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PROCESSES OF FORMING CAST STRUCTURES IN A PIPE BLANK MADE OF ZIRCONIUM-NIOBIUM ALLOY

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The presented material is part of a comprehensive study on the manufacture of fuel rod cladding tubes in Ukraine. Since the production of tubes relies on high-quality tube blanks, this study examines blank manufacturing processes and determines rational ingot casting methods. The established patterns of structure formation and the proposed scheme of niobium redistribution in the zirconium alloy Zr1Nb.

INTRODUCTION

Global experience in the production of ingots from zirconium and its alloys is mainly based on the use of vacuum arc melting with a consumable electrode in a water-cooled copper crucible, followed by double vacuum arc remelting. The main problem with this smelting method is the preparation of the consumable electrode and the introduction of alloying additives. The electrode is produced by various methods, e.g., pressing briquettes followed by sintering or welding [1].

To date, the traditional method of obtaining pipe billets from zirconium alloys includes double vacuum arc remelting, obtaining large-sized ingots (350…650 mm in diameter), and forging. This scheme is used by most countries with a nuclear cycle: the USA, Japan, etc. In principle, metal produced by vacuum arc remelting has high quality; however, it has several drawbacks, in particular, it is not always possible to eliminate porosity in ingots during the first remelting (VDP ingots). Relatively low cooling rates during crystallization form a structure of columnar crystals with a coarse lamellar internal grain structure and local manifestations of coarse α-phase at the boundaries of the grains of the initial β-phase in VDP ingots, which significantly complicates their further plastic deformation [2, 3].

The zirconium-niobium alloy smelted in Ukraine differs due to the specific technology used to obtain it (calcium thermal reduction process, electron beam remelting) and, therefore, has differences in chemical composition, uniformity in the distribution of alloying elements, and structure [4].

The study evaluated the influence of these factors on deformed metal during pipe rolling and examined the effect of new proposed production processes on the quality indicators of pipe shells and their mechanical properties at room and elevated temperatures, corrosion resistance, creep, low-cycle fatigue, material texture, structure, including fine structure [5].

Previously, there was no production of zirconium alloy pipes in Ukraine, so based on a review of global experience in this field, a technology was proposed that includes new technological solutions in metal smelting, hot and cold deformation of pipes, and their processing [6, 7].

EXPERIMENTAL DETAILS

The study was performed on a cast pipe blank made of zirconium alloy with 1% niobium after electron beam melting and electromagnetic stirring. The initial diameter of the ingot was 180…200 mm. It should be noted that for the first time in zirconium alloy processing, pipe billets were manufactured using centrifugal casting, from which fuel rod cladding tubes were also manufactured.

Since the further technological processing of the cast blank is hot deformation by pressing, it was of interest to determine the processes of forming cast structures and the distribution of chemical elements, in particular niobium, in ingots and their influence on hot deformation processes. A new method of hot deformation without forging the blank by pressing with high degrees of deformation in the β-region is proposed.

RESULTS AND DISCUSSION

It is known that in very pure zirconium, the β→α transformation occurs by diffusion with the formation of equilibrium α-phase grains [8]. The variety of structures is explained not only by the smelting conditions, but also by specific mechanisms that occur during the decomposition of the solid solution (β-phase). Even small amounts of oxygen impurities (up to 0.07%) cause the appearance of needle-like structures in rapidly cooled zirconium. It is well established that zirconium obtained by industrial methods (including calcium thermal technology) contains a significant amount of interstitial impurities (oxygen, carbon, nitrogen), which control the β→α transformation in structural terms, and this effect is enhanced in Zr-Nb alloys. Impurities, in particular oxygen, shift the lines on the β→α diagrams (Fig. 1) [9].

During cooling, a needle-like (rail-like, plate-like) structure develops structure is formed in the Zr1Nb alloy, with series of plates forming groups or packets in which the transformation occurs according to one of the variants of the β→α transition orientation ratio [9]. At the same time, the first zirconium plates formed have a low niobium concentration, as can be seen from the equilibrium diagram for an alloy with 1% niobium.

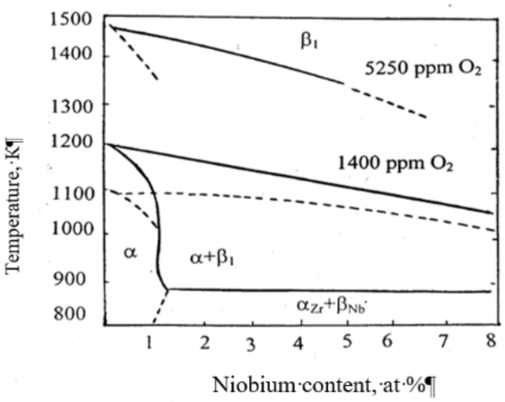


Fig. 1. Position of boundaries on the Zr-Nb phase diagram at different oxygen contents [9]

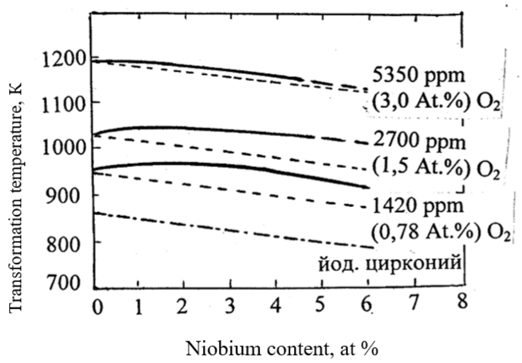


Fig. 2. Measured and calculated values of the transformation temperature for Zr-Nb alloys with different oxygen contents – measurement; calculation [9]

Therefore, at the β→α transformation front in the α phase, the concentration of niobium will increase, since some of its atoms will be removed by the drift mechanism from the transformation front in the tangential direction, as is the case with carbon during bainite (or pearlite) transformation (Fig. 2). As a result of this process, the concentration of niobium on the side surface of the α plate increases to the formation of the β-Nb phase, and its embryo grows in the direction of maximum niobium concentration, i.e., along the α-Zr plates. The final structure after such a repeatedly repeated process consists of wide α-Zr plates and narrow β-Nb layers alternating between them (Fig. 3).

Fig. 3. Microstructure of Zr1Nb alloy ingots after electron beam remelting, x500

Considering that the β-Nb phase contains 85% Nb and 15% Zr, then with 1% Nb in the alloy, the ratio should be approximately 1:50. In reality, according to estimates based on light microscope images, this value ranges from 1:10 to 1:30. This discrepancy is explained by the fact that the niobium layers are not continuous, but form a system of ribbons in one plane between two α-Zr plates in the form of a branched flat dendrite. The "transparency" of such a branch compensates for the deficit (lack) of the niobium phase in the layered structure, maintaining the average concentration of niobium in the alloy in the package (Fig. 4).

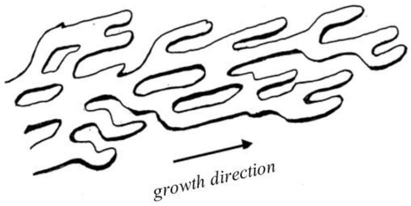
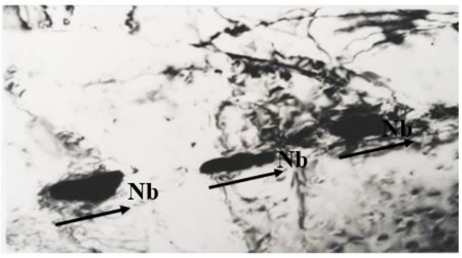


Fig. 4. Reconstruction of the structure of β-Nb phase layers in the package

The morphology of niobium layers is very diverse: from solid plates with a thickness of several microns (0.01...0.1 μm) in width and several tens or even hundreds of microns in length, to narrow strips (lines) or rods (lines of dots), visible in Fig. 5.



x20000

Fig. 5. Intermittent distribution of niobium layers in cast metal (electron microscopy)

Zirconium alloys are characterized by the presence of 4 to 12 systems of unidirectional mutually parallel plates (packages) in a single former β-crystal, the boundaries between which are almost straight lines. This morphology is characteristic of bainitic and martensitic structures, the origin of which is associated with the shear transition of one crystal lattice into another (Fig. 6).

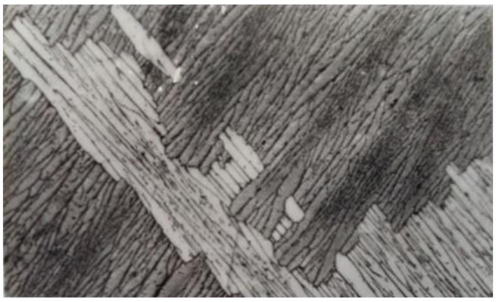


Fig. 6. Frequency distribution of α-Zr plate width in a package, x500

It can be hypothesized that in Zr1Nb alloys, the decay of the β-phase begins with the formation of an α-Zr plate by a shear mechanism, the growth rate of which is limited by the rate of diffusion removal of niobium atoms by a drift mechanism, accompanied by enrichment (creation of a concentration jump) with niobium in the boundary layer, where, upon reaching the required concentration, a β-niobium interlayer is formed, which is a drain for niobium atoms both for the already formed α-Zr plate and for the adjacent β-zirconium layer, the depletion of which in niobium facilitates the nucleation and growth (by the slip mechanism) of a second α-Zr plate, parallel to the first and having the same crystallographic orientation. The process is then repeated and the plate stack grows in both the longitudinal and transverse directions.

Thus, during the decomposition of the zirconium phase in Zr1Nb alloys during slow cooling, processes characteristic of both shear (during transition) and diffusion transformation (for the transition β Zr→α Zr + β Nb) occur. Hence, the reaction can be classified as bainitic rather than quasi-monotectoid, which is characteristic of Zr1Nb alloys with higher niobium concentrations.

In ingots obtained by EPP, the most interesting phenomenon is the periodicity of interlamellar spacing within a single package. Studies have shown that interlamellar distances in packages are not always regular and uniform. There are packages in which wide and narrow plates are located in close proximity to each other, and their location clearly shows a certain periodicity (see Fig. 6). This observation cannot be attributed to local variations in sectioning angle relative to the polished surface, since this would require a corresponding change in the habitus of the plates in the package, and observations show that the plates in one package maintain their direction quite strictly. The results indicate that the periodic variation in α-Zr plate widths within a single packet is a genuine spatial feature that develops during packet formation.

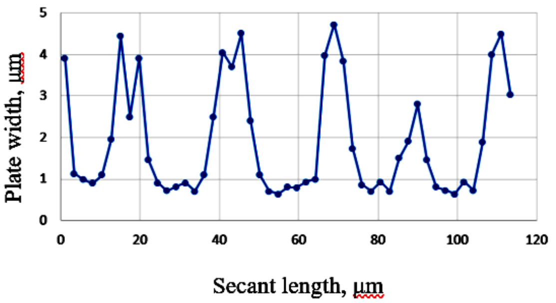


Fig. 7. Distribution of the width of the α-Zr plate in the cross section

The measurement results show that plates with a width of up to 5 µm regularly alternate with thin ones with a width of about 1 µm, with a period of approximately 20 µm (Fig. 7). This situation is more evident in Fig. 6, where the darker stripes are associated with areas of dispersed plates in the package. In addition, these zones continue on the other side of the fragment of another intersecting package.

Fig. 8 schematically shows the configuration of the β→α decay front of a solid solution in an alloy, and Fig. 9 shows the corresponding distribution of niobium concentrations in different zones of the front. It can be assumed that the surface of the front has a flat-wave configuration, where the wave crests are zones with the widest plates, and the troughs are zones with narrow plates.

The formation of a periodic structure in the package begins with the formation of a dome-shaped cross-section of the transformation front, with the leading edges of the thickened plates growing according to the shear mechanism located at the apex of the dome, and thinner ones on its slopes and in the depression.

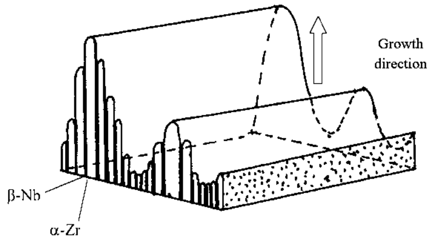


Fig. 8. Spatial configuration of the transformation front during the formation of a periodic structure in a packet

As the dome grows, niobium atoms are displaced from it by the mechanism of active diffusion and accumulate in the depression of the front, where the concentration of niobium increases and β-niobium plates nucleate more frequently (see Fig. 9).

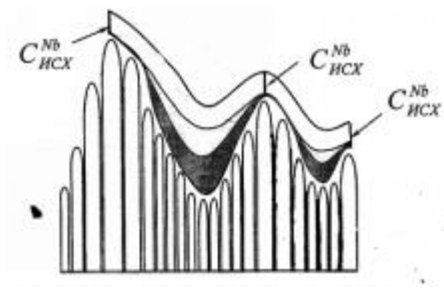


Fig. 9. Front configuration and corresponding distribution of niobium concentrations in different zones of the front

The existence of depressions and protrusions with a coordinated alternation in places where two packages with different plate habitats interact, as seen in Figs. 3 and 6, also supports this assumption. It can be seen that the widest plates, reaching the future junction with the first ones, penetrate deeper into the foreign package. That is why, if a foreign system of one to three to five plates of a different direction (package) is found in one of the packages, their width is usually much greater than that of the surrounding plates (see Figs. 3 and 6) [10].

Based on the proposed growth (structure formation) scheme, it can be assumed that zones with narrow plates have a higher specific concentration of niobium than zones with wide plates.

Thus, during the β→α transformation, a non-uniform distribution of niobium in the packets occurs. The size of the α-phase packets and their period have a decisive influence on the recrystallization processes during hot deformation.

The results of the studies characterizing the features of the formation of cast structures in the Zr1Nb alloy form the basis for recommendations for the manufacture of pipe billets.

CONCLUSIONS

1. Patterns of structure formation in the Zr1Nb alloy characteristic of new smelting methods have been

established: electron-beam melting with electromagnetic stirring.

2. A scheme for the redistribution of niobium in zirconium has been proposed, which explains the formation of striped structures in the Zr1Nb alloy packets during β→α transformation. The morphology of β-niobium layers in cast structures has been shown.

3. The influence of the smelting method on the size of α-phase packets, their periodicity, and their subsequent decisive influence on recrystallization processes during hot deformation has been established. A decrease in the width of α-phase plates during smelting increases the completeness of recrystallization of the Zr1Nb alloy during hot deformation processes.

4. It has been established that the optimal methods for smelting Zr1Nb alloy pipe billets in terms of forming a homogeneous macrostructure and mechanical properties that ensure sufficient technological plasticity of the metal during subsequent hot deformation, are vacuum-arc and electron-beam skull melting with electromagnetic stirring.

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ПРОЦЕСИ ФОРМУВАННЯ ЛИТИХ СТРУКТУР У ТРУБНІЙ ЗАГОТОВЦІ ЗІ СПЛАВУ ЦИРКОНІЮ З НІОБІЄМ

В.С. Вахрушева, Н.В. Грузін

Представлені матеріали є частиною комплексної роботи по виготовленню в Україні труб-оболонок тепловиділяючих елементів. Основою будь-якого технологічного процесу виготовлення труб є використання якісної трубної заготовки, тому в роботі розглянуто процеси виготовлення трубної заготовки і встановлення раціональних способів лиття злитків. Встановлено закономірності структуроутворення в литих трубних заготовках, і запропоновано схеми перерозподілу ніобію у сплаві цирконію Zr1Nb.