Effect of preliminary and post-SPD aging on nanostructuring and strengthening of the HPT processed 2024 aluminum alloy

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Abstract. Structural and mechanical behaviors of the 2024 commercial aluminum alloy, differed by heterogeneity of the initial structure, were examined after severe plastic deformation and further annealing. The superior strengthening of the pre-quenched alloy under high-pressure torsion (HPT) at room temperature was caused by the development of a more equiaxed nanostructure of the matrix with a lower fragment size than in pre-aged T6 condition. Post-HPT annealing at 100 °C led to extra strengthening of the pre-quenched alloy only due to the age hardening effect. The nature of the alloy behavior is discussed.

1. Introduction
Recent data have shown high potential for improving the service properties of metallic materials due to novel approaches to their structure/phase control through imparting the size less than 100 nm to the main structural components. Such nanostructuring is of particular interest for commercial materials owing to intense studies of the phenomenon of their cold severe plastic deformation (SPD) to effective strains of $e > 1$ [1]. To date, a wide range of metals and alloys has been nanostructured and several industrial applications have been announced. However, the nature and structural factors responsible for the effective formation of nanostructures of different types with a guaranteed balance of properties are still not so clear. As for age-hardenable aluminum alloys, these points are mainly controlled by the structure of the matrix and of the secondary phases. The effect of the latter is quite ambiguous and less studied. For example, it was established [2,3] that the preliminary formation and/or increase in the volume fraction of disperse phases resulted in processing of a greater fraction of nanograins of a lower size under SPD. In contrast, it was shown in [4,5] that the formation of nanosized precipitates of high densities in high-strength alloys can completely suppress recrystallization of the matrix.

One of the topical issues for the majority of wrought Al alloys is the estimation of effectiveness of post-SPD annealing in property control and aging response of the SPDed alloys. Along with the structural strengthening owing to the formation of new grains and subgrains, and an increase in the density of dislocations due to SPD, the mechanical behavior of the alloy with further annealing will be also determined by the balance of the disperse hardening and aluminum solid solution strengthening. Though the contribution of the latter to the strength of the SPDed materials is comparatively small, alloying elements should provide noticeable post-deformation hardening of the matrix with the formation of nanosized precipitates owing to decomposition of the supersaturated Al solid solution obtained under preliminary quenching or during SPD. Besides, the strengthening effect of precipitates in the severely deformed matrix should depend on their morphology as may occur in a non-conventional sequence of phase transformations [6]. Therefore, the development of the optimum
annealing regimes to realize the aging effect in the SPDed alloy is a rather important task, which requires an understanding of the nature of its structuring and strengthening. The aim of the study is to analyze the effects of preliminary and post-SPD aging on the structure and strength of the widely used commercial aluminum alloy subjected to cold high-pressure torsion (HPT).

2. Material and procedure
A hot-pressed rod of the 2024 alloy of a standard chemical composition (Al-4.2Cu-1.3Mg-0.5Mn, wt.%) was used as the starting material. Disk-shaped samples with a diameter of 20 mm and a thickness of 2 mm, cut out from the rod across its axis, were solution treated at 505 °C and quenched in water. Some of them were then aged at 190 °C for 10 hrs (T6 temper). SPD of the quenched (Q) and artificially aged (Q+AA) samples was realized through the HPT with 10 revolutions under a pressure of 6 GPa at room temperature. Post-SPD annealing was performed at 100 °C basing on the data of the previous study [7], showing that annealing at higher temperatures led to intense grain and precipitate coarsening and strong softening of the alloy. The microstructure was analyzed using transmission electron microscopy (TEM). The objects were prepared by electropolishing at -28 °C in 20% solution of HNO₃ in CH₃OH. Microhardness was determined at a load of 1N. Tensile tests were performed on an Instron 5982 at room temperature on samples with a gage part of 1 × 1 × 3 mm³, and the axis coincided with the line tangent to the circle with half the radius of the deformed discs.

3. Results and discussion
TEM analysis showed that the starting material in the Q condition had a coarse-fibered microstructure with homogeneously distributed compact Mn-rich T-phase (Al₁₃Cu₃Mn₁) precipitates of 50-200 nm in size inside the fibers (figure 1a). Further artificial aging (Q+AA state) resulted in the decomposition of preliminary supersaturated by Mg and Cu aluminum solid solution, with the formation of the main strengthening needle-like (lamellar) S-phase precipitates (figure 1e).

Due to HPT, the well-developed, non-equilibrium nanostructure of the Al matrix with an average size of near equiaxed crystallites (grains and subgrains) of about 75 nm was formed in the as-quenched alloy (figure 1 b,f). A visible difference in the structure was found in the SPDed artificially aged alloy (figure 1 c,g): it was consisted of predominantly elongated cells (fragments) ~200 nm in size. Judging by the character of both structures, one can conclude that continuous dynamic recrystallization played a dominant role in their formation, and the precipitates had quite a strong effect on its occurrence. The nature of this effect was reported in [4,5]: an increase in the number density of precipitates by one or two orders of magnitude, involving dispersoids and main strengthening phases, could lead to a strong suppression of the nanostructuring of the Al matrix via fragmentation and/or recrystallization, owing to increasing the homogeneity of plastic flow and preventing dislocation rearrangements. As a result, the dislocation pile-ups were formed instead of fragments, and the SPD structure in the pre-aged alloy was less misoriented, having a more uniform distribution of dislocations, cells, and dislocation boundaries and a lower fraction of nanocrystallites. Further post-HPT annealing did not lead to visible grain coarsening (figure 1d) amid the activation of two main processes – recovery of the structure of the matrix and decomposition of the Al solid solution.

The microstructural changes caused by the processing had a strong effect on the mechanical behavior of the alloy. Thus, the hardness of the alloy in the solid-solution hardened Q state was 125 HV and increased to ~170 HV after the T6 treatment because of the precipitation hardening induced by the formation and growth of zones and metastable S phases. After HPT, the hardness in both alloy conditions increased to practically the same value of ~270 HV, testifying a strong dependence of the SPD strengthening on the initial structural heterogeneity of the alloy (figure 2). That is, the strain hardening of the pre-quenched alloy was more intense, and the strength of the preliminarily aged alloy was close to the limit reached under SPD in the Q alloy due to the formation of a more developed nanostructure. Such behavior cannot be explained from a common point of view based on the well-known structural effects, involving the Hall-Petch strengthening and dispersion hardening. This behavior is most likely caused by the interaction of dislocations with the clusters containing Cu and
Figure 1. TEM structures of the 2024 alloy in the pre-quenched (a,b,f,d,h) and further aged at 190 °C for 10 hrs (c,e,g), before (a,e) and after HPT (b,c,f,g), and post-HPT aging at 100 °C, 100 hrs (d,h).

Mg atoms, namely, the fragments of zones and metastable phases that were formed during SPD. This point of view was advanced and grounded in [8] and our results support it.

The same situation was found in tensile tests: the yield stress and ultimate tensile strength, measured for both initial conditions after HPT, were also close (table 1). Moreover, their ductility values were also equal. It is also necessary to note that the tensile strength of the SPDed alloy were twice as high as those of the age-hardened initial rod, and also those after the "removal" of the SPD hardening effect by further T6 heat treatment. However, in the latter case, the elongation to failure was an order of magnitude higher than in the SPDed alloy and twice higher than in the rod. This effect was caused by the homogenization of the spatial distribution and refinement of the excess phases under HPT [9].

Figure 2. Microhardness of the 2024 alloy vs time of post-SPD annealing at 100 °C.
Post-SPD annealing at 100 °C led to the alloy extra hardening in the Q condition only with the maximum reached at 50 hours (figure 2). This behavior was caused by the effect of disperse strengthening due to artificial aging. It should be noted that the products of aging in the form of lamellar zones and precipitates of the main strengthening phases were hardly distinguished by TEM and found only in the recovered areas with a more equilibrium structure (figure 1h). The morphology of these precipitates had no obvious differences from the conventional ones, except for their lower length, limited by the inter-grain boundary spacing. Therewith the loss in the alloy strength owing to the elimination of structural defects, and also a decrease in the level of alloying of aluminum solid solution, were compensated by disperse strengthening with even more than 10% of hardness increase.

The data obtained, especially the absence of aging response in the pre-aged alloy, allow us to conclude that HPT, even to high strains, does not result in sense dissolution of main strengthening phases and sense changing of the alloying of the matrix, frequently declared in the literature.

4. Conclusions
1. The room temperature-HPT of a commercial hot-pressed rod of the 2024 Al alloy with differed heterogeneity of the initial structure, performed by 10 revolutions at 6 GPa, led to a more equiaxed and developed nanostructure of the fragmented type in the pre-quenched alloy. In the pre-aged at 190 °C for 10 hrs condition (T6 temper), a mixed (sub)grain/cellular structure, preferably composed of coarser elongated crystallites separated by dislocation boundaries, was formed. HPT resulted also in superior strain hardening with the higher impact also obtained in the pre-quenched alloy.
2. In contrast to the pre-aged alloy condition, post-SPD annealing of the pre-quenched alloy at 100 °C led to its extra hardening. Therewith, the losses in the alloy strength owing to the elimination of structural defects, and also a decrease in alloying of the aluminum solid solution, were compensated by precipitation hardening with even more than 10% of strength increase. The data obtained allow concluding that the absence of aging response in the SPDed pre-aged alloy is conditioned by the absence of sense changes in the contents of the main alloying elements in the aluminum matrix due to severe straining, and dynamic dissolution of secondary phases, frequently discussed in the literature.

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References
[1] Valiev R Z, Estrin Y, Horita Z, Langdon T G, Zehetbauer M J and Zhu Y 2016 JOM 68 1216
[2] Rabinovich M Kh, Markushev M V and Murashkin M Yu 1997 Mater. Sci. Forum 243 591
[3] Barlow C Y, Hansen N and Liu Y L 2002 Acta Mater. 50 171
[4] Markushev M V, Avtokratova E V, Krymskiy S V, Sitdikov O Sh 2018 J. Al. Comp. 743 773
[5] Markushev M V, Burbastykhu Yu L, Krymskiy S V and Sitdikov O Sh 2017 Lett. Mat. 7 101
[6] Sha G, Wang Y B, Liao X Z, Duan Z C, Ringer S P, Langdon T G 2009 Acta Mater. 57 3123
[7] Markushev M V et al. 2018 IOP Conf. Series: Mater. Sci. Eng. 447 012008
[8] Chen Y, Gao N, Sha G, Ringer S P and Starink M J 2015 Mater. Sci. Eng. A 627 10
[9] Krymskiy S V et al. 2011 Perspective Mat. 12 387 [in Russian]