 1 – Elements compositions obtained by RX diffraction of as received nickel alloy 600

Compound Name	Scale Factor	Chemical Formula
Iron	0,214	Fe
Chromium	0,061	Cr
Nickel	0,216	Ni
Chromium Nickel	0,259	Cr <sub>2</sub> Ni <sub>3</sub>
Iron Carbide	0,636	Fe <sub>5</sub> C <sub>2</sub>
Hematite, syn	0,019	Fe <sub>2</sub> O <sub>3</sub>
Manganese Oxide	0,034	Mn O
Manganese Oxide	0,069	Mn <sub>3</sub> O <sub>4</sub>

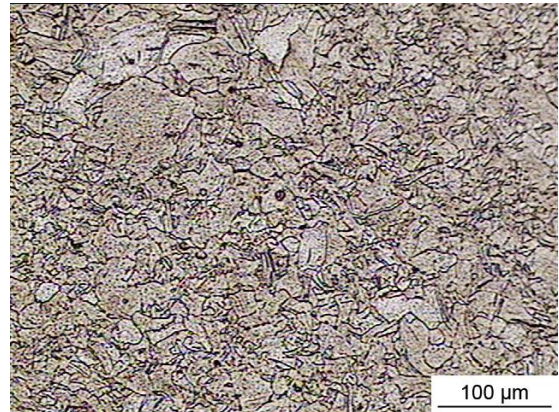
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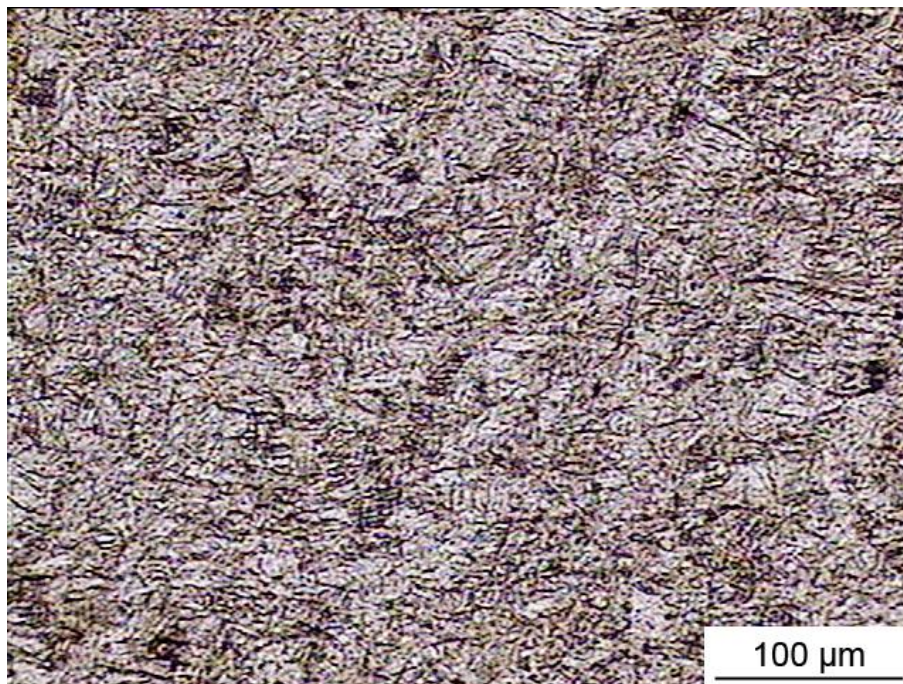
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**Figure 5** - Micrograph by optical microscopy of as received nickel alloy 600 (varying sizes of grains and presence of precipitation).



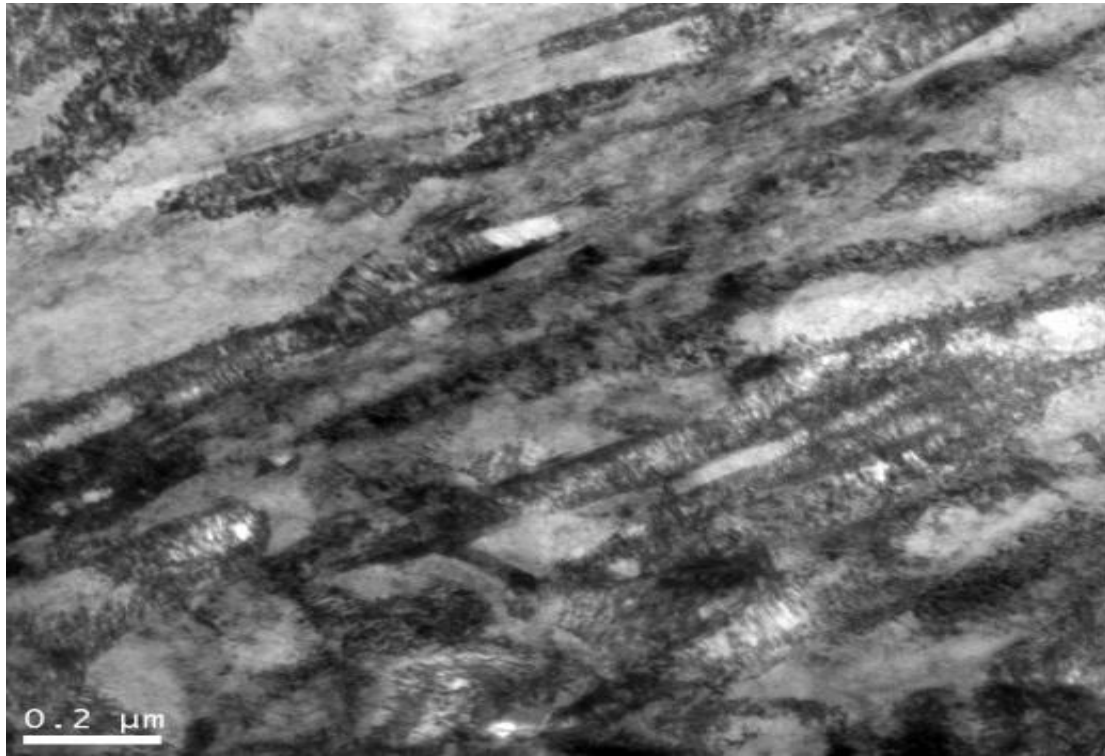
**Figure 6** - Micrograph by optical microscopy nickel alloy 600 after heat treatment at 673K after 1800s: grain size variation and carbide precipitations.



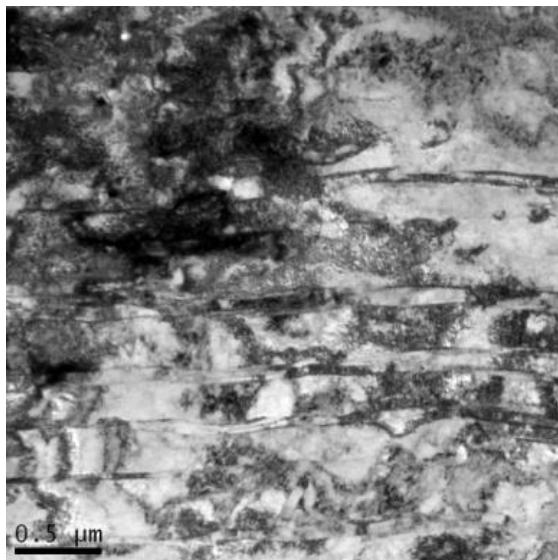
**Figure 7** – Optical micrograph of the nickel alloy 600 after heat treatment of 673K during 1800s also after four SPD passes, presence of elongated grains and precipitates identified by further EDS microanalysis - SEM, electron and x-rays diffraction results.

The observations carried out by MET revealed that after a few steps by PCE, the structure of nickel alloy 600 heat treated (673K during 1800s) presents a mixture of grain subboundary and low and high angle boundary grains. Individual linear crystalline defects (dislocations) were induced already in the early stages of extrusion and reordered to form dislocation tangles

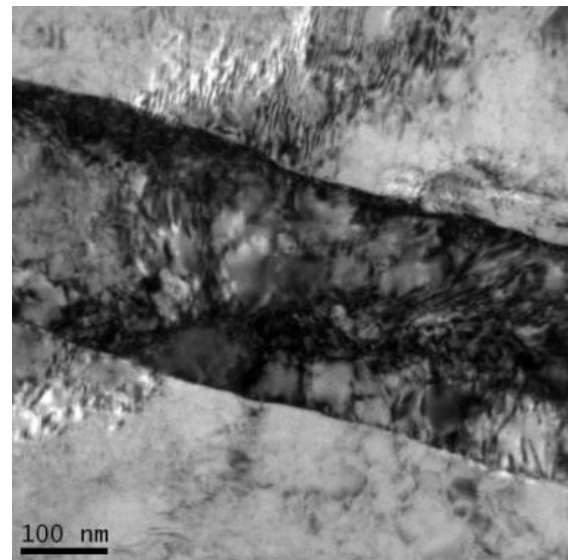
presented in new grain boundaries and grain subboundaries during the deformation process (Figures 8).



(a)



(b)



(c)

**Figure 8** - Electron micrographs (TEM) of nickel alloy 600 heat treated at 673K during 1800s followed by intense deformation –SPD process, route B<sub>C</sub>, two passes: (a), (b) General aspects of the microstructure, subgrains presence inside elongated grains in the shear process direction despite the variation in the shear process used; presence of dislocation tangles and fine precipitates (c) grains with preferred shear direction, also presence of dislocations.

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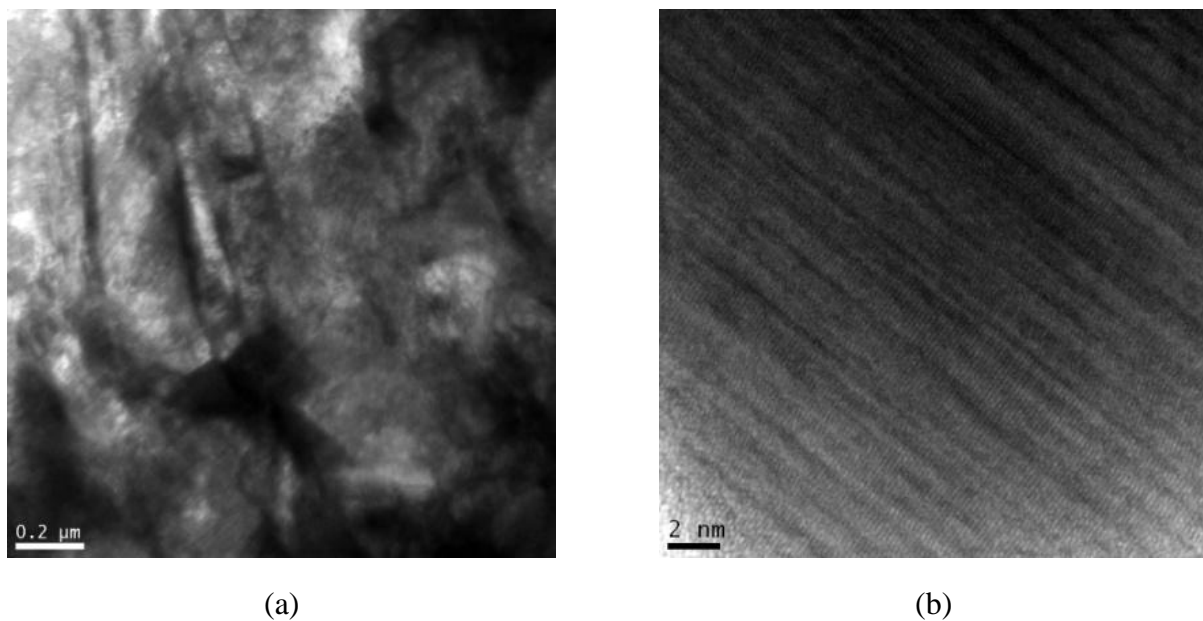
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Considering the deformation process condition the heterogeneity in microstructure formation was often observed across the bulk specimen in dependence of the introduced strain. As results of different straining applied the banded elongated subgrain structure is present due to dominant shear strain.

With increasing deformation, a part of the dislocations was absorbed by subboundaries and enlarged the disorientation among the subboundaries changing into boundaries of low and high angle.

It is also observed a reduction in the number of low angle boundaries and increased the number of high angle boundaries with the rise of the number of passes made in nickel alloy 600 and contributes to a substantial number of activated slipping plans of the same family.

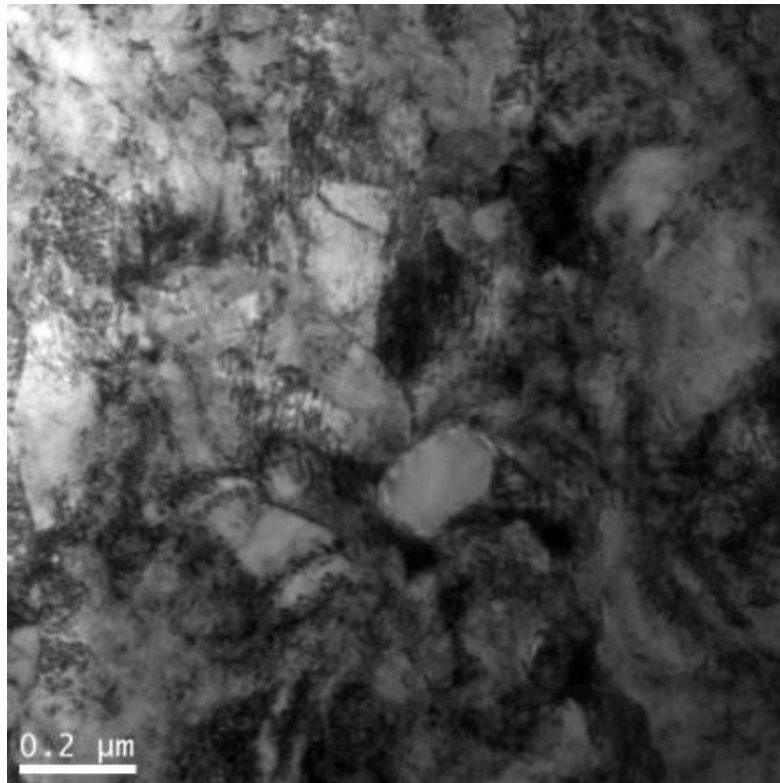


**Figure 9** - Electron micrograph (TEM) of nickel alloy 600 heat treated in 673K during 1800s followed by the intense deformation processing (SPD). Route B<sub>C</sub>, four passes. (a) Elongated grains and subgrains with dislocation tangles; (b) Details of the presence of microtwins inside subgrains with preferred shear deformation direction that motivates possible subsequent formation of nanograins in the twin walls during shear rotations by SPD process.

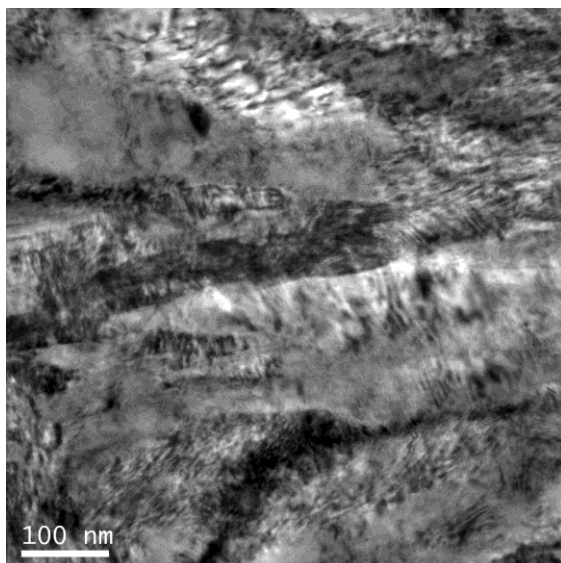
The microstructural characterization by TEM shows an indication of parallel arrangements of microtwins formation inside the grains observed in addition to have in several regions intersections between them.

There are also presence of some elongated grain and in some places along the boundaries of elongated grains are also observed embryos of new grains (nanograins of 200 nm).

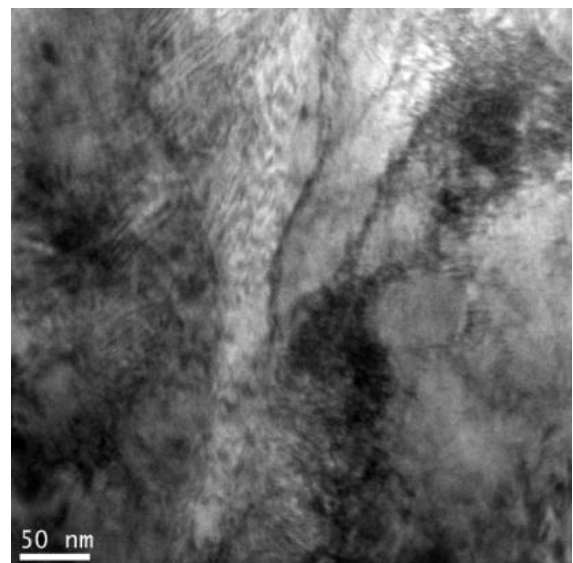
The presence of a large number of dislocations takes some difficult to this measure (Figures 9 and 10). The analyzed diffraction patterns indicate polycrystalline material with random grains orientation.



(a)



(b)



(c)

**Figure 10** - Electron micrograph (TEM) of nickel alloy 600 heat treated at 400° C during 1800s followed by the intense deformation process (SPD - B<sub>C</sub> route after 6 passes). (a) Presence of dislocations tangles inside the grain size some of them reduced probably due to additional energy of successive shear passes occurred during the process; (b, c) subgrains of varying sizes with dislocations, dislocation rings as well thinner precipitates, presence of microtwins inside the subgrains due to shear applied that may indicate possible subsequent formation of nanograins on your walls.

Seemingly these microtwins can act as new grains nuclei (supposed nanograins) due to the: (a) generation of a high density of dislocations within the microtwins; (b) subsequent formation of deformation groups of inside the microtwins. Initially, during the SPD process, dislocations can pile-up near the walls of the microtwins giving possibility to produce new grain nuclei and also some atomic planes during rotation process divided the existing microtwins, producing new nano domains with a possible nanograins formation [11].

## Conclusions

Regarding the microstructural observation of nickel alloy 600 subjected the procedure PCE (Bc route) it can be concluded that the intense deformation process produces a microstructural variation with respect to the size of the nanograins obtained after PCE process. The multiple deformations of the microtwins accommodating of plastic deformation applied and the presence of second phases cooperates to the nanograins mechanisms by creating a large number of dislocations and also the interaction dislocations-precipitates.

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