EAST EUROPEAN JOURNAL OF PHYSICS
East Eur. J. Phys. Vol.4 No.3 (2017) 36-42 DOI:10.26565/2312-4334-2017-3-05

PACS: 28.41.Qb, 81.40 Ef, 61.10.Nz

INFLUENCE OF COLD ROLLING TO THE TEXTURE PARAMETER OF PURE HAFNIUM AND ZIRCONIUM

G.P. Kovtun, K.V. Kovtun, D.G. Malykhin, T.S. Yurkova, T.Yu. Rudycheva
National scientific center "Kharkov Institute of Physics and Technologies"
1, Akademicheskaya st., Kharkov, Ukraine, 61108
E-mail: dmitr.malykhin@gmail.com
Received August 12, 2017

Results of X-ray investigations of dependence of the crystallographic texture of hafnium and zirconium on degrees of cold rolling are presented. To obtain numerical principles, the method of inverse pole figures (IPF) with calculation of the texture parameters of Kearns (TP) for a chosen direction has been used. Measuring was carried out in the plane of the plates, as well as in the other two directions relating to rolling. The trigonal diagram of TP changes with degrees of deformation was built. Two stages of TP changes with deformation degrees are revealed: rate of the TP changes at the initial stage and, in part, characteristics of the subsequent stage depend on the initial texture of the materials. The second stage is characterized either by achievement of a minimum of TP in the rolling direction of the plates, or by moderate and even slight changes of them. In an example of hafnium, it has established that the increased rate of initial changes in TP is exclusively associated with intensive twinning predominantly by the $\{10\bar{1}2\}\langle\bar{1}011\rangle$ system. Arguments for dominance of twinning in the texture formation in hafnium and zirconium on the subsequent stage of rolling deformation are given.
KEYWORDS: X-ray analysis, texture, inverse pole figures, texture parameter, cold rolling, hafnium, zirconium, twinning

ВПЛИВ ХОЛОДНОЇ ПРОКАТКИ НА ТЕКСТУРНИЙ ПАРАМЕТР ЧИСТОГО ГАФНІЮ ТА ЦИРКОНІЮ

Г.П. Ковтун, К.В. Ковтун, Д.Г. Малихін, Т.С. Юркова, Т.Ю. Рудичева
Національний науковий центр «Харківський фізико-технічний інститут»
Україна, 61108, м. Харків, вул. Академічна, 1

Наведено результати рентгенівських досліджень залежності кристалографічної текстури гафнію і цирконію від ступеня холодної деформації прокаткою. Для отримання численних закономірностей використано метод зворотних полюсних фігур (ЗПФ) з розрахунком текстурних параметрів Кернса (ТП). Зйомки проведено в площині пластин, а також в двох інших напрямках, пов'язаних з прокаткою. Побудовано тригональну діаграму змін ТП зі ступенем деформації. Виявлено дві стадії змін ТП зі ступенем деформації: темп змін ТП на першій стадії і, почасти, характеристики другої стадії залежать від початкової текстури матеріалів. Друга стадія характеризується досягненням мінімуму ТП, виміряних у напрямку прокатки пластин, або помірною чи слабкою їх зміною. На прикладі гафнію встановлено, що підвищений темп початкових змін ТП виключно пов'язаний з інтенсивним двійникуванням переважно за системою $\{10\bar{1}2\}\langle\bar{1}011\rangle$..Наведено аргументи на користь домінування двійникування у формуванні текстури гафнію і цирконію на другій стадії деформації прокаткою.
КЛЮЧОВІ СЛОВА: рентгеноструктурний аналіз, текстура, зворотні полюсні фігури, текстурний параметр, холодна прокатка, гафній, цирконій, двійникування

ВЛИЯНИЕ ХОЛОДНОЙ ПРОКАТКИ НА ТЕКСТУРНЫЙ ПАРАМЕТР ЧИСТОГО ГАФНИЯ И ЦИРКОНИЯ

Г.П. Ковтун, К.В. Ковтун, Д.Г. Малихін, Т.С. Юркова, Т.Ю. Рудичева
Национальный Научный центр «Харьковский физико-технический институт»
Украина, 61108, г. Харьков, ул. Академическая, 1

Приведены результаты рентгеновских исследований зависимости кристаллографической текстуры гафния и циркония от степени холодной деформации прокаткой. Для получения численных закономерностей использован метод обратных полюсных фигур (ОПФ) с расчётом текстурных параметров Кернса (ТП). Съёмки проведены в плоскости пластин, а также в двух других направлениях, связанных с прокаткой. Построена тригональная диаграмма изменений ТП со степенью деформации. Выявлено две стадии изменений ТП со степенью деформации: темп изменений ТП на начальной стадии и, отчасти, характеристики последующей стадии зависят от исходной текстуры материалов. Вторая стадия характеризуется достижением минимума ТП, измеренных в направлении прокатки пластин, либо умеренным или даже слабым их изменением. На примере гафния установлено, что повышенный темп начальных изменений ТП исключительно связан с интенсивным двойникованием преимущественно по системе $\{10\bar{1}2\}\langle\bar{1}011\rangle$. Приведены аргументы в пользу доминирования двойникования в формировании текстуры гафния и циркония на последующей стадии деформации прокаткой.
КЛЮЧЕВЫЕ СЛОВА: рентгеноструктурный анализ, текстура, обратные полюсные фигуры, текстурный параметр, холодная прокатка, гафний, цирконий, двойникование

Zirconium and hafnium are the basis of materials intended for use in the construction of nuclear reactors. Products made of such materials, especially from zirconium-niobium alloys, undergo a special thermo-mechanical treatment aimed to maximize their satisfactory performance. Their crystallographic texture is an important characteristic on which the mechanical and radiation properties of these materials depend, as well as concerning to other hcp metals. Obtaining skills to predict its development after a necessary cycle of treatments is of practical interest. Study of principles of its changes after elementary processing cycles and analysis of the structural mechanisms responsible for these changes is

of scientific interest.

The aim of these studies is to reveal principles of changes in the crystallographic texture of pure hafnium and zirconium in process of cold rolling deformation, starting from annealed state of their plates. An exceptional feature is the subject of research – the Kearns texture parameter (TP) [1], which characterizes the general directionality of the crystallographic axes "c" of grains in hcp metals towards the investigated direction of the sample.

The world experience of research of this kind is practically absent. At present, the X-ray method of direct pole figures (DPF) is widely used for crystallographic texture research. The advantages of this method is simplicity of texture representing – in the form of a spatial distribution of orientations of the "c" axes, and the disadvantages are the difficulties of obtaining them due to limitations in the orientation of the samples and other technical reasons. This greatly reduces accuracy in the event of subsequent data processing. In fact, the DPF is a qualitative result of such studies.

In contrast, the suggested approaches described below make it possible to determine with satisfactory accuracy the TP as a characteristic convenient for both quantitative analysis and revealing numerical principles. This aspect also has no precedents and is therefore of especial interest.

The second exceptional feature of the research is the determination of TP in three orthogonal directions associated with rolling of plates.

Results will obtained can give some information about laws of rolling texture developments in the materials, and about accompanying structural mechanisms of it.

TECHNIQUE AND MATERIALS

To determine the TPs of hafnium and zirconium plates, the X-ray analysis of their texture by inverse pole figures (IPF) is used [2-4]. In contrast to the DPF method, in this approach, a usual X-ray optical scheme with Bragg-Brentano focusing is used. In particular, the present investigations were carried out using an X-ray diffractometer DRON4-07 in the radiation of CuK_α. To eliminate the vertical divergence of the X-ray beam, the pair of Soller slits was used.

According to this method, for the selected measuring direction (*j*), the pole density values P_{ji} (i.e. $P_{j,(hkl)}$) are calculated – this is an analogue of the distribution of crystallographic orientations (*hkil*) in this direction. These quantities are proportional to the experimental values of the integral intensity I_{ji}. The corresponding coefficient (R_j), in turn, is proportional to the flux of radiation incident on the sample. In general, the values of P_{ji} may be determined according to the following formulas:

$$P_{ji} = \frac{1}{R_j}\frac{I_{ji}}{I_{0i}}; \quad R_j = \sum_i A_i \frac{I_{ji}}{I_{0i}}; \tag{1}$$

where I_{0i} is the standard set of integrated intensities of reflection from an ideally non-textured (crystallographically isotropic) sample of such material; A_i is the quote of own orientation space (of a grain) for the *i*-th reflection, is used as the statistical weight [4]. In fact, the pole density is normalized to unity and would equal to unity for such non-textured material. Values I_{0i} either may be measured in advance, if there is a non-textured sample, or calculated [5].

The TP may be determined on the basis of the calculations of P_{ji} (1) using the following formula:

$$f_j = \left\langle \cos^2 \alpha_i \right\rangle_j = \sum_i A_i P_{ji} \cos^2 \alpha_i; \tag{2}$$

where α_i (α_{ij}) is the angle between the "c" axes of crystallites and the normals (*hkil*) in their reflecting position, i.e. in the direction of measuring. The meaning of the TP can be explained by an elementary example: the TP of a grain is equal to 1, if its axis "c" is oriented in the measuring direction, – is equal to 0 if it is perpendicular to them, and takes intermediate values in other cases. The TP of the material is averaged over all the plurality of grains.

The method was tested at its application to the Zr-2.5% Nb alloy texture investigations. So, the refined intensities I_{0i} were obtained by averaging the results for the samples of this alloy, obtained in three projections [6]. After comparing the results with the calculated values, the final set of values was obtained, the error of each of them is estimated at a limit of 5%. The values of A_i were calculated mathematically. Their set may be applicable to titanium, hafnium, zirconium and its industrial alloys.

Texture measurements of hafnium and zirconium samples were carried out in the rolling plane (normal direction, – ND), in the cross-section (rolling direction, – RD), and in the longitudinal section of the plates (transverse direction, – TD).

As samples for three-dimensional studies, hafnium plates HFE-1 (of Ukrainian specification) 2 mm thick annealed at 850°C for 1 hour, with a residual zirconium content of less than 0.2 wt. %, and other impurities – less than 0.1 wt. % are used. For the studies, samples were obtained by subsequent rolling up to 5, 15 and 30%.

Plates of iodide zirconium (99.9 mass%) with a thickness of 5 mm are also investigated in “3D” after both annealing at 600°C for 1 hour and subsequent deformation by 6, 10, 15, 20, 30 and 50%. Measurements in RD and TD were carried out from the surfaces of the central cut of plates to eliminate the boundary effect of rolling.

Additionally, the TP parameter in the normal direction of hafnium plates 5 mm thick, of the same kind and annealing mode, deformed to 5, 10, 15, 20, 30 and 50%, is investigated. For this material, the designation “$Hf^{(2)}$” will be further used, as well as “$Hf^{(1)}$” for its previous analogue.

The investigated surfaces of the plates were preliminary processed by grinding and etching.

RESULTS

In Figure 1, the IPFs of the $Hf^{(1)}$ plates are shown in three measuring directions. The values of the pole density (0002), $(10\bar{1}0)$ and $(11\bar{2}0)$ are displayed.

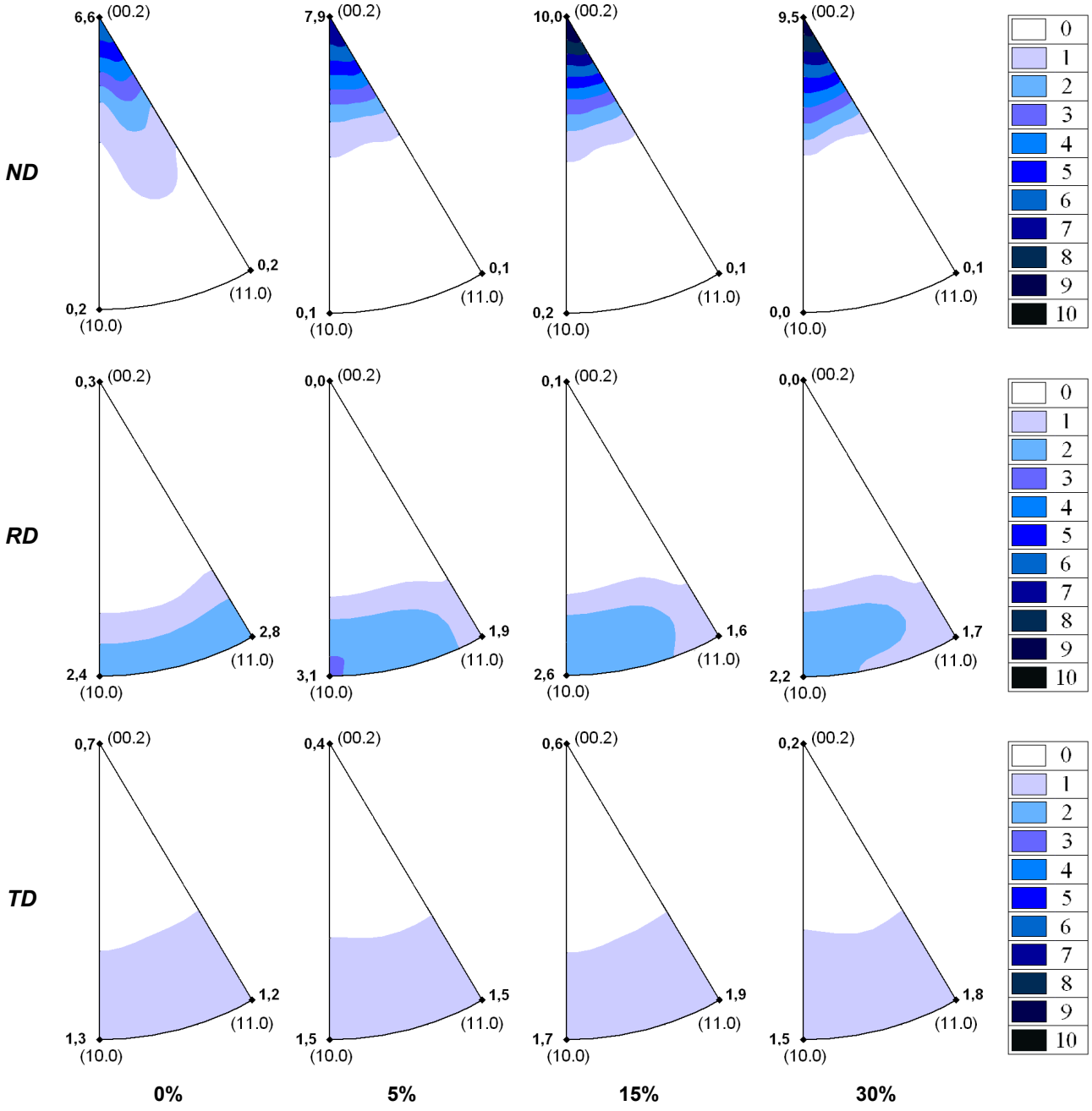

Fig.1. IPFs in three measuring directions for the original and deformed $Hf^{(1)}$ plates. The directions and the degree of deformation are indicated.

In Figure 2, with the same designations, IPFs of zirconium plates are given.

Based on the results of measurements from the plane of the initial and deformed $Hf^{(1)}$, $Hf^{(2)}$, and Zr plates, the graphs are given for the TP values in the ND direction (f_{ND}, Fig. 3) calculated by the formula (2).

The TP values determined for the three directions of the $Hf^{(1)}$ and Zr plates are displayed using a trigonal diagram (to be continued in the context). The diagram is built according to the principle of representing sections of ternary phase diagrams. The basis of this building is the trigonometric regularity:

$$\sum_j \cos^2 \alpha_{ji} = 1 \rightarrow \sum_j f_j = 1 \quad (j = 1, 2, 3). \tag{3}$$

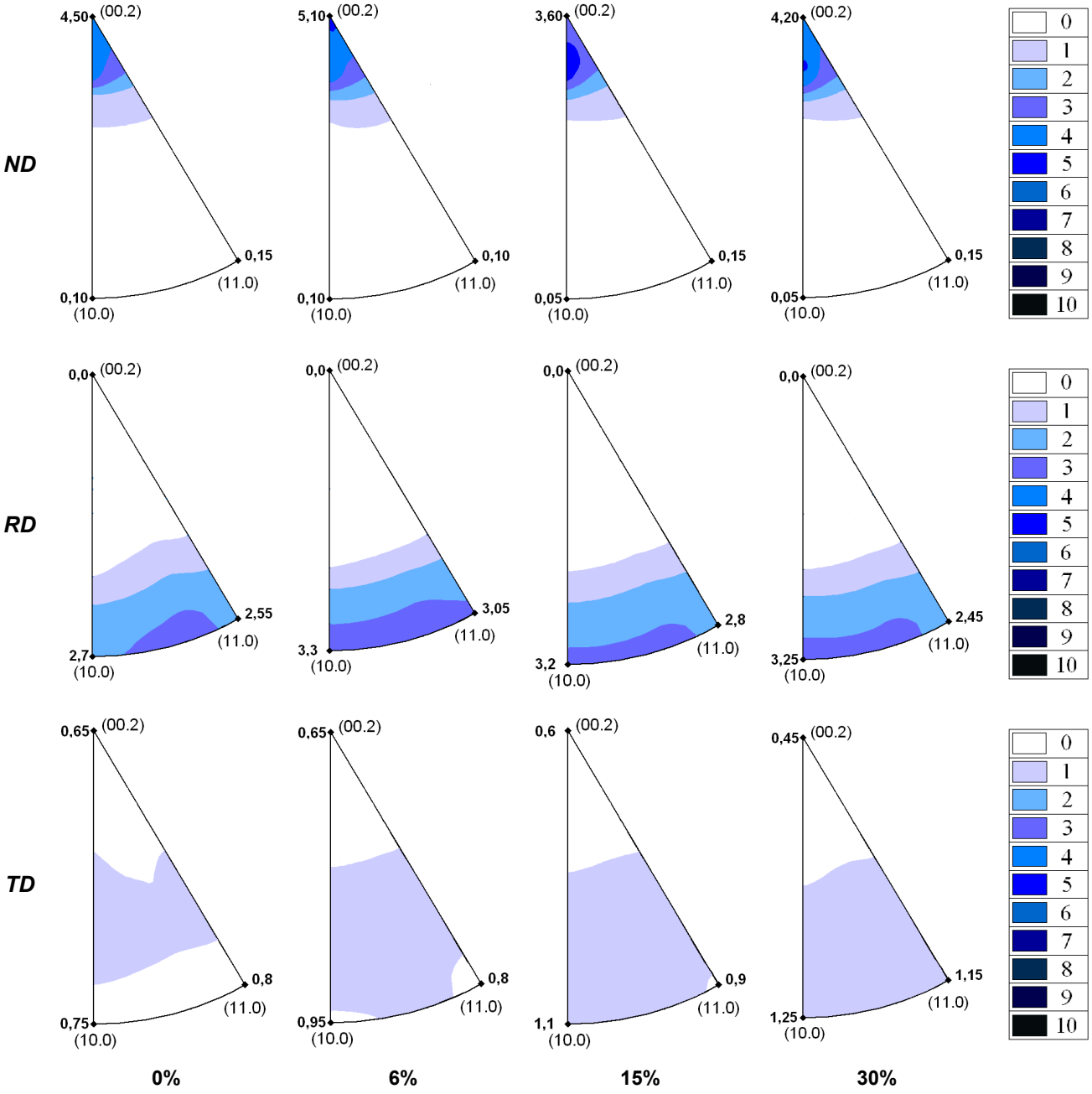

Fig.2. IPFs in three directions for the original and deformed plates of zirconium with indication the directions and the deformation degrees.

ANALYSIS OF THE RESULTS AND DISCUSSION

First of all, an ambiguity of the results obtained on the $Hf^{(1)}$ and $Hf^{(2)}$ samples (Fig. 3) attracts an attention. This circumstance, apparently, is due to differences in the making of the original plates.

A common principle of the graphs (Fig. 3) is moderate changes of the TPs for deformations of more than 5%. In the region below it and somewhat higher, the commonality of the graphs run is absent. The rate of the changes at up to 5%, as can be seen, depends on the initial values of the TPs. Partially, this also relates to changes at increased deformations, as can be seen in the example of the $Hf^{(1)}$.

Existence of such differences is confirmed by figure 4, where the arrows indicate the direction of the changes, starting from the initial state. So, on the one hand, it can be seen from the figure that the texture parameters of the original Zr plate fit in the direction of their further change, although at the same time, there is an increased initial rate. In part, this circumstance can also be noted for the $Hf^{(2)}$ (Fig. 3). Probably, the original plates of the $Hf^{(2)}$ and Zr were made by rolling, as well as subsequent samples, and at the same time – with achievement of high texture. On the other hand, for the $Hf^{(1)}$ (Fig. 4), the initial stage is more clearly detected, due to both the lower initial f_{ND} value (Fig. 3) and the increased TP in the RD (f_{RD}; Fig. 4).

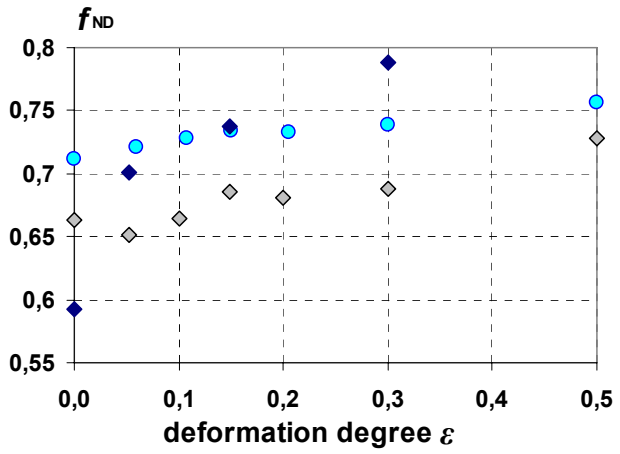

Fig.3. Changes of the f_{ND} with deformation degrees of the Hf$^{(1)}$ (◆) [7], Hf$^{(2)}$ (◆) and zirconium plates (○).

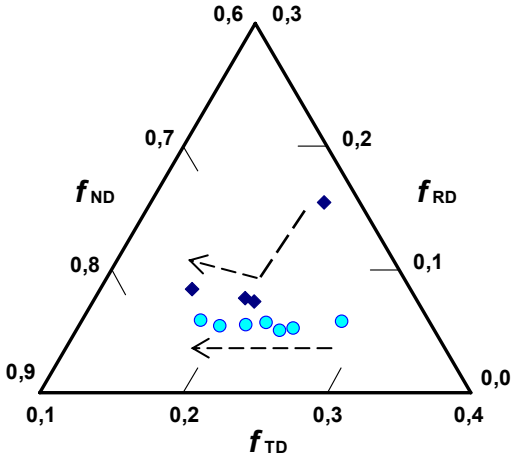

Fig.4. Diagram of the TPs changes for the Hf$^{(1)}$ (◆) [7] and zirconium plates (○) at the cold rolling.

In general, there are two principles in the obtained results. Firstly, in the changes of TPs with deformation degree of hafnium and zirconium, the stationary (moderate) stage exists. Its feature is small changes in f_{RD} (Fig. 4). In zirconium, for example, this parameter at such stage practically does not change, taking on value 0.06. Secondly, in the changes of TPs, at depending on their initial values, the initial stage as a rule can exist, differing by an accelerated run and a differed direction on the texture diagram. Partly, it is observed on the plates of Zr and is especially obvious for the Hf$^{(1)}$ plates (Fig. 3.4), and even is noticeable for Hf$^{(2)}$ (Fig. 3).

To define the structural nature of changes of TPs, first of all, their evaluation was carried out from the standpoint of homogeneous deformation at the scales of grains and higher. It was understood that such deformation is provided by the dislocation slip mechanisms. It was assumed that the contribution of slip to the formation of a typical rolling texture, if this could be, can be due to the binding of both the basal planes (0002) of the crystallites and their "c" normals – to the geometry of such kind deformation. According to this principle, during deformation, these planes are permanently approaching to the rolling plane by their orientation. In this calculation, changes of the normals "c" incline are taken: $\alpha_{ij} \rightarrow \alpha'_{ij}$. The texture features of the initial and deformed plates allow us to use for estimation such formula: $\tan\alpha'_{ij} = k\tan\alpha_{ij}$, – where k depends on the deformation degree ε and takes the value $(1 - \varepsilon)^n$. The value n is equal to 2 for the longitudinal section of the plates (RD↔ND), and to unit for the cross section (TD↔ND) [7].

According to this scheme, the evaluation was carried out on a sequence of values of the pole density $(h0\bar{h}l)$, aligned along the $\cos^2\alpha$ scale. In this regard, in Fig. 5, the deformation dynamics of the experimental sequence of values $P_i(\cos^2\alpha_i)$ and calculated one – $P_i(\cos^2\alpha'_i)$ – are given. The values of $P_i(\cos^2\alpha'_i)$ were calculated on the basis of the data for the initial sample.

As can be seen from figure 5, the experimental and calculated data significantly differ. In view of this, it should be assumed that at the rolling process of the plates, dislocation slipping in the grain body is distributed irregularly. In such cases one considers that it develops a tendency to localize in the grain boundary region. Such a state, apparently, has little effect on the changes in the crystallographic orientations of grains. The reason of the texture changes in this case remains to be attributed to twinning. Data from other studies confirm formation of twins in zirconium during rolling [8].

From what has been said, in particular, it follows that in the initial changes in the Hf$^{(1)}$ texture, which are the most evident in comparison with other ones (Fig. 3, 4), preference should also be attributed to twinning.

For a detailed explanation of this, a calculation of the $f_{RD}(hki0)$ values, characterizing the directionality of normals to the prismatic planes along the RD (instead of the "c" axes, as is accepted for TP), has been carried out. Practically it has taken in account the directions within 30º around the RD. An analogue of formula (2) has been used with the replacement $\cos^2\alpha_{ij} \rightarrow \sin^2\alpha_{ij}$:

$$f_{RD}(hki0) = \sum_i A'_i P_i \sin^2 \alpha_i , \tag{4}$$

where A'_i is the statistical weight of the poles, additionally bounded by the such angular limits and so equal to either A_i, or zero, or intermediate values. All values refer to the RD.

The meaning of this task is to analyze the juxtaposition of quotes of prismatic – $(hki0)$ – and basal (axis "c") orientations along RD, and their changes during deformation.

In this regard, in Figure 6 the graph of correlation of f_{RD} with $f_{RD}(hki0)$ for all deformation degrees of the Hf$^{(1)}$ plates is presented. As can be seen, the graph obviously represents a linear correlation of these values. Significantly, its course is extrapolated to coordinates (1; 0). So, if $f_{RD}(hki0)$ could reached its maximum, i.e. unit, then the parameter f_{RD} (i.e. $f_{RD}(0002)$) according to formula (3) would vanish.

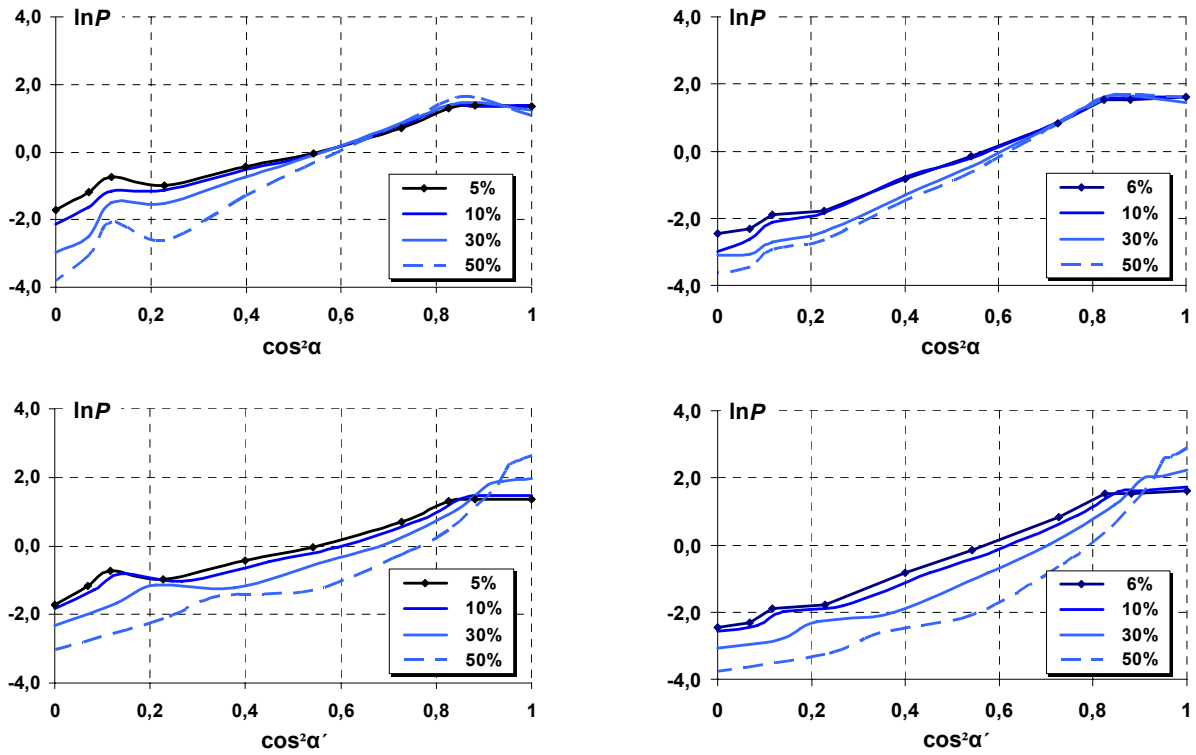

Fig. 5. Graphs of changes in the experimental (upper figures) and calculated sequences of values of the pole density (lower figures) measured in normal direction of the deformed Hf(2) (left) and Zr samples (right-hand).

It follows from this that the initial rotation of the "c" axes from the RD towards the ND (and a slight reverse for deformations above 5%) is done with immediate exchanges (0002) ↔ (*hki*0). In other words, the turn acts are made practically on 90°. It is meant, intermediate orientations do not participate in this. By the way, they are practically absent (Fig.1, RD).

This shows that the changes of the TPs at the initial stage of deformation of the Hf(1) are carried out exclusively by intensive twinning, and predominantly by the $\{10\bar{1}2\}\langle\bar{1}011\rangle$ system, which rotates the axes by 85° [9.10].

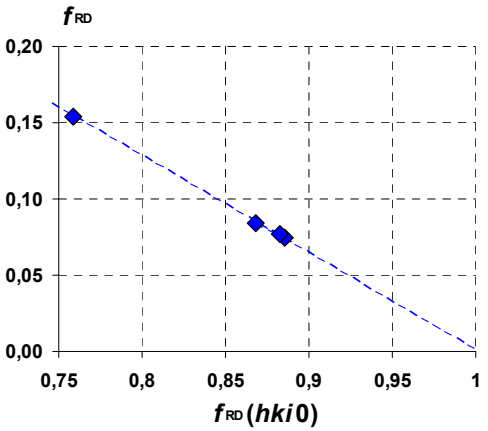

Fig.6. Correlation between f_{RD} and $f_{RD}(hki0)$ parameters at all the deformation degrees for the Hf(1).

For the Hf(2) and zirconium, the conditions for this are insufficient. Evidently, such a process in these materials could end at the stage of making the original plates. An initial stage of TPs changes in pre-annealed zirconium plates can be expected if the initial f_{RD} parameter will noticeably exceed a value of 0.06.

The results obtained are in general consistent with existing concepts, according to which twinning exhibits high activity at deformation of hcp metals [11], significantly affects their texture [10-12] and plays an exclusive role in texture formation in zirconium alloys [13]. Moreover, it does a significant contribution to the mechanical properties of hcp metals [14].

CONCLUSIONS

By the method of inverse pole figures, X-ray studies of dependence of the crystallographic texture of hafnium and zirconium plates, in particular, the texture parameter of Kearns (TP) on degree of cold deformation by rolling are carried out. Measurements are done in the rolling plane of plates and in two other orthogonal directions.

In the changes of TPs with the degree of deformation of hafnium and zirconium, the presence of two stages has been revealed. Unlike the initial one, the subsequent stage is characterized by a moderate rate of change of TPs, in particular, by small changes in the rolling direction.

The level and rate of changes of the TPs at the initial and, in part, the characteristics of the subsequent stage depends on the initial state of the material, in this case on degree of difference of the initial TPs from their expected values in the subsequent stage.

Using the example of hafnium, which is characterized by a relatively high value of the initial TP in the rolling

direction, it is found that the increased rate of initial changes of this parameter is exclusively associated with intensive twinning, predominantly of its $\{10\bar{1}2\}\langle\bar{1}011\rangle$ system. The arguments for dominance of twinning in formation of the texture of hafnium and zirconium in the subsequent stage are given.

REFERENCES

1. Kearns J. Thermal expansion and preferred orientation in Zircaloy // USAEC WAPD-TM-472. Nov. 1965.
2. Ageyev N.V., Babareko A.A., Betsofen S.Ya. Method of inverse pole figures // Izvestiya AN SSSR. Ser.: Metals. – 1974. – No. 1. – P. 94-103.
3. Harris. G.B. Quantitative measurement of preferred orientation in rolled uranium bars // Phil. Mag. – 1952. – Vol. 43. – No. 336. – P. 113-123.
4. Morris P.R. Reducing the effects of non-uniform pole distribution in inverse pole figure studies // J. Appl. Physics. – 1959. – Vol. 30. – No. 4. – P. 595 -596.
5. Mirkin L.I. Reference book on X-ray analysis of policrystals. Red. prof. Umanskij. Moscow. State ed. phys.-mat. liter. – 1961. – 864 P.
6. Grytsyna V., Stukalov A., Chernyayeva T. et al. Destruction of crystallographic texture in zirconium alloy tubes // Fourteenth International Symposium: Zirconium in Nuclear Industry. Stockholm, Sweden, June 13-17, 2005. ASTM Stock. Number: STP1467. Bridgeport, NJ. – 2006. – P. 305-329.
7. Malykhin D.G., Kovtun G.P., Potina T.S. Features of texture, microhardness and deformation mechanisms in cold-rolled hafnium // Voprosy atomnoi nauki i tehniki. Ser.: «Vacuum, pure metals, superconductors (21)». – 2016. – No.1(101). – P. 35-40.
8. Luan B.-F., Ye Q., Chen J.-W. et al. Deformation twinning and textural evolution of pure zirconium during rolling at low temperature // Trans. Nonferr. Met. Soc. China. – 2013. – No. 23. – P. 2890-2895.
9. Chernyayeva T.P., Grytsyna V.M. Characteristics of HCP metals defining its behavior at mechanical, thermal and radiation treatment // Voprosy atomnoi nauki i tehniki. Ser.: «Physics of radiation damages and radiation material science» (92). – 2008. – No. 2. – P. 15-27.
10. E. Tenckhoff. Review of deformation mechanisms, texture and mechanical anisotropy in zirconium and zirconium base alloys // Proceedings of XVI International Symposium «Zirconium in Nuclear Industry». West Conshohocken, ASTM STP1467. – 2006. – P. 25-50.
11. Vishnyakov Ya.D., Babareko A.A., Vladimirov S.A. et. al. Theory of formation of textures in metals and alloys. Moscow.: Nauka, 1979. – 345p.
12. Calnan E.A., Clews C.J.B. // Phil. Mag. – 1952. – Vol. 42A. – No. 328. – P. 919-931.
13. Tome C.N., Lebensohn R.A., Kocks U.F. A model for texture development dominated by deformation twinning: application to zirconium alloys // Acta Metall Mater. – 1991. – Vol. 39. – No. 11. – P. 2667-2680.
14. Tome C.N., Agnew S.R., Blumenthal W.R. et al. The relation between texture, twinning and mechanical properties in hexagonal aggregates // Materials Science Forum. – 2002. – Vol. 408-412. – P. 263-268.