

Effects of severe plastic deformation on microstructure and mechanical properties of phosphorous sintered steel parts

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Severe Plastic Deformation (SPD) became a known method for crystal grain of metals refining, improving, on this way, their mechanical properties. However, up to day, its application to sintered materials has been less investigated, in spite of its high potential of crystal grain refining but also of densification by pore closing. Consequently, the possibility of its application to sintered structural steel parts, in conditions of preserving their required shape and dimensions, is being considered in this paper. By analysing the known SPD variants, one was established that the most appropriate for this purpose is a combination of Constrained Groove Pressing (CGP) with Bulk Mechanical Alloying (BMA). It consists in sintered part repressing in a die of similar, but larger cavity, of the required final dimensions, producing compression and shear stresses and transverse deformation with pore closing and microstructure refining. Considering this potential, its application for sintered structural part densification and their grain refinement, parts obtained from one of the most used powder, PASC 60, low alloyed with phosphorous, due to its ability to form a transient liquid phase at sintering, improving, on this way, the sintering process but, facilitating, in the same time, the crystal grain growth, has been investigated.

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1. Introduction

Phosphorus is added in sintered steels to activate the sintering process at common sintering temperatures (1120÷1150 °C) by facilitating neck formation between adjacent particles of the base steel powder, resulting in pore dimensions reduction and their, at least, partial spheroidisation, both leading to mechanical properties enhancement [1]. Two mechanisms are responsible for these effects:

i) formation of a transient liquid phase, in small proportions, by melting of the Fe-Fe₃P eutectic at 1048 °C (Fig.1) [2];

ii) notable increasing of self-diffusion coefficient (of ~ 300 times) at Fe_γ to Fe_α transformation [3] while P concentration decreases below 2.6 wt.%.

Really, at an enough high P content in steel powder, during sintering at 1120 °C, the P concentration at the surface of the steel powder particles temporarily exceeds 2.6 wt.% - limit of P solubility in Fe_α (see Fe-P phase diagram in Fig. 1), and the Fe-Fe₃P eutectic forms at the iron particle surface, determining their superficial melting and forming a liquid phase. So, the sintering process transforms from solid phase in liquid phase sintering, in which, as it is known [2], necks between adjacent particles are forming much faster. Simultaneously, P – of a higher concentration at the particle surface, diffuses deeper into the iron core and, consequently, its concentration at the surface drops below 2.6 wt. %. As a result, the liquid phase disappears (this is why it is called “transient”). But, the P concentration dropping below 2.6 wt.%, to ~ 0.5

wt.%, leads, according to the phase diagram, to Fe_γ (austenite) into Fe_α (ferrite) transformation. As the self-diffusion coefficient of Fe is about 300 times greater in ferrite than in austenite, the sintering mechanisms occur much faster [4].

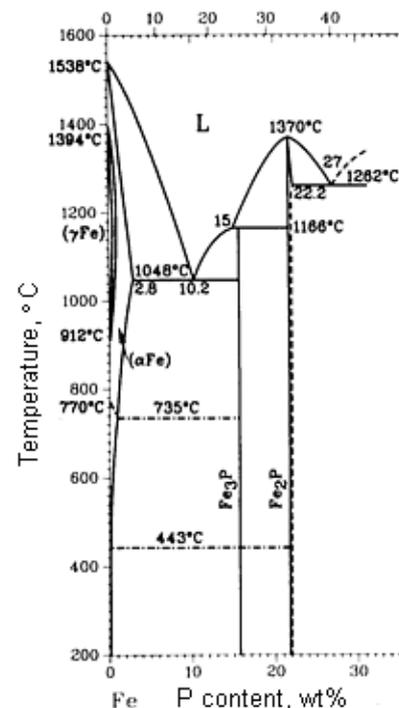


Fig. 1. Fe-P phase diagram [2].

However, both the presence of liquid phase and the higher self-diffusion coefficient in ferrite, beside the favourable effects of sintering activation, simultaneously determine a notable grain growth, which leads to the mechanical strength decreasing [5]. To avoid this undesired effect, it is necessary either to prevent the grain growth during sintering – which counteracts the beneficial effect of sintering activation – or to subsequently refine microstructure of sintered steel parts.

The last solution is, certainly more advantageous. An efficient way for this purpose seems to be Severe Plastic Deformation (SPD) [6]. It became a consecrated method for obtaining very fine crystalline and even nanocrystalline structure in bulk metals and alloys, including steels [7, 8], having as a result their mechanical properties enhancing and even a superplastic behaviour at lower temperatures [9]. For a full practical exploitation of these effects, numerous procedures of SPD application, such as Equal Channel Angular Pressing (ECAP), High Pressure Torsion (HPT), Constrained Groove Pressing (CGP), Repeated Die Forging (RDF), Bulk Mechanical Alloying (BMA) etc. have been developed [6, 8,10-12]. Also, an intensive research activity is performed to establish their potential in microstructure refining and mechanical properties of various types of bulk metallic materials improving [13].

In spite of this proved potential of microstructure refining and properties enhancing, there are only few published papers concerning SPD application to obtain the same effects in sintered materials, including sintered structural steel parts, even if, beside grain refining, SPD is expected to lead to their densification by pore closing under the effect of applied shear stresses. Or, it is known that the inherent porosity, which diminishes mechanical properties of sintered steel parts in comparison to of their wrought counterparts, is the main drawback that limits their extending to a wider range of applications. This is in spite of the recognized advantages of Powder Metallurgy (PM) in sintered part fabrication in comparison with classical metallurgy [1, 14]. So, the most SPD applications in PM refer to metal powders or metal-non-metal powder mixtures advanced consolidation to obtain metal, respectively composite billets [15-17] and also to the sintered porous semifabricates densification [18] by the most investigated SPD procedure - ECAP or by its variants (BP-ECAP, ECAP-FE, FE-ECAP), all revealing improved specific properties (relative density, homogeneity of reinforcing particles distribution, hardness) obtaining. However, being, in fact, an extrusion process [19], ECAP can not be applied for densification and crystal grain refining of the common near-net shape sintered parts, other SPD procedures being more appropriate for this purpose. Among these, very good results in grain refining and mechanical properties improving have been obtained by HPT [20] and also by CCDF (Cyclic Close Die Forging) [21], but they can be applied only to disc, respectively rectangular parts. More appropriate for the common sintered part seems to be CGP. A variant of this procedure has been investigated by J. Asami et al. [22], by adopting, in the process of sintered structural steel part fabrication, of die compaction at 2000

MPa to obtain green compacts, realizing, on this way, a SPD of powder particles. Subsequent sintering of these green compacts in appropriate conditions led to mechanical strength equivalent to that of the full dense compacts. So, they established that green densities up to 7.6 g/cm^3 and a fine-grain microstructure can be achieved on this way. Moreover, these properties can be strictly controlled by selecting degree of deformation and applied pressure.

However, any of the published information has been found concerning SPD application directly to the already sintered structural parts in condition of keeping their shape and dimensions. Even this possibility has been considered in the present research, it being focussed on the further densification as well as on the crystal grain refinement and, on this way, on mechanical properties improving of the above specified phosphorous containing sintered steel – one of the most used for structural sintered part applications [1]. By analysing the above-presented SPD variants, was established that the most appropriate for sintered parts in order to keep the part shape and assure obtaining of the required precision is also CGP, but combined with BMA. It consists in the part, realized by the common cold compaction-sintering route, but at smaller dimensions than of the final part, re-pressing with an appropriate pressure, in a die having cavity of similar shape, but of the larger required final dimensions. According to the Schmid's law [5], even to an applied axial compression stress, shear stresses also appear in the transverse directions, determining the material deformation both by compression and by shear. In the case of porous materials, this determines, beside crystal grain refining as in bulk materials, the pore shear and can lead to their closing by the walls cold welding. Consequently, it was adopted in this research. The effects of the cold compaction pressure and sintering parameters, used for the part processing, as well as of the re-pressing pressure, on the final microstructure, density, hardness, ultimate tensile strength and elongation of structural parts obtained from one of the most used phosphorous containing prealloyed steel powder have been established.

2. Experimental procedure

Höganäs PASC 60 powder, containing 0.6 wt.% P, has been adopted for phosphorous steel sintered samples realization [23]. For the sample preparation one has been adopted a similar technique as in [24], where the SPD effects on the sintered low alloyed with Cu, Ni and Mo has been investigated. So, the adopted powder has been uniaxially cold compacted at different compacting pressures (200, 400, 600 and 700 MPa) in nine cylindrical samples for each pressure, with a diameter of 9.24 mm (cross sectional area, $S_0 = 67.06 \text{ mm}^2$) and a height of 8 mm, in a pressing tool with floating die, with die wall lubrication (Zn stearate). Beside these cylindrical samples designed to density determination, three tensile test samples (ISO 2740:2009) have been realized but only at the highest above-specified compacting pressure (700

MPa). All green samples were sintered in industrial conditions, in a belt sintering furnace, at 1120 °C, in endogas with a dew point of - 40 °C, for 40 min, followed by cooling in the water-jacket cooling zone of the sintering furnace (cooling rate ~30 °C/min).

The obtained cylindrical samples were characterized by microstructure examination in an axial section through optical microscopy, after etching with Nital 3%, on an Olympus 50 x ÷ 1000 x inverted metallurgical microscope with digital JPG image camera, density and hardness determination by the Archimedes' method (ISO 2738:1999/ASTM B962) and by Rockwell B method (ISO 6508-1:2005/ASTM E18) respectively.

In agreement to the adopted combined CGP-BMA variant of SPD, for the sintered cylindrical sample re-pressing a rigid die of a diameter of 11.28 mm ($S = 100 \text{ mm}^2$) has been adopted, respectively with a degree of transversal deformation, $\delta_{tr} \approx 33 \%$, with pressures of 1400, 1600 and 1800 MPa and die wall lubrication using the same lubricant as for the initial pressing. To avoid the die crack, it has been consolidated with a 205CrII5 high strength steel (ASTM A-120) clamping ring. Three cylindrical samples realized at each compacting pressure and sintered in the above mentioned conditions, have been re-pressed at each adopted re-pressing pressure (i.e. a total of 45 samples). However, in the case of tensile test samples, because the construction of a die of a higher cross section than of the standard one and also its consolidation was more difficult, for their re-pressing has been adopted the same die as for their compaction and only one re-pressing pressure, respectively 1200 MPa. To accomplish the condition of sample transversal deformation during re-pressing in the same die as for compaction, beside their cross section reduction by shrinkage at sintering, a supplementary reduction has been realized by uniform grinding along the contour with a cutting depth of 0.86 mm, to assure a transverse

deformation degree as that adopted for cylindrical samples ~33%).

The re-pressed cylindrical samples were characterized by microstructure optical examination, density and hardness determination in the same way as before re-pressing. Both as-sintered and re-pressed cylindrical samples were subjected to crystal grain size distribution analysis, in a similar axial section as for metallographic examination before re-pressing, using the facility of the above-specified microscope, while tensile samples to tensile test at room temperature, in conditions of the above mentioned ISO 2740:2009 with a loading speed 1.00 mm/min, on the universal testing machine Sun 5 (Galdabini, Italy), with the stress – strain diagram simultaneously recording.

3. Results and discussion

In Fig. 2 there are comparatively presented the microstructures of sintered samples before and after SPD by CGP-BMA application. As can be seen, there is a notable difference between them: while in microstructure of sintered samples before repressing (Fig. 2-a) quite equiaxial crystal grains and rounded pores of dimensions up to about 20 μm (of a dark colour) can be distinguished, a dense microstructure has been formed by re-pressing (Fig. 2-b), in which porosity has been eliminated to a great extent - the remaining pores being much smaller, flattened and elongated toward the direction of plastic deformation. Also, fine elongated crystal grains oriented toward the main direction of plastic deformation (by combined effect of compression and shear) and a pronounced fragmentation of eutectic occurred. Certainly, these effects depend upon the deformation degree and re-pressing pressure, but also upon the initial density, respectively porosity, and, implicitly, upon the compacting pressure.

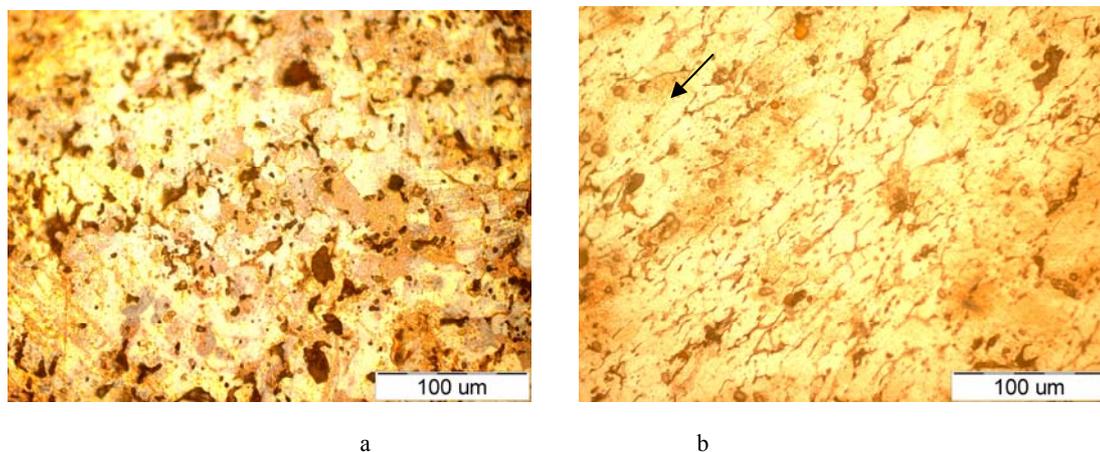


Fig. 2. Typical microstructure of sintered samples: a) before re-pressing; b) after re-pressing with 1800 MPa. Deformation degree: 33 %. The main deformation direction: along the arrow.

The correlation between density – compacting pressure and re-pressing pressure for the same degree of

transversal deformation is graphically presented in Figure 3. For better reflecting the effect of re-pressing pressure on

the densification process, on the right Y axis of the graph has been represented the compactness, i.e. relative density, values - established in respect to theoretical density of PASC 60 material, calculated taking into consideration its composition {(0.6 wt. % P and 0.03 wt.% C – balance Fe (99.37 wt.%))[23] and density of components respectively. As can be seen, by the common PM route of cold compaction and sintering (0 value of re-pressing pressure on diagram), the obtained density values are in the limits provided by the PASC 60 powder producer, Höganas AB co. [23], proving that these operations occurred normally. Instead, as has been expected, a notable increasing of density, respectively of compactness occurred by re-pressing. Certainly, the highest density (filled points) / compactness (un-filled points) values have been obtained for the highest re-pressing pressure (1800 MPa) and for the highest compaction pressure used in the green compact realization (700 MPa), but well above 90 % relative densities have been also obtained for the re-pressing pressure of 1600 MPa and even of 1400 MPa as well as for the compacting pressures of 600 MPa and even of 400 MPa. All these findings prove that the method can be applied, without difficulty, at the industrial scale as 1600 and 1400 MPa re-pressing pressures can be easier realized than 1800 or, certainly, than 2000 MPa above mentioned as adopted by J. Asami et al. in their research [22]. Also, 600 and 400 MPa compacting pressures are more often used than 700 MPa in sintered structural part fabrication to prevent excessive energy consumption and wear of pressing tools [1].

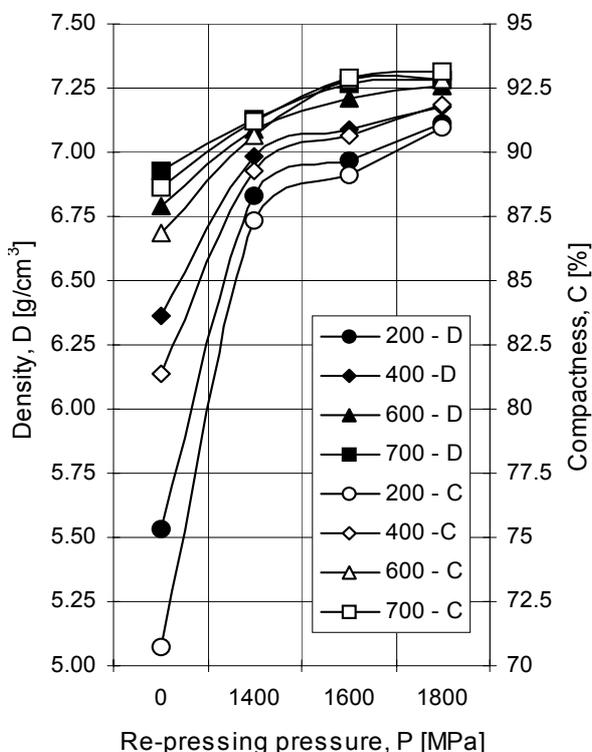


Fig. 3. Effect of re-pressing pressure on density and compactness of sintered compacts for the used compacting pressures. Deformation degree: 33 %.

To evaluate the re-pressing behaviour of sintered PASC 60 steel parts and the work hardening influence on re-pressing without their intermediate annealing – advantageous from the cost point of view – the compactness increasing as a function of the re-pressing pressure has been calculated and also the main mechanical properties were determined. In Fig. 4 is presented the compactness variation as a function of re-pressing pressure for all the used compacting pressures. Certainly, the increasing of this characteristic by re-pressing is as high as re-pressing pressure is higher, but also as the cold compaction pressure is smaller (initial compactness is lower). However, its values exceeds 6 ÷ 10 % for the common compacting pressures (400 ÷ 600 MPa) and the easier accessible 1400 ÷ 1600 MP re-pressing pressures, proving that the above conclusion concerning the possibility of SPD application, at the industrial scale, for densification of sintered structural parts made of PSC 60 powders, in advantageous conditions, is valid. It is to be observed that even for the compacting pressure of 700 MPa, a densification of over 5 % can be obtained for the same moderate re-pressing pressures, in spite of the higher increase in volume associated to higher spring back determined by the more pronounced cold hardening occurring in these more severe compacting / re-pressing conditions [4]. This last observation proves that densification of sintered part at SPD occurs, in principal, by pore closing, the adverse effect of steel cold hardening produced by SPD being smaller.

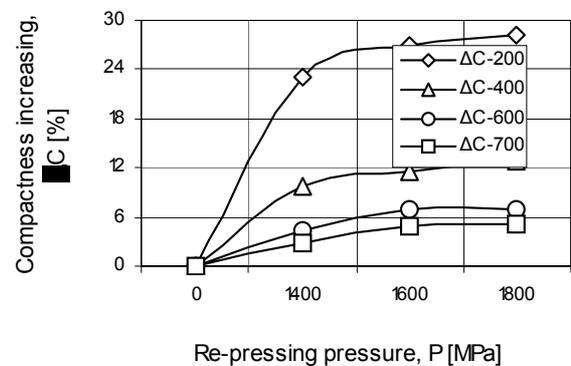


Fig. 4. Effect of re-pressing pressure on compactness of sintered compacts for the used compacting pressures. Deformation degree: 33 %.

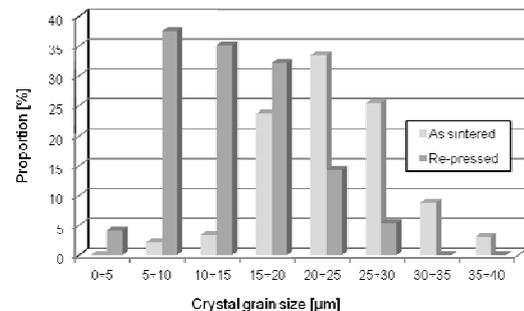


Fig. 5. Crystal grain size distribution of sintered samples compacted at 700 MPa, before and after re-pressing with 1800 MPa. Deformation degree: 33 %.

Beside the above-analysed densification by pore closing, a significant reduction in crystal grain dimensions and their alignment in the predominant deformation direction occurred. Even if it is clearly highlighted by the sample microstructures before and after SPD comparative presentation in Figures 2-a and b, a more comprehensive illustration is given by the grain size analysis, presented in Figure 5, quantitatively proving this crystal grain refining. As can be seen, the initial dimensions of the crystal grains are approximately distributed upon a Gauss curve, with the a medium grain size of $20\div 25\ \mu\text{m}$, while after re-pressing, a similar Gauss curve of the grain size distribution has been obtained, but pronounced shifted to the left, with the medium grain size of $5\div 10\ \mu\text{m}$, but with more than 70 % of the total crystal grains with average dimensions less than $15\ \mu\text{m}$. However, it is to be remarked that the dimensions of the elongated crystal grain size formed by re-pressing included in diagram from Fig. 5 are the average of their length and thickness, i.e. a certain error in their true dimension expression could occur.

As has been expected, densification, cold hardening and crystal grain refining produced by SPD have a notable effect on mechanical properties of the considered sintered parts. So, notable hardness increasing occurred by re-pressing. For the graphical representation clarity, in Fig. 6 is presented its variation as a function of the re-pressing pressure only for the compacting pressure of 700 MPa, but similar curves have been obtained for all the adopted compacting pressures. One can be remarked that hardness increases as re-pressing pressure increases only up to $1500\div 1600$ MPa. It is to be observed that this is in correlation with the densification increasing presented in Figure 4, which, for the compacting pressure of 700 MPa, also becomes stable for re-pressing pressures over ~ 1600 MPa. This proves that densification process has a predominant effect on the hardness of considered sintered part increasing by SPD, higher than cold hardening.

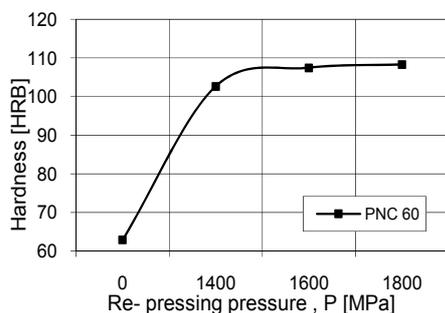


Fig. 6. Effect of re-pressing pressure on the hardness of the obtained samples, for the cold-compacting pressure of 700 MPa. Deformation degree: 33%.

Both densification and crystal grain refining have a notable effect on the tensile behaviour of the considered sintered steel. This is convincingly illustrated by the stress-strain diagram recorded during the re-pressed tensile

sample testing, presented in Figure 7. As can be seen, not only tensile characteristics have been changed, but also the steel behaviour in tensile. While before re-pressing a yield stress appeared at 257.2 MPa and the fracture occurred at a tensile strength of 308.2 MPa and an elongation of $\sim 8\%$, by re-pressing a toughening effect occurred, determining the tensile strength increasing to 411.4 MPa and also a notable increasing of Young's modulus, but also, as expected, a certain decreasing of elongation, to $\sim 4.8\%$. As can be remarked, a notable tensile strength increasing, of 33.48 %, has been obtained by the sample re-pressing with only 1200 MPa – an accessible pressure for SPD industrial application.

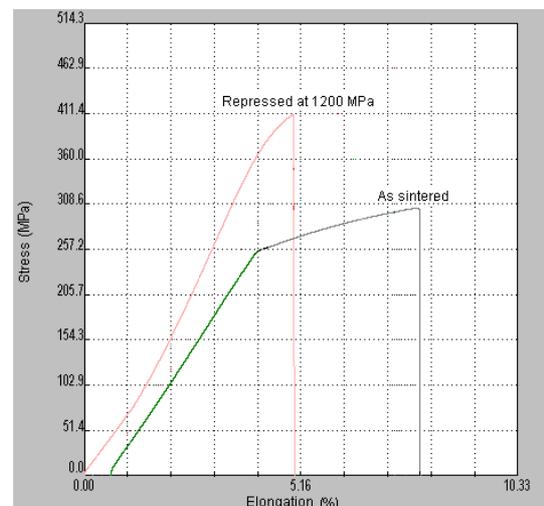


Fig. 7. Tensile Stress-Strain curves of the initial, cold pressed with 700 MPa and sintered compact and of the re-pressed compact with a re-pressing pressure of 1200 MPa. Deformation degree: 33 %.

4. Conclusions

The above presented results prove that SPD in the combined CGP-BMA variant can be successfully applied for density increasing and especially crystal grain refining, determining, on this way, mechanical properties improving, not only to Distaloy type sintered structural steels, previously investigated by the authors [24], but also to sintered steels low alloyed with phosphorous – one of the most used in sintered structural parts fabrication due to its ability to sinter with a transient liquid phase formation, improving on this way, the sintering efficiency. In the GCP-BMA variant, i.e. by the sintered compact further die compaction with transverse deformation of material, SPD can be applied to parts of a high complexity, preserving their geometrical shape and precision. There were established that density increasing occurs, in principal, by the pore closing, the hardness increasing being close related to this densification. Beside densification, a notable crystal grain refining occurs, a decrease, by re-pressing with 1800 MPa, of the medium grain size from $20\div 25\ \mu\text{m}$

to 5÷10 µm, but with more than 70 % of the total crystal grains less than 15 µm.

For the common industrially used compacting pressures (400÷600 MPa) and accessible re-pressing pressures, of 1400÷1600 MPa, an increase in density of about 6 ÷ 12 %, in hardness of over 30 %, in UTS of more than 33 %, accompanied by a notable increase in the Young's modulus with a certain decrease of elongation, but its keeping at reasonable values, of ~(4.5÷5.0) % - the last two denoting a high enough toughness of the processed PASC 60 sintered steel, can be achieved in quite simple, safe and controllable conditions, replacing the classical calibration operation with SPD, preserving, in the same time, the part shape, dimensions, precision and surface quality.

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