NEUTRON RADIOGRAPHY AS TOOL FOR THE DETECTION AND MEASUREMENT OF HYDROGEN DISTRIBUTIONS

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ABSTRACT. The uses of neutron radiography for detection of hydrogen and the measurement of hydrogen distributions are discussed. The most severe limitation is due to the heavy scattering of thermal neutrons by hydrogen. The applications can be divided into three groups:
1) Detection of hydrogen in heavier materials.
2) Detection of hydrogen in the presence of deuterium.
3) Biomedical uses of neutron radiography.

1) INTRODUCTION

Neutron radiography is an extensively expanding branch of nondestructive testing. The basis of neutron radiography is, first, the fact that the mass of attenuation coefficient for thermal neutrons changes irregularly from element to element compared with X-rays (Fig. 1). A second advantage is the fact that the cross-section varies even from isotope to isotope of the same element in many cases.

Many of the non-nuclear applications of neutron radiography are based on the high attenuation of thermal neutrons by hydrogen. This makes it possible to detect hydrogen under very different experimental conditions.

Most applications of neutron radiography are in nuclear industry up to now. Here it is used mainly for investigations of nuclear fuel and fuel elements. So it is possible to measure the arrangement of fuel pellets in a rod, the enrichment of pellets, the burnup and dimensional changes of fuel due to reactor operation.

The nuclear reactor is the most frequently used source for neutron radiography. The arrangement used at the 250kW Triga Mark II reactor of Vienna is shown in Fig. 2. An 8 cm thick Bi-filter reduces the background of gamma rays in the neutron beam. The conical collimator produces a "point source" of neutrons. As neutron detectors we use either the conventional 25 μm Gd-converter together with Osray TA T4 TW film or track-etch foils CA 15 type B of Kodak-Pathe. These are cellulose nitrate foils coated with a boron-compound. The advantage of these foils is their insensitivity to gamma rays.

For some uses of neutron radiography it would yet be desirable to have a portable neutron source for in situ and field measurements. The development of Cf–252 makes it possible to construct rather simple portable neutron sources for neutron radiography. (1). The sketch of a portable source is shown in Fig. 3 (2). The assembly is 66 cm long with a diameter of 56 cm. It contains the Cf–252 source, a zirconiumhydride moderator, a device for mounting a collimator, a Be-reflector and as shielding materials a boron-carbide-epoxy mixture and depleted uranium. Using a source of 268 μg Cf–252 a thermal neutron flux of 1.2×10^6 cm^{-2} s^{-1} was obtained.

One main limitation to the use of neutron radiography for the detection and measurement of hydrogen is imposed by its high scattering cross section. Because of this fact a severe image degradation results because the scattered neutrons can reach the film, producing an intense noise of the image. Therefore it is in general not useful to investigate thicknesses of hydrogen containing substances which correspond in their hydrogen content to water thicknesses of more than about 1 cm. A reduction of the number of scattered neutrons reaching the film can be achieved by putting a second collimator, a so-called "anti-scatter grid" between object and film. This is a fine collimator. If the image of the collimator is not to appear on the film, the grid must be moved during exposure. The effect of such a grid can be measured by the improvement of the sharpness of the image of a knife-edge, the so-called "edge-spread function". Preliminary results of such investigations were reported earlier (3).

The applications of neutron radiography for the detection of hydrogen can be divided in three major groups:

2) DETECTION OF HYDROGEN IN THE PRESENCE OF HEAVIER MATERIALS

Referring to Fig. 1 it can be seen that hydrogen has a mass attenuation coefficient which is more than 2 orders of magnitude greater than that of most other elements. Based on this fact hydrogen compounds can easily be detected where other experimental methods fail or give bad results. Special applications in this case are: investigation of water or organic inclusions in metals, detection of hydrogeneous substances behind thick metal walls and in metal tubes or metal containers, investigations of explosives, inspection of metal adhesions etc. In the case of hydrogen compounds in steel, use of cold neutrons has a great advantage in increasing the contrast (4). This is due to the high coherent scattering of iron which has its Bragg-edge at 0.005 eV. For lower energies the Bragg-equation cannot be fulfilled any more and therefore the penetration of cold neutrons through steel is much better than that of thermal neutrons. A beam of cold neutrons can simply be produced by installing a Be-filter in the neutron beam. It was reported recently (5) that using an intense beam

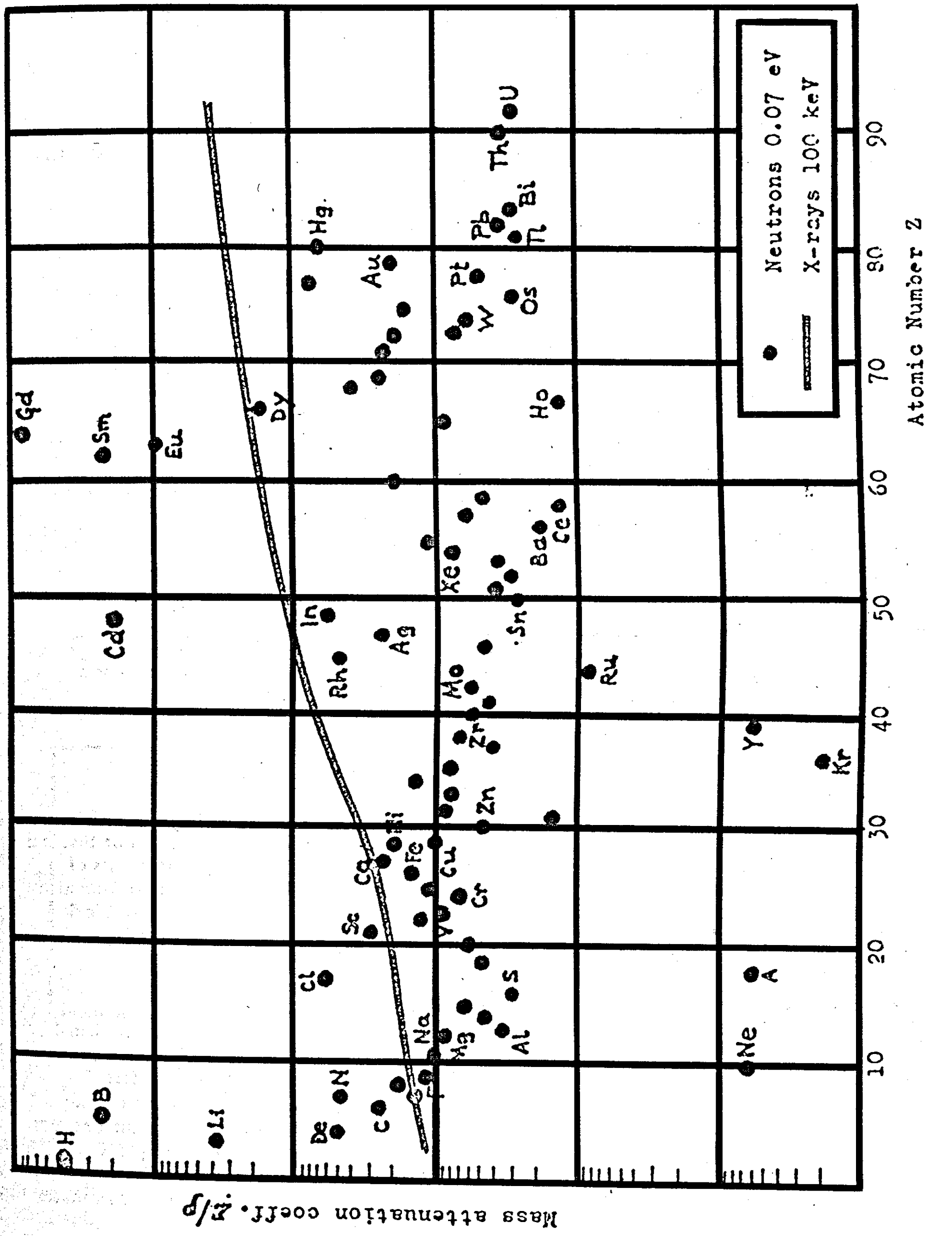

Fig. 1. Mass attenuation coefficient of the elements for thermal neutrons and X-rays (from AE—484)

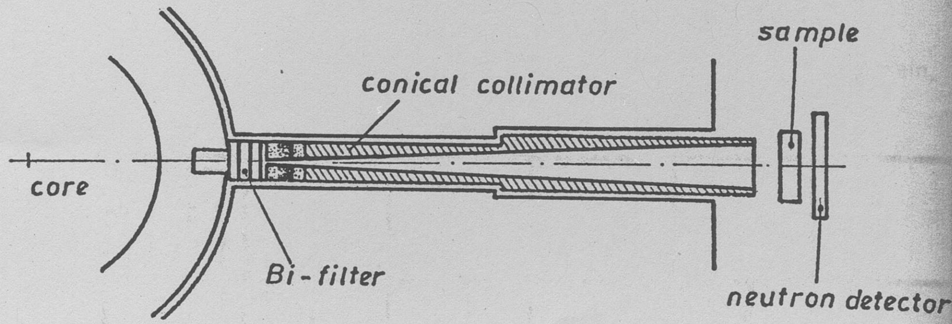

Fig. 2. Neutron radiography setup at the Triga reactor Vienna

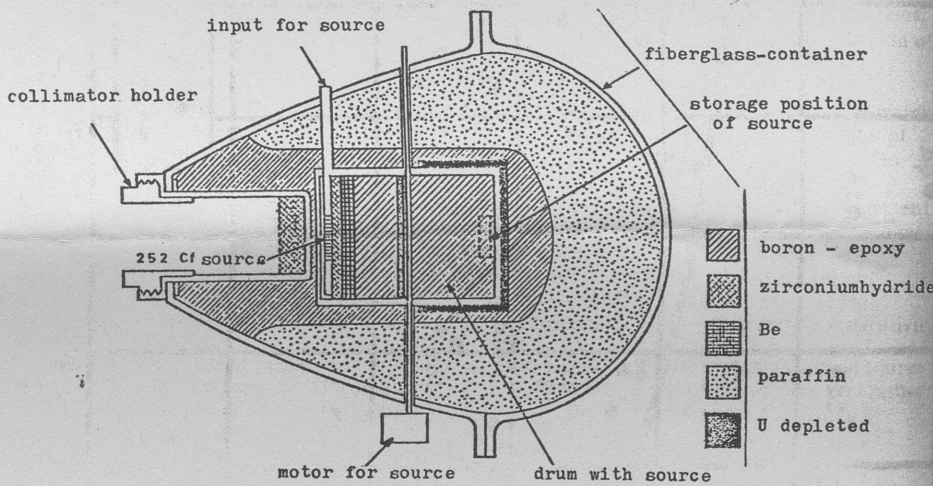

Fig. 3. Portable Cf–252 neutron source (see ref. /1/ & /2/)

of cold neutrons it was possible to detect Perspex less than 2 mm in thickness in the centre of 15 cm of steel, the corresponding values for X-rays being nearly one order of magnitude worse. For thermal neutrons the penetration through steel is limited to about 5 cm. In recent time measurements of water motion in concrete under a temperature gradient were performed successfully at Atominstitut Vienna. Other experiments have been made on the diffusion of hydrogen in zirconium (6). Work on a quantitative determination of diffusion coefficients of hydrogen in various metals using neutron radiography will be published in the near future.

3) DETERMINATION OF HYDROGEN IN THE PRESENCE OF DEUTERIUM

These applications are based on the fact that the attenuation coefficient of deuterium for thermal neutrons is much lower than that of the common isotope H–1. This factor was used to measure quantitatively the diffusion in the systems (7) $H_2O - D_2O$, $CH_3OH - CD_3OD$ and $CH_3OD - CD_3OD$. The measurements were performed by putting in a thin Al-container two layers of the substances investigated on top of each other. Afterwards the smearing out of the borderline between the two liquids was observed by neutron radiography as a function of time, the density profile of the films was measured afterwards with a densitometer. Comparing these density values with those obtained with a series of standard mixtures, concentration profiles with diffusion time as parameter were obtained. Fig. 4 shows such a typical concentration profile.

From these concentration profiles the value of the diffusion coefficient can be determined by fitting error functions which are the solutions of the ordinary diffusion equation for infinite medium to the concentration profiles obtained. By a more elaborate procedure the diffusion coefficient as a function of concentration can be determined. The measurements on the methanol system indicated that the methanol molecule diffuses as a whole. This can be seen by the fact, that the values of the diffusion constant are exactly on the same straight line in Fig. 5 independent of which hydrogen atom

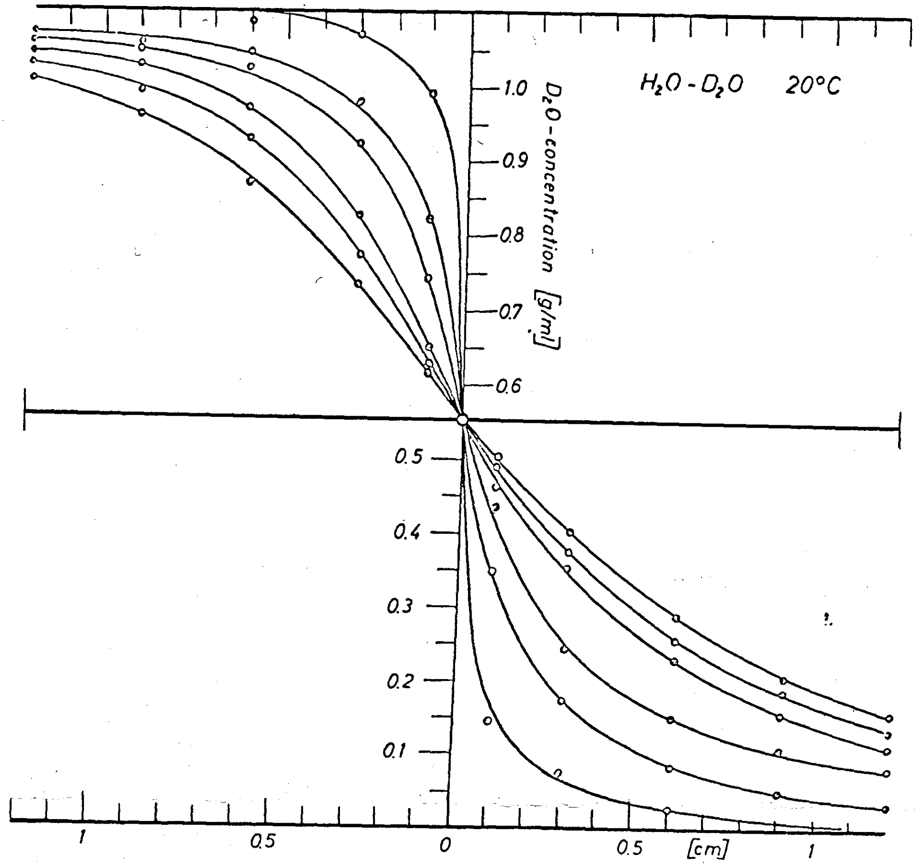

Fig. 4. Concentration profile of the diffusion of $H_2O - D_2O$ obtained by neutron radiography

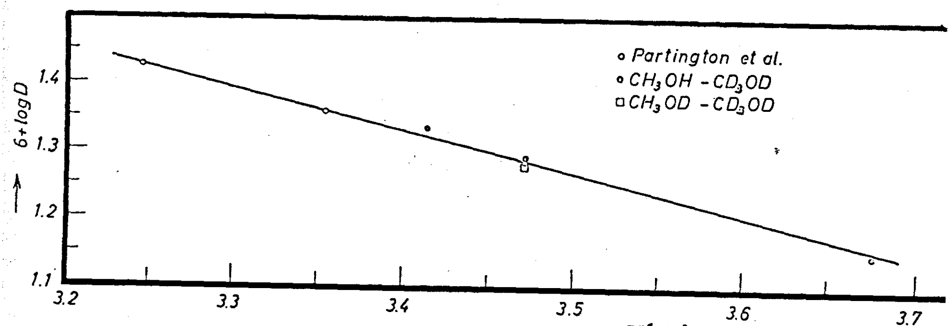

Fig. 5. Diffusion coefficient of methanol as a function of temperature obtained by neutron radiography

4) NEUTRON RADIOGRAPHY OF BIOMEDICAL OBJECTS

A severe limitation is here again the thickness of most of the biological objects. It is quite easy to obtain radiographs of small plants or insects, but there exists no worthwhile application of neutron radiography to those small objects up to now. An advantage of neutron radiography here would be the fact that neutrons can penetrate bone much easier than soft tissue. Most promising seems therefore to be the application of neutron radiography to osseous tumors and to dental problems (8, 9). Reijonen et al. (10) have demonstrated the applicability of neutron radiography to thin pathological samples of bones. Here a good resolution could only be obtained for samples thinner than 1 mm; the results show quite promising applications in the in vitro study of deseases. For in vivo studies (11) the use of epicadmium neutron beams seems to have some useful potentialities because of the lower

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