New method for the measurement of hydrogen diffusion in metals

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Neutron radiography is established as a method to measure hydrogen diffusion in metals. Measurements on β-Ti are reported.

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Radiography with thermal neutrons is a very sensitive method to measure hydrogen under various experimental conditions.[1,2] This fact is due to the high attenuation of the hydrogen isotope H^1 for thermal neutrons, which is large compared to most other materials. Neutron radiography has already been used to measure the mutual diffusion of light and heavy water[3–5] where the method was calibrated with a series of standard mixtures; a similar calibration procedure for the measurement of hydrogen diffusion in metals is rather cumbersome in its experimental verification and it requires the use of some other method for the measurement of the hydrogen content. One of the main emphases of the present work was to overcome this problem.

We use the direct method of neutron radiography with a 25-μm-thick Gd converter on the back side of an Osray TA T4 DW film. The developed radiographs were scanned with a microdensitometer with a slit aperture of 75×2500 μm^2. The characteristic curves, which represent the optical density as a function of the exposure, are S shaped and possess a part where the density is proportional to the logarithm of exposure E. Within this limit we can write

$$D = G \log E + D_0, \tag{1}$$

where D is the optical density, G is the gradation, and D_0 is a residual density of the film. Therefore, the density difference for two different exposures with and without a neutron absorbing or scattering material in the beam is

$$D_2 - D_1 = G \log(I_2/I_1), \tag{2}$$

where the intensities of the neutrons striking the converter are related by the well-known experimental relation

$$I_2 = I_1 \exp(-N\sigma d). \tag{3}$$

Here σ is the total microscopic cross section of the material, N is its atomic density, and d is its thickness. Thus we obtain for the density difference

$$D_2 - D_1 = -0.4343 GN\sigma d. \tag{4}$$

The optical density therefore is a linear function of additional material in the beam if the exposures are restricted to the range mentioned above.

To test the validity of the method, six plates of β-titanium[6] (with 13% V, 11% Cr, 3% Al) each of size $10\times65\times1.2$ mm^3 were loaded electrolytically up to one-half their length with hydrogen. The attained hydrogen concentration was 12%. These plates were then sandwiched to form a sample 7.2 mm thick in order to increase the neutron radiographic contrast. The total hydrogen amount was low enough so that at the object-converter distance of 30 cm the number of scattered neutrons which reached the converter was less than 1% of the transmitted ones. The sample was mounted in a furnace with aluminum walls to allow the radiographs to be taken without a temperature interruption. The diffusion process was observed at 55, 80, and 110 °C. For each of these temperatures neutron radiographs were taken at different times.

Typical densitometric scans of the radiographs are shown in Fig. 1 and, in view of the above and disregarding the noise of the film, are directly proportional to the hydrogen concentration. These experimental curves can be represented by error functions which are the solutions of the one-dimensional diffusion equation for a step function as initial distribution for a non-concentration-dependent diffusion coefficient and for an infinite system; this latter point is fulfilled as long as the concentration variation due to diffusion has not yet reached the boundary of the diffusion medium.[7] The derivative of these hydrogen concentration profiles is directly related to the diffusion coefficient.[7] In particular, when using the slope k_0 at the position of the initial concentration step, the diffusion coefficient is

$$D = (4\pi k_0^2 t)^{-1}, \tag{5}$$

where t is the time elapsed since the beginning of diffusion. The diffusion coefficients thus obtained are

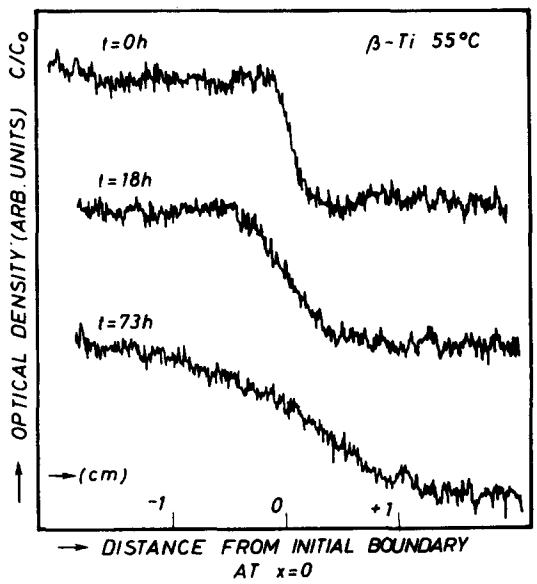

FIG. 1. Densitometer scans at different times. Disregarding the noise, the curves directly represent hydrogen concentration profiles.

TABLE I. Diffusion coefficients of H in β-Ti.

T (°C)	D (10^{-10} m^2/s)
55	0.72 ±0.14
70	1.17 ±0.23
110	2.11 ±0.42

presented in Table I. These values agree within the experimental accuracy with those obtained by other methods.[8]

In summary it appears that the main advantage of the neutron radiographic method for studying hydrogen diffusion in metals is the characteristic that the hydrogen distribution in the sample is obtained directly with no separate calibration procedure required.

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