

Effect of interlayer macrostress on hydride orientation in shell tubes from Zr-based alloys

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Abstract. Methods of texture analysis allowed to show that inhomogeneity of the crystallographic texture in shell tube wall is responsible for rise of tangential macrostress in the tube. This inhomogeneity predetermines a difference of thermal expansion between outside and inside layers of tube at its exploitation temperature. The indicator of such stress is formation of radially-oriented hydrides in the transverse section of tube, subjected to treatment in autoclave.

1. Introduction

Shell tubes from Zr-based alloys are responsible construction elements of atomic reactors. During exploitation these tubes undergo to oxidation and hydrogen saturation, which can become a reason for their fracture. Crystallographic texture is among the factors that have a very significant and sometimes decisive influence on the corrosion behavior of zirconium alloy products. In particular, the unfavorable hydride orientation in shell tubes from Zr-based alloys under operating conditions of the reactor dramatically reduces their service life. When considering the influence of texture on the hydride orientation in tube wall, one should take into account, that in most cases this effect is detected indirectly, in particular - by initiating a tensile stress due to non-uniformity of texture. At the same time, crystallographic texture is the primordial cause responsible for that or another parameters of corrosion process. In this paper, authors are considering the mechanisms of texture layering in tubes from Zr-based alloy on its corrosion and hydride orientation, irrelative of chemical or phase composition of these alloys.

2. Studied samples and methods of investigation

Studied samples were cut out from several series of shell tubes, submitted to regular technological treatment. These samples were measured by X-ray texture method of direct pole figures (PF) [1]. At that electro-erosion cutting of tubes into separate segments and assembling of these segments into a square sample were used. For composite samples $\text{PF}(0001)_{\alpha\text{-Zr}}$ were constructed and integral Kearns parameters were calculated. The similar fragments of the same tubes were saturated with hydrogen in the vapor-water atmosphere within an autoclave during 42 hours at 350°C for. Then cross-sections of tubes were studied using optical microscopy in order to reveal hydride particles [2].

3. Experimental results and discussion

Application of precise contemporary X-ray methods by study of shell tubes from Zr-based alloys permitted to reveal a number of structure features, able to quicken diffusion of oxygen and hydrogen into the matrix. First of all this relates to inhomogeneity of the crystallographic texture through the tube wall, arising due to different values of Q-factor, calculated for layers, which adjoin to inside and outside surfaces of tube. It is equal to the ratio of reduction by tube wall $\Delta t/t$ to reduction by its diameter $\Delta D/D$, where t – thickness of wall and D – diameter of tube. As Q-factor increases, texture maxima in $\text{PF}(0001)$ shift to the center of stereographic projection, moving away from the transversal tube direction T to the tube radial direction R . Q-factor is the major technological parameter of the tube rolling and



determines a stress state in tube wall during its deformation. Therefore, Q-factor influences on the crystallographic texture, forming at these conditions.

Usually Q-factor is calculated, when starting from the thickness of whole tube wall. However if we distinguish in the tube at least two layers and determine for them values of Q-factor by assumption, that deformation through these layers is homogeneous, we would see that obtained values of Q-factor inevitably differ from each other. Though at the first approximation textures of these layers are visually similar, their corresponding integral Kearns parameters f_R , f_T and f_L , used for quantitative estimation of texture in tubes from Zr-based alloys and calculated as projections of the basal axes distribution on radial, tangential and axial directions of tube [3], prove to be somewhat different. Influence of this effect on hydride orientation in shell tube wall is considered lower.

Results of tube texture analysis are presented in Fig. 1. At the left part of Fig. 1 a fragment of shell tube and its flat sections, studied by X-ray texture measurements, are depicted, whereas at its right part – results of measurements. From above the partial PF(0001) (with angular radius 80°) for the outside layer of tube with calculated Kearns parameters are shown, while from below – the same for the inside layer of tube. Further equatorial sections R-T of PF(0001) for both layers are constructed (Fig 1-c). The difference of Kearns parameters for studied layers corresponds to the limiting sensitivity of the method for their determination [3], but the following example in Fig. 2 visually shows that this difference is sufficiently big for rise of essential tangential macrostress.

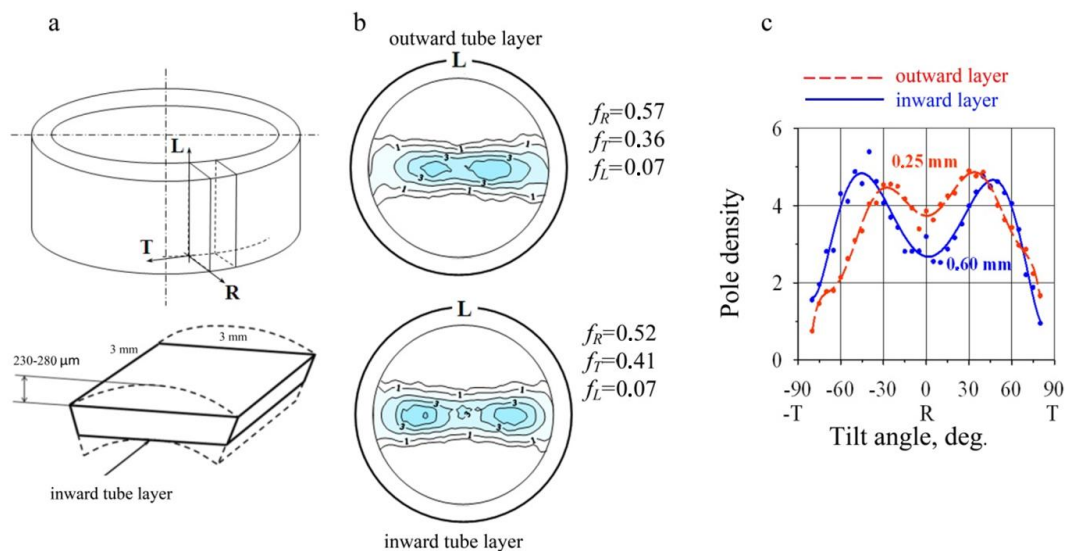


Figure 1. Scheme of cutting and preparing studied inside and outside surfaces of tube (a), PF (0001) of different tube layers (b) and distributions of basal normals in R-T section (c)

Coefficient of thermal expansion for any direction in tube, for example – for tangential direction T, is connected with texture Kearns parameter f_T by the following way:

$$\alpha_T = \alpha_c f_T + \alpha_a (1 - f_T),$$

where α_c and α_a – coefficients of thermal expansion for α -Zr single crystal along axes c and a in its crystalline lattice. At that, Kearns parameter is calculated namely for the direction T. Mean values of coefficients α_c и α_a at 350°C , found for a number of literature data [4], are equal to 5.5 deg^{-1} and 8.8 deg^{-1} , respectively.

Differences in thermal expansion of outside and inside layers along the tangential direction of tube at its exploitation temperature is a reason for rise in it of tangential macrostress.

When the coefficient of thermal expansion α_T of inside layer exceeds the analogous coefficient of outside layer, tangential tensile stresses operate at the outside layer of tube.

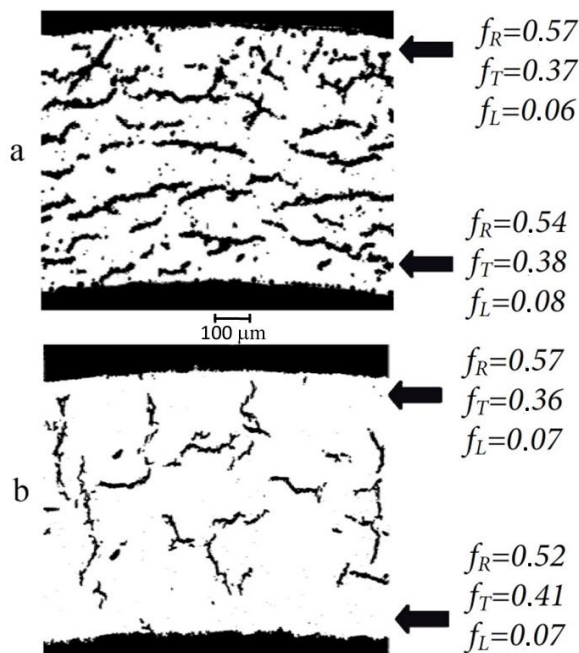


Figure 2. Radially-oriented hydrides in the cross-section of tubes, submitted to heating up to 350°C in a autoclave

A reliable indicator of such stresses is rise of radially-oriented hydrides in the cross-section of tube, submitted to keeping in autoclave (Fig. 2). By heating of tube up to 350°C solubility of hydrogen in α -Zr increases more than by 10 times, and by next cooling this hydrogen enters in the composition of hydride phase, precipitating in tube in accordance with direction of operating macrostresses, when taking into account, that precipitation of hydrides is a mechanism of these macrostresses relaxation.

Cross-sections of two shell tubes, presented in Fig. 2, differ in arrangement of hydride phase precipitations. At that, in the first tube hydrides are situated approximately along tangential direction, while in the second tube – along radial direction. It is known, that Hydride precipitates in α -Zr matrix have habit planes of numerous types, including basal planes and grain boundaries. However, by presence of tensile stresses they prove to be the strongest factor among those, determining orientations of hydride precipitates, which line up by perpendicular to operating stress. Namely because of this circumstance hydride

precipitates reorient in the course of delayed hydride cracking in the field of stresses. Thus, the distribution of hydrides in Fig. 2-a responds to their arrangement by habit planes, whereas the hydride distribution in Fig. 2-b confirms existence of tangential macrostress in the second tube before precipitation in it of the hydride phase. This macrostress promotes penetration of oxygen inside the tube wall.

It is seen in Fig. 2-b, that under the influence of tangential macrostresses lamellar hydride particles take up radial positions, perpendicular to outside surface of tube, and coincide apparently with grain boundaries, having analogous orientations and being ways of active hydrogen diffusion under operation of tangential macrostresses. On the contrary, in the first tube (Fig. 2-a) in the presence of tangentially-oriented hydrides penetration of hydrogen inside the tube wall is difficult because of absence of tangential stresses as well as transparent radial ways of hydrogen diffusion, generated by these stresses.

Calculated deformations of shell tubes in the tangential direction as a result of their heating up to 350°C are presented in Table 1 in order to illustrate, how insignificant can be layer-by-layer texture inhomogeneity in tube, causing nevertheless a rise of essential macrostresses, responsible for precipitation of radially-oriented hydrides and promoting diffusion of oxygen inside the tube wall. Here ε_T – strain relative tangential deformation of the corresponding tube layer, and $\varepsilon_T^{\text{in}} / \varepsilon_T^{\text{out}}$ – the ratio of these values for inside and outside tube layers.

Table 1. Thermal expansion of different layers of shell tube by its heating up to 350°C

Sample number	Depth of layer, t, μm	f_T	$\alpha_T 10^{-6}$, deg.	$\varepsilon_T, 10^{-3}$	$\varepsilon_T^{\text{in}}/\varepsilon_T^{\text{out}}$
1	250	0.37	6.721	2.2179	1.005
	600	0.38	6.724	2.2288	
2	240	0.36	6.688	2.2070	1.025
	620	0.41	6.853	2.2615	

Comparison of deformation textures for inside and outside layers of tubes testifies, that in the second tube the relative tangential deformation of inside layer by $\Delta f_T = 0.05$ already exceeds the deformation, responding to the yield stress. When taking into account values $\varepsilon_T^{\text{in}}/\varepsilon_T^{\text{out}}$, presented in the last column, it becomes understandable, that by control of production the first tube was accepted as satisfying to technical conditions, whereas the second tube was rejected as spoilt.

4. Summary

As obtained experimental data show, the layer-by-layer texture gradient is sufficient for noticeable variations of dimensional thermal changes in successive layers. In the case, when the coefficient of thermal expansion for the inside layer exceeds that for the outside layer, tangential tensile stresses operate at the outside tube surface. As a result, intensification of corrosion processes takes place, including diffusion of oxygen and hydrogen.

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