

RETENTION OF ATOMIC HYDROGEN IN PYROLYTIC GRAPHITE

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ABSTRACT

When irradiating a graphite target with an atomic hydrogen beam a dose dependent retention is observed which varies from 4% of the incident atoms at a fluence of 10^{15} cm^{-2} to 0.24% at 10^{19} cm^{-2} . The trapping efficiency rises continuously with the temperature up to 700 K and then decreases drastically.

1. INTRODUCTION

Graphite has been proposed as a nonstructural first wall material for a tokamak fusion device because of its low Z-number and is already used as material for a limiter in a tokamak. Furthermore graphite samples are used to determine the hydrogen fluxes to the first wall of tokamaks. For these applications the knowledge of the interaction of hydrogen atoms and ions with graphite is essential. Above all the retention is important for the tritium inventory in a fusion reactor and for recycling of fuel material and its reactions for the erosion problems and impurity production. We have measured the retention of hyperthermal (2700 K) hydrogen atoms on pyrolytic graphite as a function of the graphite temperature and the irradiation dose (1) complementary to the already known trapping of H^+ and D^+ ions with energies between 50 eV and some keV (2) and earlier results on the retention of atomic hydrogen at very small doses (3,4).

2. EXPERIMENTAL

The trapping of hydrogen atoms is measured by irradiating a graphite sample with a hydrogen atom beam containing tritium as tracer and determining the tritium activity of the probe after the irradiation. The amount of retained hydrogen can then be calculated from the mixing ratio of hydrogen and tritium in the beam assuming the absence of an isotope effect. The irradiation set up is shown in Fig. 1. The atomic beam is produced by thermal dissociation of hydrogen in a heated tungsten tube (2700 K) and reaches an intensity at the target up to $10^{16} \text{ atoms cm}^{-2} \text{ s}^{-1}$. A velocity selector between the source and the diaphragm eliminates the tungsten impurity of 0.1%. The graphite probes are made from pyrolytic carbon (Union Carbide) in the form of ribbons of 0.02 cm thickness by machine tooling. The probe is pretreated before irradiation by an one hour baking at 2100 K in order to ensure that the sample is hydrogenfree. The probes are irradiated in C-direction and the temperature is controlled by a pyrometer. After the irradiation the probes are brought into a separate device for a complete combustion (Fig. 2). The tritium activity of the collected water is determined in a liquid

scintillation counter.

3. RESULTS AND DISCUSSION

The retention of atomic hydrogen on pyrolytic graphite has been measured for three different graphite temperatures and for irradiation doses between 10^{15} and 10^{19} atoms cm^{-2} (Fig. 3 and 4). The amount of retained hydrogen is dose dependent and varies from 4% of the incident atom beam at a fluence of 10^{15} cm^{-2} to 0.16% at $5 \cdot 10^{19} \text{ cm}^{-2}$, i.e. $8 \cdot 10^{16} \text{ cm}^{-2}$ hydrogen atoms were found to be trapped in the high dose region. This value is one order of magnitude higher than the saturation value reported by Staudenmaier et al. (2) for an irradiation with 50 eV hydrogen ions. We interpret this difference as follows. The trapping coefficient for thermal atomic hydrogen is rather small and therefore diffusion and recombination determine the trapping rate even at small doses. When irradiating with energetic ions practically all ions are trapped up to a critical dose. Further trapping above this dose would be due to bulk diffusion rather than direct implantation. This contribution would be small and is not investigated. On the other hand, the diffusion of hydrogen in graphite could be influenced by radiation induced defects and their interaction with hydrogen. If this is true completely different processes should be responsible for the trapping of hydrogen by irradiation with thermal atoms than in the case of implantation of energetic ions.

Fig. 3 and 5 also show the trapping of atomic hydrogen at higher temperatures of the probe. The trapping efficiency rises with temperature. The reason can only be a different temperature dependence of the diffusion constant and the recombination rate at the surface. Above 700 K the trapping efficiency decreases as can be seen in Fig. 4. The reason for this decrease could be a very fast diffusion above 700 K or the chemical interaction of hydrogen with carbon. Unfortunately, large differences of temperature dependencies for the production rate of methane (5,6) have been observed. Therefore, any discussion on this basis could only be highly speculative. Irradiation with molecular hydrogen on the other hand, leads to negligible trapping. Our measured value is at least 3 orders of magnitude smaller than that obtained with atoms.

The main assumption for the interpretation of our results is: The absence of an H/T-isotope effect. This assumption has been checked by tracer experiments with two different mixing ratios of hydrogen and tritium and with a mixture of deuterium and tritium without drastic discrepancies. Additionally we have also measured the deuterium concentration in the probe after the irradiation via the $\text{D}(\text{He}, \text{p})\text{He}$ nuclear reaction using the experimental set up in Garching (2) and found a relatively good agreement within 30%. By adding up all the uncertainties the error for the absolute value of retention should not exceed 100%.

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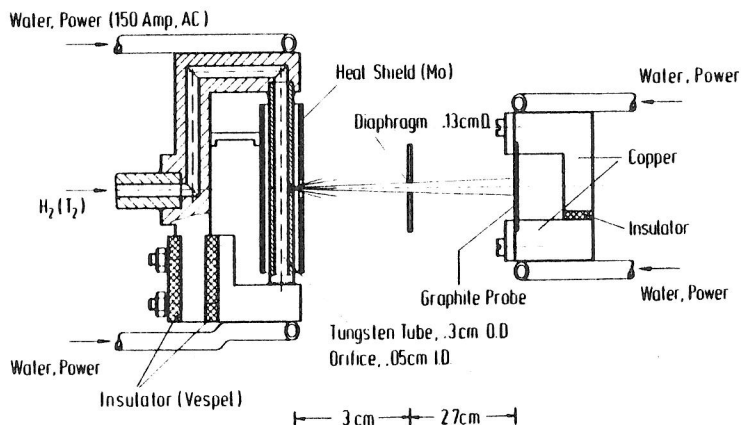


Fig. 1 Irradiation set up and beam geometry. The atomic beam is produced by thermal dissociation of $H_2(T_2)$ in the tungsten tube at 2700 K in a differentially pumped chamber. Between the tungsten tube and the diaphragm a velocity selector is installed to eliminate the tungsten impurity of the beam (0.1%).

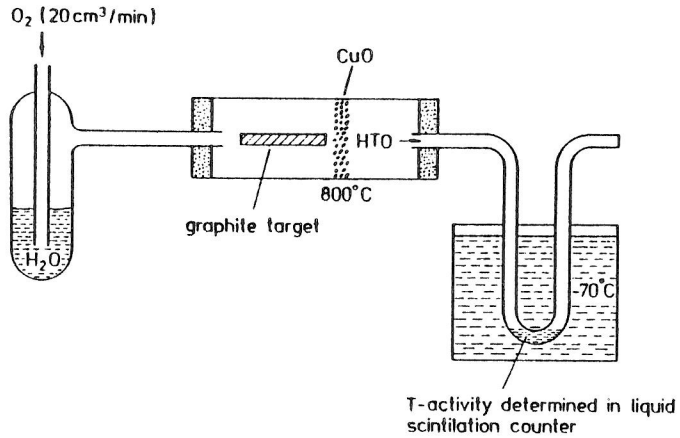


Fig. 2 Apparatur for the combustion of the irradiated graphite. The quartz tube with the graphite probe is heated in a muffle furnace.

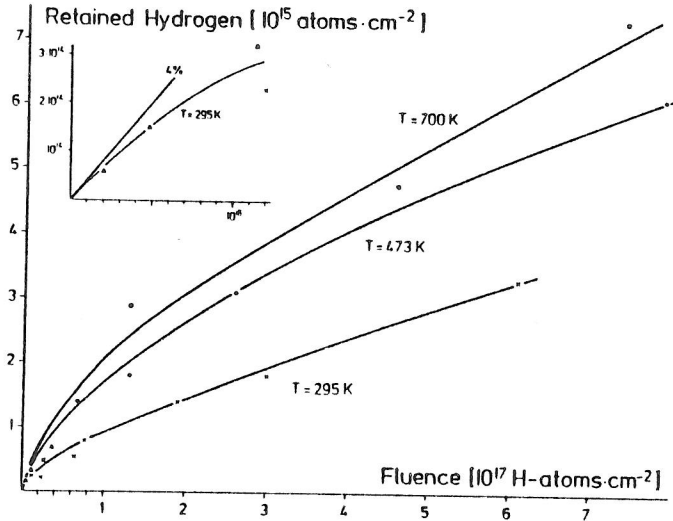


Fig. 3 Retention of hydrogen in graphite as a function of the irradiation fluence for different temperatures. Mixing ratio: $H_2:T_2 = 820:1$.

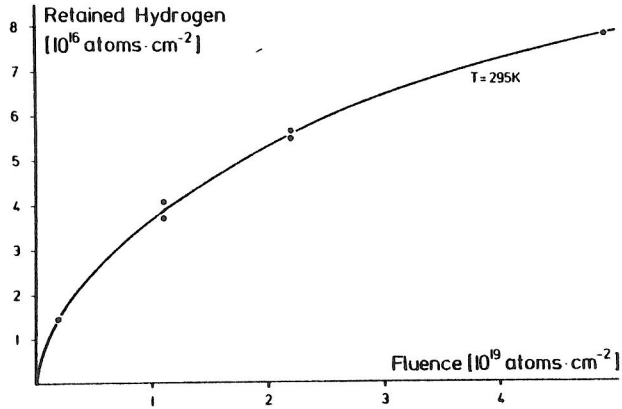


Fig. 4 Retention of hydrogen in graphite at high irradiation fluences. Mixing ratio: $H_2:T_2 = 4 \cdot 10^4:1$.

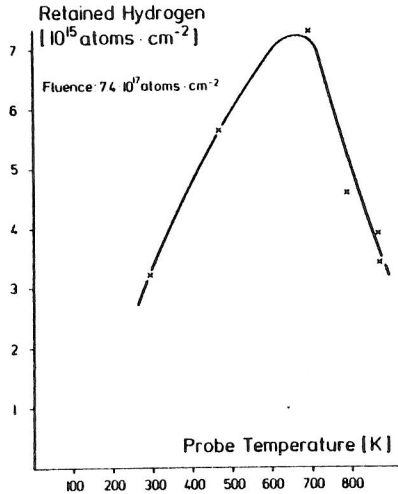


Fig. 5 Temperature dependence of hydrogen retention in pyrolytic graphite. Irradiation fluence: $7.4 \cdot 10^{17}$ cm $^{-2}$. Mixing ratio: $H_2:T_2 = 820:1$.