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ON THE POSSIBILITY OF USING TRITIUM FOR RADIOGRAPHIC STUDY OF THE DISTRIBUTION OF HYDROGEN IN TITANIUM AND ZIRCONIUM

(Presented by Academician G. V. Kurdyumov, 24 IV 1957)

In the modern use of titanium and zirconium as structural materials, much attention is devoted to the problem of the distribution in them of hydrogen, which sharply reduces the ductility of these metals and increases their notch sensitivity (*1, 2*). However, the existing methods of metallographic analysis do not give a complete picture of the true nature of the distribution of hydrogen in alloys. In view of this, an attempt to use the radioactive isotope of hydrogen (tritium) for this purpose seems timely.

There is no information in the literature on the possibility of using tritium in physical metallurgy. The principal factor hindering the possibility of obtaining tritium radiograms is the low decay energy of this isotope (0.018 MeV), as a result of which, in radiography of alloys containing tritium, the radiation emitted by a layer of metal only about 10^{-4} cm thick can act on the photographic emulsion (*3, 4*).

To create the flux of β-particles necessary from the standpoint of chemical action on the photographic emulsion during radiography (10^6–10^7 β-particles per 1 cm^2 of emulsion surface), it is necessary either to use tritium of very high specific activity (using tritiated water with an activity on the order of 1 Cu/ml), or to saturate the alloy under study with tritium to the greatest possible concentration.

The task of obtaining tritium autoradiograms of titanium and zirconium is facilitated by the high solubility of hydrogen in these metals (in comparison, for example, with steel).

Using the tritiated water at our disposal, with a specific activity of 15 Cu/ml, we succeeded in obtaining quite distinct autoradiograms of titanium and zirconium specimens by saturating them with a mixture of tritium and hydrogen to a concentration of 700–1000 ml of gas per 100 g of metal.

To obtain gaseous tritium (more precisely, a mixture of the isotopes H_2 and T_2) from tritiated water, a method was adopted involving the decomposition of

Fig. 1

Figure 1: Fig. 1

Fig. 2

Figure 2: Fig. 2

water vapor on the surface of zinc turnings in vacuum. This made it possible almost

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Fig. 1. Commercially pure cast titanium after forging and saturation with tritium at 920°. Cooling in air. Hydrogen content 1000 ml per 100 g of metal: *a* –microautoradiogram, 300×. Exposure 30 days; *b* –optical microphotograph, 300×

Fig. 2. Commercially pure cast titanium after forging and saturation with tritium at 980°. Cooling in air. Hydrogen content 2100 ml per 100 g of metal: *a* –microautoradiogram 100×. Exposure 30 days; *b* –optical microphotograph 100×; *c* –same, 300×; *d* –same, 300×

Fig. 3. Commercially pure cast zirconium after forging and saturation with tritium at 1000°. Cooling in air. Hydrogen content 1700 ml per 100 g of metal: *a* –microautoradiogram 300×. Exposure 30 days; *b* –optical microphotograph 200×

Fig. 1

Fig. 2

Fig. 3

one-hundred-percent utilization of tritium during its subsequent introduction into the metals under study.

The tritium obtained by the method described above, whose activity was 12 μCu/l ml of gas, was introduced into metal previously degassed in vacuum at a temperature above the point of the $\alpha \to \beta$ transformation.

Sections for autoradiography were prepared in the usual manner (with protection against work hardening and excessive heating during polishing). The presence on the surface of titanium and zirconium sections of a natural oxide film, which prevented chemical interaction of the metal with the emulsion, eliminated the need for artificially coating the sections with a special lacquer film. This

Fig. 3

Figure 3: Fig. 3

ensured more perfect contact between the emulsion and the metal being radiographed and excluded the possibility of strong absorption of β-particles in the film material, which is especially important for tritium as a β-emitter.

Radiography of titanium and zirconium specimens saturated with tritium was carried out on NIKFI nuclear film of type MR for 20–30 days.

The tritium autoradiograms shown in Figs. 1–3 are "negative" : the darker regions in them correspond to a higher concentration of hydrogen, the lighter ones to a lower concentration.

The high sharpness of the autoradiograms, comparable with the sharpness of optical photomicrographs, is due to the low decay energy of tritium and, as a consequence, to the insignificant scattering of radiation in the metal and in the photographic emulsion.

From comparison of the autoradiogram shown in Fig. 1 with the optical photomicrograph of a titanium specimen, it follows that the structural pattern detected in technically pure titanium is associated with the presence of titanium hydrides. Indeed, although examination of the photomicrograph does not make it possible to assert with complete certainty that the dark constituent visible under the microscope is a hydride phase (5), the fact that the tritium autoradiogram almost completely reproduces the microstructure pattern leaves no doubt about this.

The character of the arrangement of structural constituents in the autoradiograms of titanium with a relatively high hydrogen content, shown in Fig. 2, indicates that the process of phase recrystallization of the alloy takes place according to the Widmanstätten type of structure. It is known that the formation of this structure obeys the principle of the greatest orientational and dimensional correspondence, in which the displacement of atoms of the crystal lattice of the new phase relative to their former positions in the lattice of the old phase would be minimal.

In light of this, it should be assumed that the light needles of the α-phase and the dark accumulations of eutectoid, consisting of the α-phase and titanium hydride, visible in the autoradiograms (Fig. 2), are arranged along the crystallographic planes of the former atomic lattice of the β-solid solution, preserving a crystallogeometric relationship with them.

Such an arrangement of the structural constituents, one of which (titanium hydride) is very brittle, must naturally lead to a sharp decrease in the plastic properties of the alloy and to an increase in its notch sensitivity.

Attention is drawn to the fact that the α-phase needles visible in the autoradiograms produce very slight darkening of the photoemulsion during radiography in comparison with the regions of eutectoid distribution that cause strong blackening, the hydrogen content of which is close to 40 at. %. This clearly confirms the proposition that the solubility of hydrogen in the α-phase at room temperature is very small (1). As is known, confirmation of this fact by other research methods is very difficult (6).

Figure 3 presents an autoradiogram and an optical photomicrograph of a zirconium specimen saturated with tritium at a temperature of 1000°. Compa-

comparison of the autoradiogram with the optical image makes it possible to establish that the structural constituent located along the grain boundaries is zirconium hydride (or, more precisely, a eutectoid consisting of hydride and the α-phase). At the same time, the distribution of hydride inclusions in zirconium, revealed on the autoradiogram (Fig. 3), does not reproduce the microstructural pattern, as was the case in titanium; this evidently indicates the absence of complete structural analogy between the Ti—H and Zr—H systems.

Mastery of the method for obtaining tritium radiograms from titanium and zirconium specimens made it possible to establish a number of important regularities in the distribution of hydrogen in alloys based on these metals, including in cases where, in metallographic examination, the character of the hydrogen distribution in the alloy is not visible at all.

Tritium was used to study the distribution of hydrogen during the welding of titanium. It was of interest to determine whether titanium hydrides accumulate in the fusion zone, owing to the inequality of the solubility of hydrogen in solid and molten titanium and to the presence of a temperature gradient during crystallization of the weld metal.

Radiographic investigation established that the transition of hydrogen from the base metal into the weld metal takes place uniformly, without any noticeable concentration of the hydride phase at the fusion boundary.

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