TRAPPING OF DEUTERIUM AT DAMAGE IN GRAPHITE

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ABSTRACT

Enhanced retention of deuterium (D) and tritium (T) in graphite due to neutron damage could result in large T inventories in future DT fueled fusion devices such as CIT and ITER. This paper describes experiments done to characterize the effect of lattice damage on retention of D in graphite. Lattice damage was produced in several types of graphite by irradiation with 6 MeV C+ ions or with neutrons. The damaged graphite was then heated to 1200°C and exposed to D_2 gas. The concentration of D retained in the graphite was measured by nuclear reaction analysis and compared to the level of damage. In POCO, N3M and H451 graphites, D retention increased with damage at low damage levels but saturated above 0.04 dpa at a D concentration of about 650 atomic ppm. D retention in damaged highly oriented pyrolytic graphite was an order of magnitude smaller than in the other graphites indicating that crystalline microstructure is an important factor. These results indicate that neutron damage in CIT and ITER may cause retention of large inventories of tritium in graphite components.

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1. Introduction

Large amounts of graphite are used as the plasma-contacting material in tokamaks. In DT fueled machines, the amount of tritium (T) which might be absorbed into the bulk of the graphite must be considered. For example, the 5 grams of T allowed for DT operation in the Tokamak Fusion Test Reactor (TFTR) distributed through the ~2000 kg of graphite in TFTR would correspond to a T concentration of only 10 atomic parts per million (appm). This concern prompted measurements of the D concentration in the TFTR limiter after extensive exposure to high-power D plasmas [1]. The result that less than 1 appm of D was retained showed that retention of T in bulk graphite was unlikely to significantly affect DT operation in TFTR.

The situation may be different for the next generation of tokamaks such as the International Thermonuclear Experimental Reactor (ITER). Such a machine is expected to produce enough DT fusion neutrons during its lifetime to cause lattice damage to a level of about 10 displacements per atom (dpa) in graphite. Furthermore there is evidence that lattice damage may increase retention of hydrogen in graphite [2]. Enhanced uptake of tritium in bulk graphite due to neutron damage could lead to an unacceptably large tritium inventory. The experiments described in this paper were done to characterize the effect of lattice damage on the trapping of hydrogen isotopes in graphite.

2. Experimental

Various types of graphite were irradiated with 6 MeV C^+ ions to produce lattice damage without changing the chemical composition. The graphites used in this study included: H451 graphite from Great Lakes Carbon Company,

a neutron damage resistant graphite with a grain size of 0.7 mm which is a design base material for high temperature gas-cooled reactors; POCO an isotropic graphite with a mean grain size of $10\mu m$ from POCO Graphite Inc.; Graphnol N3M with a mean grain size of 30 μm from Oak Ridge National Laboratory; and compression annealed highly-oriented pyrolytic graphite (HOPG) which is a single crystal. The samples were near room temperature during the ion irradiation. In addition a sample of H451 graphite was damaged by neutron irradiation in the High Flux Irradiation Reactor (HFIR) at Oak Ridge National Laboratory. During neutron irradiation the sample temperature was 875°C.

After irradiation the samples were exposed at 1200°C to D_2 gas at a pressure of one atmosphere for three hours and then held at 1200°C for an additional hour under vacuum before cooling. This temperature was chosen to represent an average operating temperature for the ITER limiter. The concentration of D in the samples was then measured using $^3He(D,H)\alpha$ nuclear reaction analysis. Figure 1 shows measurements of the D concentration within about 1 μm of the surface, versus lateral position across the ion-irradiated region. Data is shown for four types of graphite and for three different irradiation doses. Samples irradiated to fluences of 10^{16} and 10^{17} C^+/cm^2 have much higher D concentrations in the damaged areas than in the adjacent undamaged regions.

The damage produced by the ion irradiation varies with depth so it might be expected that the concentration of trapped D would also depend on the depth. The depth profile of D in the graphite samples was determined by measuring the nuclear reaction yield versus energy of the ^3He analysis beam. Higher energy ^3He probes D at greater depths into the material. Since the

stopping power and nuclear reaction cross sections are well known, the results can be unfolded to yield the D concentration versus depth. The mathematical details of how this is done are described in Ref. 3. ^{3}He energies up to 2.8 MeV allowed the D concentration to be determined to a maximum depth of about 8 μm.

Figure 2 shows the D concentration versus depth for the various samples. In the N3M and H451 graphite samples irradiated to fluences of 10^{15} and 10^{16} C$^+$/cm^2 the D concentration increased with depth down to 5 microns from the surface and then decreased at greater depths. The POCO graphite irradiated to 10^{17} C$^+$/cm^2 had about 600 appm of D from the surface to about 5 microns, and less at greater depths. The sample of highly oriented pyrolytic graphite (HOPG) irradiated to 10^{17} C$^+$/cm^2 had about 70 appm of D over the entire depth probed by the analysis.

Figure 3 shows the damage produced in graphite by 10^{17} C$^+$/cm^2 at 6 MeV calculated using the TRIM [4] Monte Carlo particle transport code. The damage increases from about 0.2 dpa near the surface to more than 10 dpa at the end of range 5 μm beneath the surface. This depth dependence of the damage is reflected in the profiles of D concentration versus depth shown in Fig. 2 for the two samples irradiated to 10^{15} C/cm^2. The D concentration varies less with depth for higher fluence irradiations due to saturation of the D retention at high damage levels.

The near-surface D concentrations in Fig. 2 can be correlated with the damage obtained from Fig. 3 scaled according to the $^+$C ion dose. For example the POCO graphite irradiated with 10^{17} $^+$C/cm^2 has a near-surface D concentration of 600 appm and a corresponding near-surface damage level of 0.2 dpa. Figure 4 shows a plot of the D concentration versus damage level

obtained in this way. The points for POCO and HOPG at 10±5 dpa are from the observation that the concentrations of D in the POCO and HOPG do not increase with depth (Fig. 2) whereas the damage is almost 2 orders of magnitude greater at 5 μm than near the surface.

The N3M, H451 and POCO graphites appear to behave similarly. The D concentration in these materials increases with increasing damage levels and saturates near 10^3 appm at damage levels above about 0.1 dpa. In the POCO graphite irradiated with 10^{17} C/cm^2 the concentration of retained D was about 600 appm at the end of range 5 μm beneath the surface where the damage was on the order of 10 dpa (see Figs. 2 and 3). Also shown in Fig. 4 is the D concentration retained in a sample of H451 graphite damaged by neutron irradiation in the HFIR reactor to a dose of 1.5x10^{22} n/cm^2 of neutrons with energy greater than 50 keV. This irradiation produced about 10 dpa. The concentration of D retained in the neutron damaged H451 graphite was 650 ±250 appm. In other words, our measurements show that retention of D in graphite damaged by neutron and ion irradiation is similar.

A parameterization for c the concentration of retained D as a function of damage d is given by

$$c = c_s \ (1\text{-}\exp(\text{-}d/d_s)) \ . \tag{1}$$

The fit of Eq. 1 to the data shown in Fig. 4 gives the values $c_s \approx 650$ appm and $d_s \approx 0.035$ dpa for the two parameters. This parameterization may be useful for modeling of tritium inventory in design studies.

The concentration of D retained in damaged HOPG graphite is much smaller than in the other graphites. The apparent saturation level for D retention

in HOPG is c_s=80 appm. The fact that D retention in the single crystal HOPG is much lower than in the other graphites indicates that crystalline microstructure is an important factor for damage induced D retention in graphite.

3. Consequences for Tritium Inventory in CIT and ITER

Current design studies for the next generation of DT fueled tokamaks such as CIT and ITER, call for large quantities of graphite as the plasma-contacting material. These machines will be the first to produce enough fusion neutrons to cause significant lattice damage in the graphite. This damage may cause enhanced retention of DT in the graphite resulting in large inventories of tritium. Here we estimate the tritium inventories at the end-of-life for CIT and ITER. These estimates are based on the assumptions that the concentration of DT in the graphite increases with damage as found in our simulations, and that the DT concentration will be uniform throughout the bulk of the graphite since the neutron damage will extend throughout the graphite. Table 1 summarizes the estimates. The damage is calculated from the neutron fluence using the conversion factor 0.7 $dpa/(10^{21}$ $n/cm^2)$ [5]. The concentration c of retained DT is obtained from the damage using Fig. 4 or Eq. 1. The tritium inventory is given by T = a c m, where m is the mass of graphite and a = 1.25 x 10^{-7} /appm. This analysis shows that neutron damage could significantly increase the inventory of tritium in bulk graphite in CIT and ITER.

The tritium inventories listed in table 1 probably represent upper limits. The following effects might reduce tritium inventories. Tritium may not penetrate through the entire bulk of the graphite. Our experiments

show only that it penetrates to about 5 μm in graphite. The depth of penetration will be examined in a future study. Also the uptake of tritium from a plasma may be slower than uptake of D from gas at a pressure of one atmosphere, as used in the present study. On the other hand the exposure time will be much longer in CIT and ITER than was used here. Finally, the temperature of the graphite during irradiation might be higher in CIT and ITER than in our studies. This might cause some annealing of the damage and a lower level of accumulated damage and a corresponding lower level of retained tritium.

4. Conclusions

In POCO, N3M and H451 graphite lattice damage greatly increases the retention of hydrogen isotopes. In these materials D retention increases with increasing damage at low damage levels but the effect saturates above 0.04 dpa at a D concentration of about 650 appm. The concentration of D retained in damaged HOPG is about an order of magnitude smaller than in POCO, N3M and H451 graphites suggesting that crystalline microstructure is an important factor for damage induced D retention in graphite. The fact that D remained in the graphite after vacuum annealing at 1200°C shows the binding energy of the D is several eV/atom suggesting that covalent chemical bonds, possibly C-D bonds may be may be causing the D retention.

Retention of DT in graphite due to neutron damage in CIT and ITER could result in accumulation of large tritium inventories. Assuming uniform DT concentrations at levels observed in our experiments the end-of-life tritium inventories are estimated to be about 17 grams for CIT, and on the order of a kilogram for ITER. The initial physics phase of operation in ITER may

already produce enough damage to approach the level where damage-enhanced DT retention begins to saturate.

Table 1. Estimated upper limit of tritium inventory at end of life due to trapping at neutron damage in graphite.

Machine	Graphite (10^3 kg)	neutron fluence (10^{20} n/cm^2)	Damage (dpa)	DT (appm)	T inventory (grams)
CIT	2	0.06	0.004	70	17
ITER	20				
Physics Phase		<0.7	<0.04	<440	<1100
Technology Phase		210	12	650	1600

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FIGURE CAPTIONS

Fig. 1.   Concentration of D within about 1 μm of the surface in graphite
          damaged by ion bombardment with 6 MeV C^+ versus lateral position
          across the damaged region.   The different symbols correspond to
          different types of graphite and different C^+ fluences.   The bar
          indicates the lateral extent of the analysis beam.

Fig. 2.   Concentration of D versus depth in graphite damaged by ion
          bombardment with 6 MeV C^+.   The different symbols correspond to
          different types of graphite and different C^+ fluences.   At low
          fluences D retention is greater where the damage is greater, but
          the D retention saturates at high damage levels.

Fig. 3.   Damage versus depth for graphite irradiated with 10^{17} C^+/cm^2 at
          6 MeV calculated using the TRIM particle transport code.

Fig. 4.   D concentration versus damage in graphite irradiated with 6 MeV C^+
          (filled symbols) and neutrons (open circle).   Neutron and ion
          damage have similar effects on D retention.   D retention increases
          with damage at low damage levels but saturates above about
          0.04 dpa.   The line shows the fit of Eq. 1 to the data for POCO,
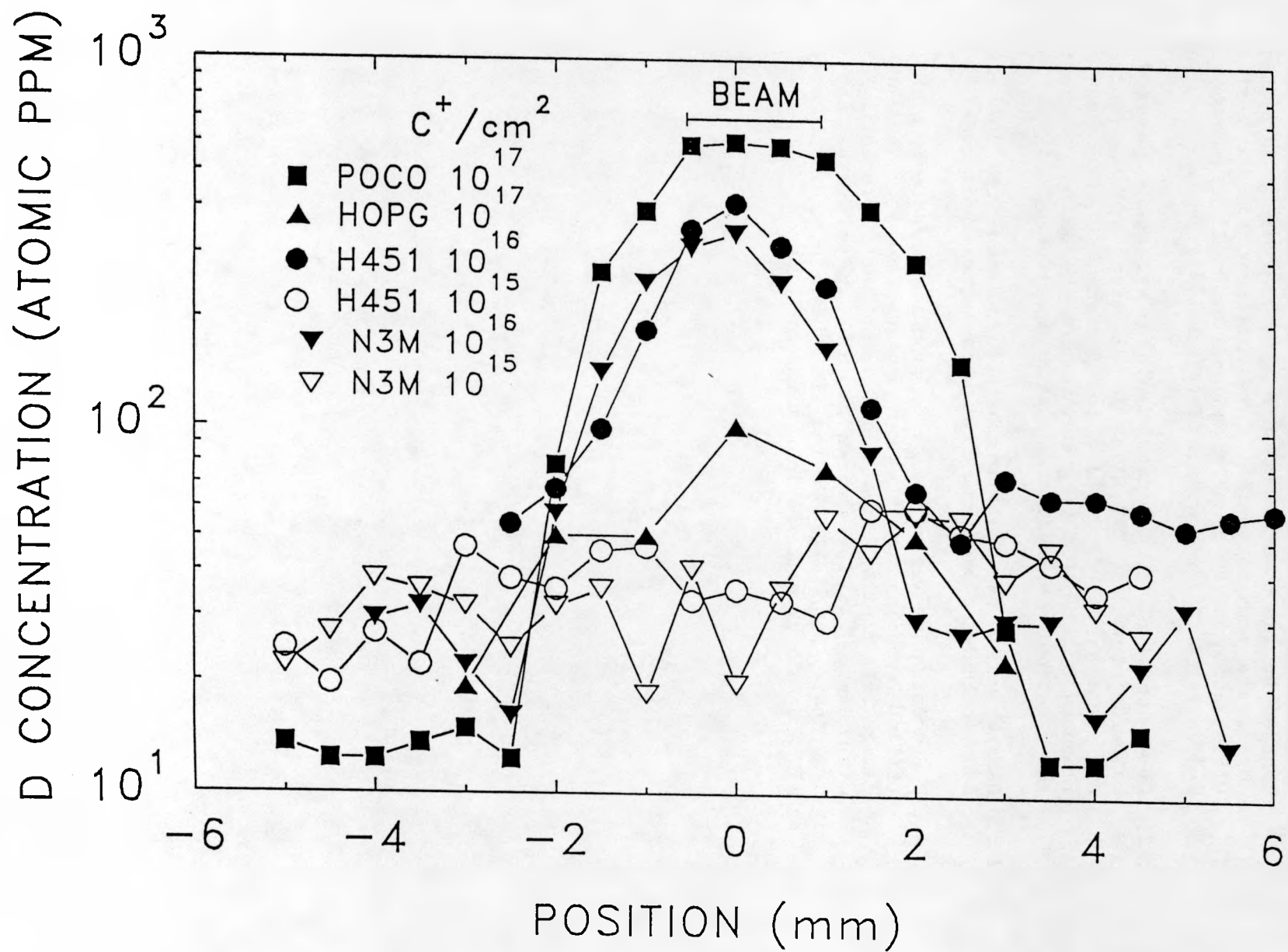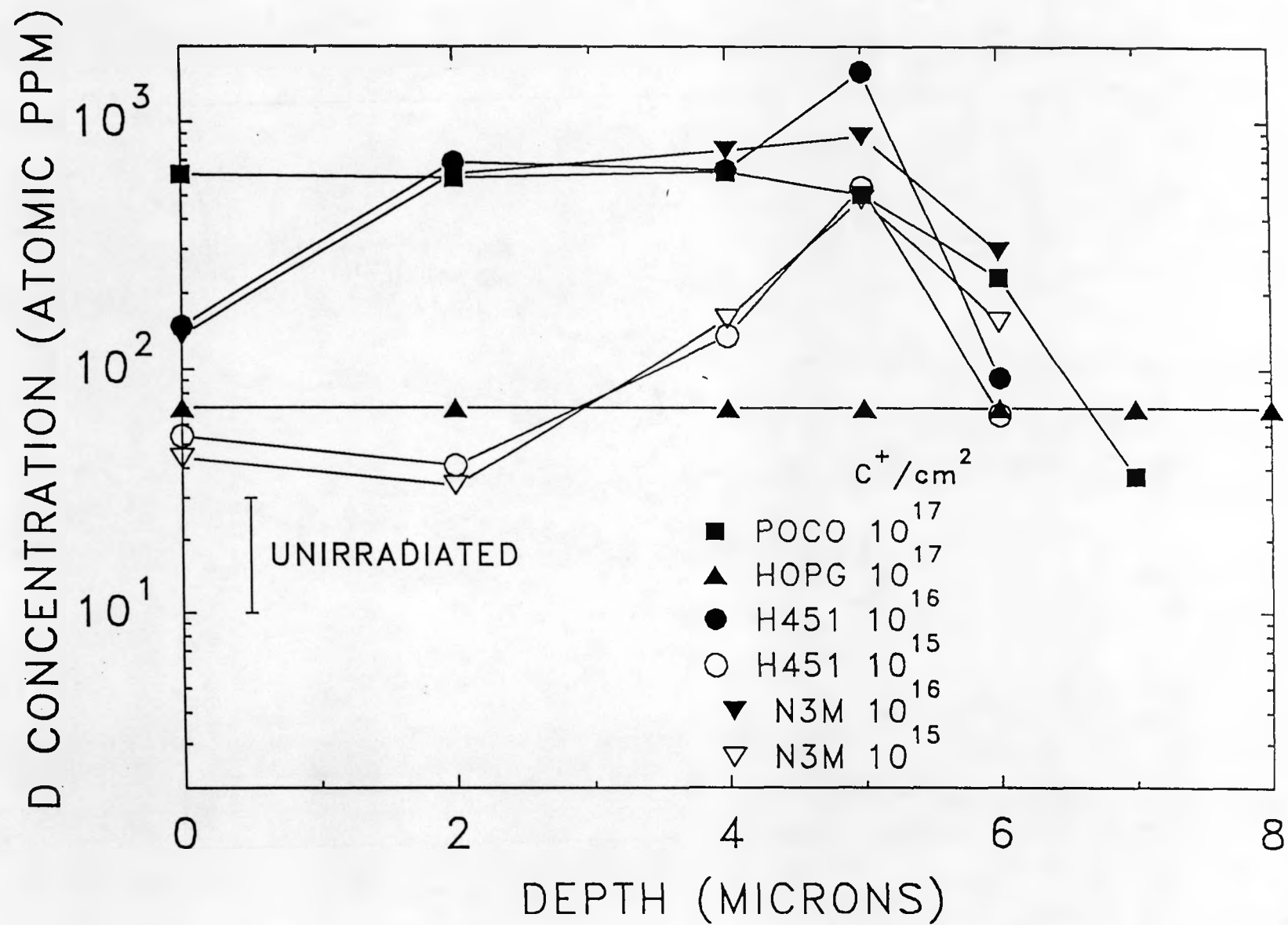   N3M and H451 graphites which behave similarly.
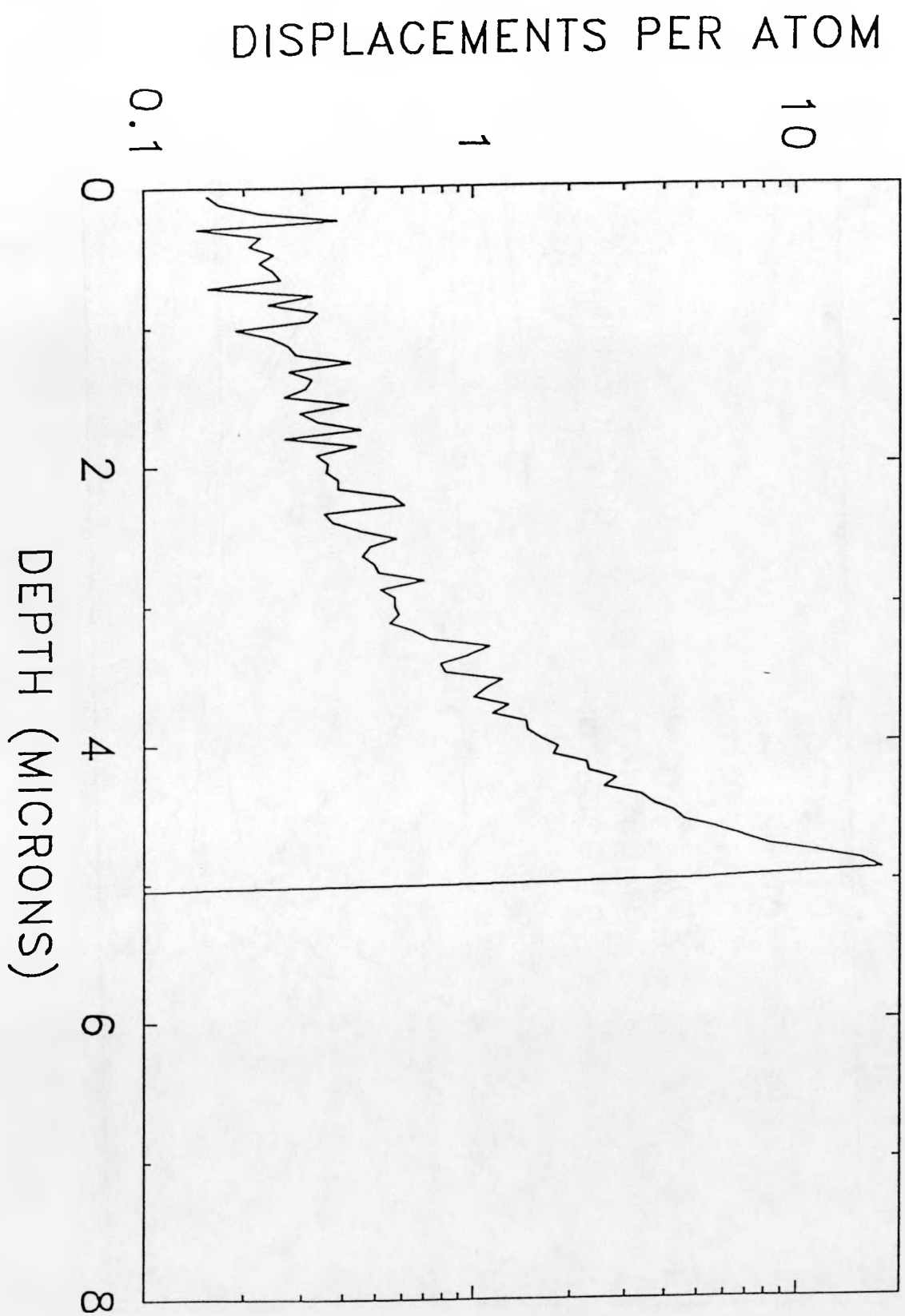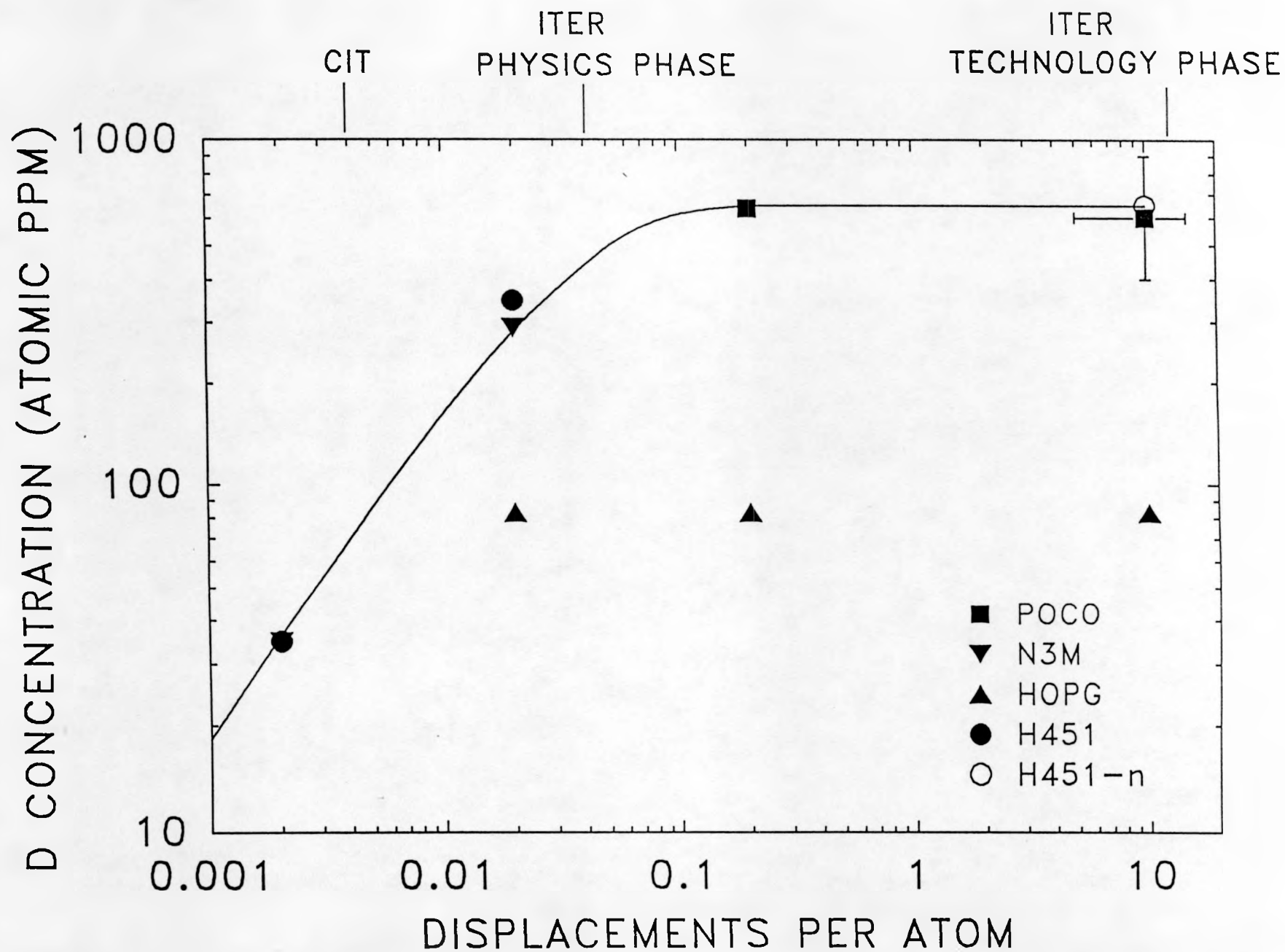
Figure 1

Figure 2

Figure 3

Figure 4