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Features of nanostructure and functional properties formation in Ti-Ni shape memory alloys subjected to quasi-continuous equal channel angular pressing

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Abstract. Influence of various deformation regimes, included severe plastic deformation by multi-pass rolling, rotary forging and equal channel angular pressing (ECAP) in the normal and quasi-continuous modes, on the structure and properties of Ti-Ni shape memory alloys was studied and compared. Features of structure formation providing a combination of high functional properties were analyzed. Investigation of the texture development, depending on the ECAP regimes and post-deformation annealing, was made.

1. Introduction

Ti-Ni-based shape memory alloys (SMA) are advanced functional materials. They allow implementing the performance characteristics of devices and technologies that are not achievable by using other materials [1-3]. Severe plastic deformation (SPD) is one of the most attractive methods of thermomechanical treatment that allows obtaining ultrafine-grained (UFG) structure and considerably improving mechanical and functional properties of Ti-Ni SMA without changes in chemical composition [4-8]. UFG structure is generally ranged into nanocrystalline (grain/subgrain size is less than 100 nm) and submicrocrystalline (grain/subgrain size from 100 nm to 1 μm) structures [9]. In accordance with the previous experience the best combination of mechanical and functional properties in Ti-Ni shape memory alloys can be achieved after formation of a completely nanocrystalline structure, with the size of structural elements 40-80 nm [10]. This structure was obtained in thin samples with a thickness of about 0.1 mm after multi-pass cold rolling. In order to obtain bulk nanostructured billets from Ti-Ni SMA the most promising SPD processes such as equal channel angular pressing (ECAP) and rotary forging at low temperatures were used [11, 12]. It was shown in previous studies that six-eight passes of traditional ECAP with the channels intersection angle of 110° and at the temperature not higher than 450 °C provides formation of mainly equiaxed ultrafine-grained (submicrocrystalline) structure. For the further grain refinement the number of passes was increased, and the deformation temperature was decreased. However, even after ECAP at 350 °C, the grain size remained in the submicrometer range of sizes (100-200 nm) [12]. Thus, for the very last time, the ECAP method allowed obtaining only a mixed submicrocrystalline structure. In this context, quasi-continuous ECAP was performed with the channel intersection angle of 120° at a temperature of 400 °C in order to obtain completely nanocrystalline structure in bulk samples [13]. The main features of the structure and functional properties in combination with the analyzes of the texture, obtained after ECAP and rotary forging with post-deformation annealing (PDA), in comparison with the multi-pass cold rolling, are described and discussed in the present work.

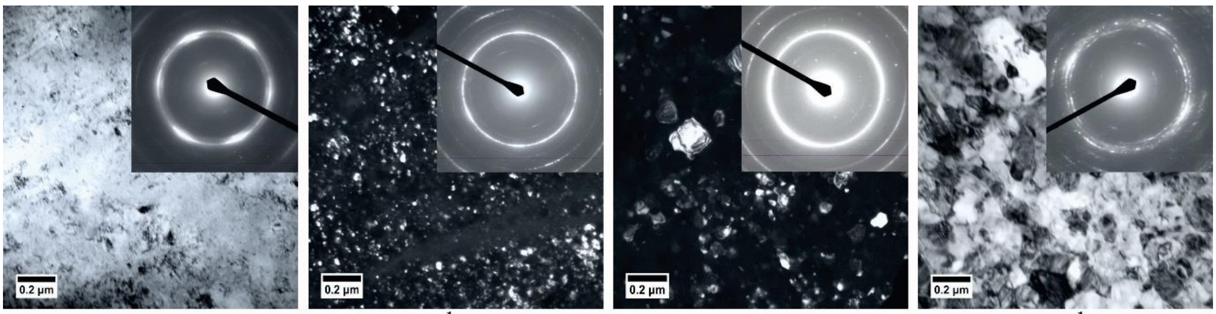
2. Experimental procedure

In the present work, TN-1 (from Ti-50.0 to Ti-50.2 at.% Ni) alloy, supplied by “Industrial Center MATEK-SMA Ltd.”, was studied. The billets for rotary forging and ECAP, rods 20 mm in diameter, were produced by screw rolling at 850-950 °C with reduction of 7-20 % per pass and interpass heating (hot-rolled state). ECAP was carried out in the traditional regime with reheating between passes at 450 °C for 20 passes and in the quasi-continuous regime at the temperature of 400 °C for 3, 5, and 7 passes. The channels intersection angle for traditional and quasi-continuous modes was 120°. The rotary forging was made from a diameter of 20 to 5 mm at 450 and 350 °C with relative strain from 1 to 5 % per one pass. The structure was studied at room temperature using "JEM-2100" transmission electron microscope. To analyze the crystallographic texture of deformed samples by direct pole figures (DPF) automated diffractometer "DRON-3", equipped with texture set, was used. This method consists in the consequential recording of the integrated intensity of the selected X-ray reflection from the {hkl} planes with rotation and inclination of the sample around the normal to the selected sample section [14]. The reflection intensity {hkl}, recorded for different sample orientations, was used to obtain a partial textural direct pole figures DPFs {100}, {110} and {112} for section perpendicular to the axis of the studied rod. The main axes of deformation are the compression direction (CD) and the tension direction (TD) for the last pass. For investigation of B2-austenite texture, the sample was heated to a temperature of 80-120 °C. A texture comparison was performed after ECAP and after rotary forging. The mechanical properties were determined at room temperature by the uniaxial tensile tests using universal tensile machine "INSTRON 3382" with a deformation rate of 4 mm/min. The maximum completely recoverable strain was estimated by a thermomechanical method using band samples.

3. Results and discussion

3.1 Microstructure

The structure of flat samples with a thickness of 0.13 mm obtained by multi-pass cold rolling and post-deformation annealing (PDA) by various regimes is shown in Figure 1. Directly after multi-pass cold rolling (accumulated true strain e = 1.9) a mixed nanocrystalline and amorphous structure was formed (Figure 1). During the PDA, the amorphous structure crystallizes into a nanocrystalline, the new grains (obtained both due to deformation and crystallization of the amorphous structure) grown, but their average size is less than 100 nm up to 400 °C. The main features of this structure are, firstly, its "bimodal" character: some grains retain a size of 5-20 nm, while others grow to a size of 30-80 nm and, secondly, a low dislocation density after crystallization from the amorphous phase.



a b c d

Figure 1. Microstructure of the TN-1 alloy subjected to multi-pass cold rolling (MPCR) according to various regimes: (a) MPCR, (b) MPCR + 250 °C, (c) MPCR + 350 °C and (d) MPCR + 400 °C. Transmission electron microscopy: dark-field images, electron-diffraction patterns in the insets.

During the process of ECAP, principally different mechanism of the ultrafine-grained structure formation as compared with cold rolling appears, comprising dynamic polygonization and recrystallization. Due to the repeated changes in the deformation direction, band structures (deformation localization bands) with a multidirectional orientation and great number of their intersections form. This leads to a considerable grain refinement and a continuous increase in the grains misorientation. In this case the dynamic polygonization and formation of subgrains precedes the dynamic recrystallization. A small size of these subgrains is mainly responsible for a high deformation hardening. The supposed mechanism of grain refinement after SPD at elevated temperatures consists in the gradual transformation of the originally formed subgrained dislocation substructure into a final structure with submicron and then nanocrystalline grain size. The lowering of the deformation temperature leads to a decrease in the intensity of dynamic softening processes (dynamic recovery) and, as a consequence formation of grains with smaller average size. The dynamic properties of various dislocation substructures, the acting slip systems and the deformation modes also change. All these aspects affect the properties of the finally formed microstructure.

The structure of alloy under all studied regimes of ECAP is shown in Figure 2.

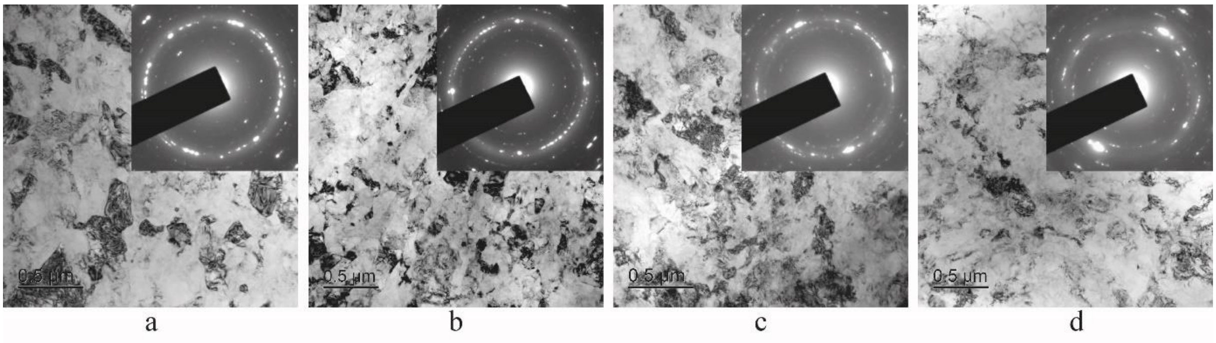


Figure 2. Microstructure of the TN-1 alloy subjected to deformation according to various regimes: (a) ECAP-20, (b) ECAP-3, (c) ECAP-7, and (d) ECAP-7 + 400 °C. Transmission electron microscopy: dark-field images, electron-diffraction patterns in the insets.

According to the results of the microstructural analysis, in the case of ECAP-20 at 450 °C, a mixed ultrafine grained/subgrained structure with high density of free dislocations is defined. The average linear size of the structural elements is 171±10 nm, therefore, this structure can be referred to a submicrocrystalline (Figure 2 a). The decrease in the deformation temperature to 400 °C and the exclusion of interpass heating lead to a refinement of structural elements. After ECAP-3, a mixed grained/subgrained structure with a large number of grains/subgrains with a size of less than 100 nm is produced. The average linear size of structural elements is 115±5 nm, i.e., it approaches the nanometer range from above. With an increase in the number of quasi-continuous passes to seven, the average size of structure elements does not significantly change, it rather stays at the boundary between the nanometer and submicrometer ranges (103±5 nm). The high density of free dislocations within the structural elements remains. Annealing after ECAP-7 at the deformation temperature (400 °C, 1 h) visually leads to a decrease in the dislocation density inside the grains/subgrains. Thus, after ECAP in 7 quasi-continuous passes, a mixed nanostructure of B2-austenite is produced. The size of structural elements of this nanostructure significantly exceeds the size, which was obtained by PDA after multi-pass cold rolling (40-80 nm), but the difference between completely recoverable strain values, obtained by PDA after cold rolling on a thin sample and ECAP on a bulk sample, is not so significant.

3.2. Comparison of mechanical and functional properties
In order to understand the main features of the structure, obtained after multi-pass cold rolling, and allow achieving the best combination of functional properties, such as recovery stress and completely recoverable strain, it is necessary to determine the conditions providing reduction of hindrances for reverse movement of shape recovery carriers. The first condition consists in the relatively high value of the austenite dislocation yield stress σ_y that serves as an approximate measure of the recovery stress value σ_r. The second condition is a relatively low value of the critical stress σ_{cr} for reorientation or

formation of martensite. The maximum difference between these two values $\Delta\sigma = \sigma_y - \sigma_{cr}$ was obtained directly after multi-pass cold rolling and allow achieving an ultimately high combination of functional properties (completely recoverable strain of 8.0 %, and recovery stress of 1400 MPa) (Figure 3) [15].

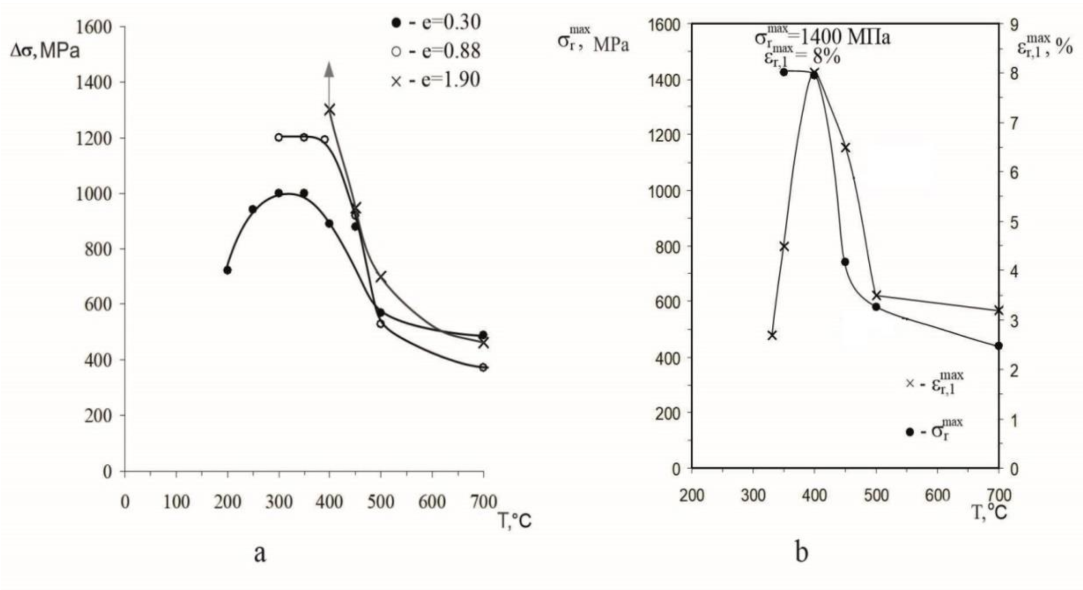


Figure 3. The values of $\boldsymbol{\Delta\sigma}$, $\boldsymbol{\sigma_r^{max}}$, $\boldsymbol{\varepsilon_{r,1}^{max}}$ after multi-pass cold rolling as function of the temperature of PDA. Arrow means that dislocation yield stress wasn't achieved during the tests [adapted from 10].

Formation of nanoscale grains with optimal degree of dispersion (40-80 nm) as a result of PDA at 400 °C after multi-pass cold rolling with accumulative true strain e = 1.9 increases the value of σ_y and the value of $\Delta\sigma$, but dislocation density in the obtained structure is low in comparison with the polygonized structure, when the high value of σ_y defined by high dislocation density. PDA at higher temperatures leads to the growth of austenite grains, consequent softening and degradation of functional properties. The structure with smaller grain size (less than 40 nm) after PDA at 350 °C, inhibits development of martensitic transformation and thus decreases the recoverable strain to 4.5 % and below. Nevertheless, the recovery stress remains at a very high level (1400 MPa) [15].

Equal channel angular pressing with the channel intersection angle of 120° at 400 °C for seven passes in the quasi-continuous regime allows achieving a mixed nanocrystalline and nanosubgrained structure with the average size of structure elements of 103±5 nm. As mentioned above, that is larger than the optimal range of 40-80 nm. Obtained structure is characterized by high dislocation density and the high ultimate tensile strength (1000-1150 MPa) in combination with the elongation to failure more than 20 %. The maximum completely recoverable strain in bending tests (9.5 %) was achieved after additional PDA at 400 °C for 1 h. Increase of the completely recoverable strain from 7.3 % after ECAP to 9.5 % after ECAP + PDA seems to be consequence of a decrease of the transformation yield stress and the retention of the high dislocation yield stress and, as a result, of an increase in their difference $\Delta\sigma$ [10].

3.3. Features of texture development during ECAP and rotary forging

The formation of crystallographic texture as a result of warm rotary forging (350-450 °C) and ECAP under different regimes described above has been studied. Partial DPFs {110} of rods deformed by warm rotary forging are shown in Figure 4. Partial DPF {110} of rod deformed by ECAP-20 with heating between passes up to 450 °C is presented in Figure 5a, and PPF {110} of rod deformed by ECAP-3 in quasi-continuous regime at the nominal temperature of 400 °C for three passes is shown in Figure 5b. All DPFs are presented for the section perpendicular to the axes of the rods.

According to the results obtained (Figure 4a), in the case of rotary forging at a temperature of 450 ° C, an axial tensile texture, at which the <110> axis is oriented along the direction of drawing of the rod, is formed in TN-1. Such a texture is typical for the deformation of the ordered crystals with a CsCl-type lattice along the slip systems {110} <001> [16]. When the deformation temperature is reduced down to 350-400 °C, an additional component with an axis <100> appears in the rod texture. Formation of this component can be connected with activation of other slip systems, for example, {001} <100>, as it was observed in the rolling of single crystals [17].

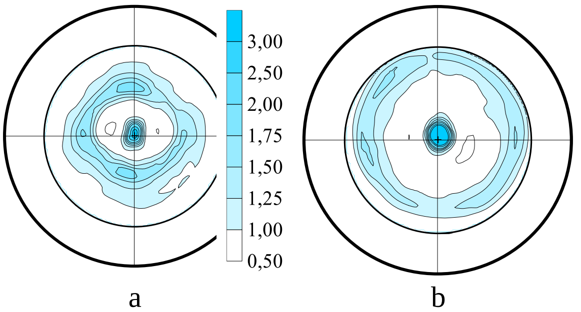


Figure 4. Partial DPFs {110} of rods, which were rotary forged at temperatures 350 (a) and 450 °C (b)

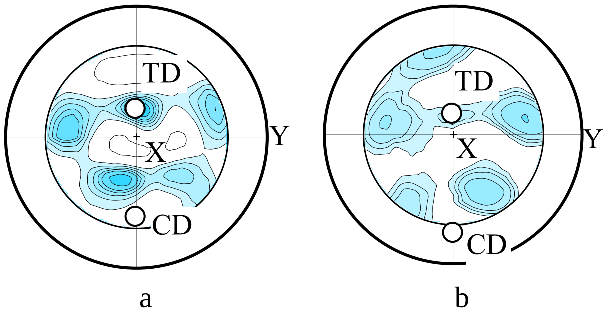


Figure 5. Partial DPFs of the TN-1 rods subjected to deformation according to various regimes: ECAP-20 at 450 °C (a) and ECAP-3 at 400 °C (b)

The partial DPFs {110} for different ECAP modes are shown in Figure 5. The outer axes of the rod after ECAP are denoted. The X axis is directed along the axis of the rod after deformation and displaced in the center of the stereographic projection. The Z axis is displaced at 120° relative to X-axis and characterizes the direction of movement of the rod at the beginning of the passage. The Y-axis is perpendicular to the plane of the intersecting channels directed along axes X and Z. The Z axis isn't shown in Figure 5. Earlier [17-18], it was shown that the ECAP texture can be represented as a rolling texture rotated about a transverse direction Y by some angle. The angle of rotation is determined by the angle of the channel intersection. The maximum shear plastic deformation develops in the rod when it passes the curved part of the channel. It is in this region that the rotation of the PPFs occurs, due to the reorientation of grains in accordance with the crystallographic mechanisms of plastic deformation acting there and the stressed state of the rod, given by the shape of the channels.

Let us consider deformation of TN-1 rod at the temperature 450 °C. As a result of 20 passes of ECAP, a clear crystallographic texture forms. It was shown in [18-19] that at each successive stage, a finite deformation texture that is characteristic of a stress state realized for this final stage can be formed, provided that the crystallographic mechanisms inherent in the material under consideration are not suppressed. This is due to the fact that the deformation at each stage exceeds 1. For forging at a temperature 450 °C, it is shown that crystallographic slip {110}<001> is activated, then in the case of rolling the ordered crystals with a CsCl-type lattice a {111}<110> texture is formed [16]. In accordance with the obtained results (Figure 5), it is this texture component that is rotated by an angle of about 100 ° relative to the first channel of the mold or Z-axis around Y-axis. The location of the TD and CD directions for the tool under consideration is shown in Figure 5.

In the case of ECAP at a temperature of 400 °C, the rolling texture {111}<110> still rotates and the increased scattering of texture is observed. However, the angle of rotation increases, which is due to a change in the coefficient of friction of the rod with the mold when the temperature changes. The observed scattering of the crystallographic texture can be associated with a change in the mechanisms of plastic deformation, which was also observed in the case of warm forging, as well as a result of grain refinement and activation of grain boundary slip. With the increase in the number of passes of ECAP, the texture maximum in PPF {110} approaches to the X axis.

Thus, the rods obtained by the warm rotary forging and ECAP in different modes are characterized by different crystallographic texture, the PDA also affects the final texture. Taking into account that the shape memory effect (SME) is orientationally dependent, the differences in the texture of the studied samples can affect the SME value.

4. Summary

1. The realization of the best combination of functional properties of Ti-Ni SMA may be explained by the peculiarities of mixed nanograined/nanosubgrained structure which forms as a result of annealing after severe plastic deformation. The increase in the yield stress due to the grain refinement down to a critical size of 40-80 nm in thin objects is comparable to the growth of the dislocation yield stress in bulk billets with a mixed nanograined/nanosubgrained structure with a high dislocation density after ECAP in the quasi-continuous mode and prevents plastic deformation by dislocation slip during deformation inducing SME. Besides, a positive effect of the post-deformation annealing is expressed in elimination of excessive work hardening and decrease of the transformation yield stress, which increases the completely recoverable strain.

2. It was found that when the temperature of the warm rotary forging of the TN-1 alloy is reduced from 450 to 350-400 °C, along with slip systems {110} <001>, operating at 450 °C, additional plastic deformation systems {100} <001> are activated.

3. In the case of ECAP-20 with intermediate heating up to 450 °C, the same slip systems act as in the case of rotary forging, which determine the formation of a characteristic rotated rolling texture at ECAP. The reduction in the pressing temperature and the absence of intermediate heat between the ECAP passes contribute to scattering of the crystallographic texture while maintaining the predominant texture component inherent in the deformation of ECAP-20.

Acknowledgements

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