Deuterium depth profiling with $D(^3\text{He},p)\alpha$ nuclear reaction

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Abstract
The seminar is intended to describe the method of nuclear reactions $D(^3\text{He},p)\alpha$ to detect deuterium in materials. Following the presentation of the nuclear reaction used for the analysis I concentrate on the description of the concepts needed to calculate the proton yield. I present related cases of measurements at the tandem accelerator laboratory of Jožef Stefan Institute. The described examples of analysis include deuterium depth profiling in tungsten exposed to atoms of deuterium and $^3\text{He}$ microbeam analysis.
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1 Introduction

One of most critical issues in the construction of thermonuclear reactor based on magnetic confinement in the tokamak configuration is the construction of the inest wall. During plasma operation of such device, the wall is subject to severe physical conditions, a combination of neutron and charged particle bombardment, heavy and uneven thermal load photon irradiation and atomic hydrogen exposure (Figure 1). This leads to erosion, deposition, gas capture and lattice damage of the wall.

The build up of deposited layers also leads to fuel retention in the vessel walls as the deuterium or tritium fuel may be co-deposited along with the eroded materials. This is particularly true for carbon, which has a chemical affinity for hydrogen isotopes such as deuterium or tritium. While fuel retention is not a problem for present day tokamaks which are using deuterium plasmas, it will become an issue for next step devices, where the allowable vessel tritium inventory is limited, especially if carbon would be used as the dominant plasma-facing material. This is the main driver for developing the tungsten plasma-facing materials in view of the deuterium-tritium operation of International Thermonuclear Experimental Reactor (ITER). ITER walls will be made of Be, divertor zone of W. The most critical plasma striking zones of divertor wall will be made from carbon composites[1].

The use of carbon fiber composites (CFC), molybdenum and tungsten as plasma-facing materials in nuclear fusion research resulted in an increasing interest in quantitative profiling of deuterium in plasma-facing materials. Deuterium can penetrate these materials up to depths of several tens to several hundreds of µm, where it is trapped at low concentrations [2]. The assumption is that this deep penetration is an important process for the deuterium inventory in the tokamaks and other fusions reactors[3]. Study on temperature dependence of diffusion speed are plan to take place in the end of 2013 by Jožef Stefan Institute and Max-Planck-Institut für Plasmaphysik collaboration.

To keep within the ITER regulatory limit, the tritium inventory build up in ITER must be controlled. The build-up is extrapolated from present day machines where no serious inventory issues are foreseen if tungsten plasma-facing covers are used. Techniques for tritium removal and tritium inventory measurements are under active development since they could be needed in the case carbon plasma-facing covers are used during deuterium-tritium experiments.

The carbon materials penetration is related to the porosity of the material, while in tungsten the diffusion process is driven by the stress-field induced by implanted deuterium [2]. Poor understanding of these processes, and their investigation requires methods for quantitative depth profiling of deuterium up to depths of at least 10 (or better few tens) µm.

Figure 1: Plasma wall interaction scheme[1]
2 Methods of deuterium detection and depth profiling

In many cases, samples are from the walls of experimental fusion reactors. Consequently, the applied depth profiling method should be as insensitive to surface roughness as possible, prohibiting grazing incident or exit angles for the ion beam analysis methods, such as ERDA.

2.1 Thermal Desorption Spectroscopy (TDS)

In the Thermal Desorption Spectroscopy (TDS), the specimen is heated at a constant heating rate in a vacuum. The change in hydrogen partial pressure due to hydrogen release from the specimen is monitored as a function of temperature or time. By carrying out the TDS, desorption peaks corresponding to the trapping sites are visible in the obtained TDS spectrum, partial pressure vs. temperature curve. After acquiring several TDS spectrum at different heating rates, desorption energy for each trapping state can be determined. From this it is possible to deduce total inventory and binding energy of hydrogen isotopes[4]. However, no information on depth concentration profiles is obtained.

2.2 Secondary ion mass spectroscopy (SIMS)

This method allows us to do depth profiling of hydrogen isotopes up to a depth of about 100 µm [5]. However, it has some disadvantages as it is time consuming and not quantitative by itself. A quantification can be obtained by comparison with ion beam analysis results for the surface layer. For good accuracy the ion-beam analysed layer should be as thick as possible, which again raises the question of quantitative hydrogen isotopes analysis up to larger depths[5].

2.3 Elastic Recoil Detection Analysis (ERDA)

With ERDA (Elastic Recoil Detection Analysis) one detects lighter particles from a target sample that are recoiled by heavier incident ions from the probing beam. When a MeV He ion beam incident on a sample with H and D, the H and D in the sample will be scattered in the forward direction. Spectrometry of these forward recoiled atoms gives rise to the quantitative depth profiling of these species in the sample[6].

Main limitation of this method is that it can be used only in thin films and not to thick samples. This results also in high sensitivity on the surface of the samples if proper geometry is used. Otherwise, it produces quantitative depth concentration profiling of deuterium.

2.4 Neutron-ERDA (NERDA)

NERDA with incident 14 MeV neutrons beam from the sample back side allows depth profiling of hydrogen isotopes up to depths of hundreds of µm [2]. The principle of measurements and analyses is similar to normal ERDA, except the use of neutron instead of heavy ions. However, the depth resolution of this method is only a few tens to about 100 µm [2] and counting statistics is poor due to the low incident neutron fluxes, resulting in low sensitivity.

2.5 Nuclear reactions resonance method (NRA)

The \(D(d,p)T\) and the \(D(^3He,p)α\) reactions can be used for quantitative depth profiling of D up to large depths. The \((^3He,p)\) resonance method, exploits the broad resonance of this reaction, Figure 3 by measuring the total proton yield as function of incident energy and a calculation of deuterium depth profile becomes possible [2][7]. The resonance method uses only the total number of detected protons and can be utilized with detectors having large solid angles.

However, due to the broad resonance with a FWHM of 730 keV[2] and the non-negligible contribution of protons originating from outside the resonance region this method requires measurements at 8-10 different energies, thus being time-consuming; it requires good counting statistics, which largely compensates the advantage of a large detector; and it is often difficult to show that the derived depth profile is unambiguous.

The paper concentrates on the NRA method. It is used by several groups to create quantitative depth profiles of deuterium, some also up to very large depths (∼ 100µm). And it is also in use at Jožef Stefan Institute.
3 Nuclear reactions

During the measurement, a beam of $^3$He ion (1+ or 2+) is directed to the sample containing deuterium. The goal is to determine how much deuterium is accumulated in the sample and its depth distribution. Incident helium ions are elastically scattered in the sample, as well as induce nuclear reactions on deuterium nuclei

$$^3_2\text{He} + ^1_1\text{D} \rightarrow ^4_2\text{He} + ^1_1\text{p} \quad Q = 18.35 \text{ MeV} \quad (1)$$

$$^3_2\text{He} + ^1_1\text{D} \rightarrow ^1_1\text{p} + ^0_0\text{n} + ^2_2\text{He} \quad Q = -2.73 \text{ MeV}$$

$$^3_2\text{He} + ^1_1\text{D} \rightarrow ^2_1\text{D} + ^1_1\text{D} + ^1_1\text{p} \quad Q = -6.52 \text{ MeV}$$

$$^3_2\text{He} + ^1_1\text{D} \rightarrow ^1_1\text{T} + ^1_1\text{p} + ^1_1\text{p} \quad Q = -1.95 \text{ MeV}$$

In addition, helium ions can also interagate with carbon atoms in sample, if sample is based on CFC, which can be described by the nuclear reaction in which protons are produced,

$$^3_2\text{He} + ^{12}_6\text{C} \rightarrow ^{14}_7\text{N} + ^1_1\text{p} \quad Q = 3.96 \text{ MeV} \quad (2)$$

$$^3_2\text{He} + ^{13}_6\text{C} \rightarrow ^1_1\text{p} + ^{15}_7\text{N} \quad Q = 9.85 \text{ MeV}$$

All these reactions produce various products, which are then detected in the detection set-up. Usually high-energy protons are detected, which are best suited for measurements, as they are charged and lose small amount of energy when passing through the sample material. From the measured data and properties of these nuclear reactions (kinematic relations and scatter cross-section) it is possible to say something about the depth profile of deuterium in the analysed layer.

It should be noted that nuclear reactions (1) and (2) are not the only possible mode of interaction of ions with the helium atoms in the sample, since the incident helium ions are elasticity scattered on nucleus in target

$$^3_2\text{He} + ^2_1\text{D} \rightarrow ^2_2\text{He} + ^2_1\text{D}$$

$$^3_2\text{He} + ^{12}_6\text{C} \rightarrow ^{12}_6\text{C} + ^2_2\text{He} \quad (3)$$

$$^3_2\text{He} + ^{13}_6\text{C} \rightarrow ^{13}_6\text{C} + ^2_2\text{He}$$

These reactions in terms of depth profiling of deuterium are forming a low-energy background in the measurement. Problem is solved by placing protective film (usually Mylar or Clapton) in front of the detector. Fast protons pass, while scattered helium ions are stopped in the foil.

For practical experiments and measurements is mostly used first of the given nuclear reaction $D(^3\text{He}, p)\alpha$. Consequently, we will focus on this reaction and its application in practice. From now on if we write $\text{He}$ it means $^3\text{He}$. Due to practical reasons, we detect fast protons from the first of given $D(^3\text{He}, p)\alpha$ reactions, as they are fast and could be easy separated from other detected particles.

4 Reaction kinematics

For the beginning we must calculate amount of energy $Q$ released in reaction $D(^3\text{He}, p)\alpha$, which is defined as the difference of kinetic energy initial and final stats:

$$Q = T_f - T_i = c^2(M_i - M_f) = c^2(m_{He} + m_D - (m_\alpha + m_p + 2m_e)) =$$

$$= (2808.9 + 1876.1246 - 3727.38 - 938.27 - 2 \times 0.511)\text{MeV} = 18.3526\text{MeV} \quad (4)$$

From here we see that the reaction is exothermic ($Q > 0$), which means that the initial state will always get the selected final state, no matter what kind of energy the incident helium ion. After the formation of new particles, this energy is converted into kinetic energy of the reaction products (in this case the proton and $\alpha$ particle) in center-of-mass system.
We are interested in how the kinetic energy of the proton depends on the energy of the incident $^4$He ion. To this end, we solve the kinematic equations for a given nuclear reaction in the laboratory system\cite{7}:

\[
E_{He} + E_D = E_p + E_\alpha \\
p_{He} + 0 = p_p \cos \theta + p_\alpha \cos \phi \\
0 = p_p \sin \theta - p_\alpha \sin \phi
\] (5)

where $\theta$ and $\phi$ are angles in the laboratory system, under which proton and a particle $\alpha$ fly after reaction is finished. The $p_\alpha$ stands for the size of momentum three vectors of particles that appear in the reaction. When these equations are solved for the outgoing proton momentum\cite{7}, the result can be written as

\[
p_p = \frac{E_{12}^2 p_{He} \cos \theta \pm \sqrt{E_{12}^2 - 4m_p^2 (E_{12}^2 - p_{He}^2 \cos^2 \theta)}}{2(E_{12}^2 - p_{He}^2 \cos^2 \theta)}
\] (6)

where $E_{12}$ and $\overline{E}_{12}$ indicates

\[
E_{12} = m_D + \sqrt{p_{He}^2 + m_{He}^2} \\
\overline{E}_{12} = E_{12} - p_{He}^2 + m_{He}^2 - m_\alpha^2
\] (7)

The expression (6) has two solutions in general, but it turns out that the only physical one, where we choose positive sign before the second article in the numerator. The kinetic energy of the proton in lab system is then equal to\cite{7}

\[
T_p = \sqrt{p_p^2 + m_p^2 - m_p}
\] (8)

How $T_p$ depends on the kinetic energy of incident helium ions and the scatter angle $\theta$ is shown in the Figure 2. We can see that protons have incurred much higher energy then incident helium ions. This is due to the high $Q$, which is released in this reaction. Since the $\alpha$ particle is much heavier than the proton, proton takes the most of the energy. We obtain slow $\alpha$ particles and fast protons. From the resulting dependence of proton kinetic energy is also seen that the maximum proton kinetic energy when the proton is emitted in forward direction, and the lowest when it is emitted backwards.

Figure 2: The kinetic energy of protons, resulting from a nuclear reaction $D(^3He, p)\alpha$ as a function of the scatter angle $\theta$ and energy of incident helium ions\cite{7}.

Our detector is positioned at 135°. Higher incident energy of incident $^3He$ result in lower detected energy of proton from $(^3He, p)$ reaction.
5 Scattering cross-section

Because of the nature of nuclear reactions, i.e. the complex structure of matter and the forces which are in play in this reaction, the derivation of the scatter cross section is very complicated. Therefore, we rather lean to the experimental data. These were measured by numerous authors V.Kh. Alimov [8], W. Möller [9] and T.W. Bonner [10]. Which are otherwise given for different systems but all can be transformed in centre of mass system[7].

Figure 3: Differential scattering cross section for the reaction $D(^3\text{He},p)\alpha$ the center of mass coordinate system. The points on these graphs are shown the measured scattering cross sections, which are measured by Alimov, Möller, Bonner. In addition to the full line marked phenomenological approximation of Möller and Besenbacher [7].

It turns out that the scattering cross-section shape is resonant, as shown in Figure 3. At this point it is also worth to point out that the bell-shaped ("resonance") scatter cross-section shape is essential in profiling deuterium in the matter, because, as we shall see below, allows as to differentiate particles which are scattered at different depths in sample.

6 Energy loss and stopping power

When scattering projectiles (in our case $^3\text{He}$ ions) does not takes place on the surface of the sample, but rather deeper in the target, we have to consider that a particle loses a fraction of the energy on the path to the target nucleus with which will interact. Energy loss of the incident particle is due to particle Coulomb interaction with electrons in electron clouds on its path. Loss of energy per unit of travelled path of the particle is described by the stopping power $S(E)$, which is defined as,

$$S(E) = -\frac{1}{\rho} \frac{dE}{dz} \quad (9)$$

Energy dependence of the stopping power in the first approximation is described by Bethe-Bloch function [6], in general, is its dependence on energy is much more complex and we can not accurately calculate. In practice we use the average measured values by any combination of the projectile and the target, they can be found collected in the tables.
Figure 4: $^{3}He$ ions energy at different depths and initial energies in a target of 100% W, calculated with SRIM 2008 code [11].

The result of stopping power is that $^{3}He$ ions have only finite range in the selected target, display the Figure 4.

Figure 5: The graph shows maximum range of $^{3}He$ ions in selected target (tungsten with some proportions of deuterium) at different energy, calculated by SRIM 2008 code [11].
7 Proton yield

So far we have developed and describe all the mechanisms and processes that occur in scattering of $^3He$ nuclei on the nuclei of deuterium. In the experiment a beam of helium ions with energy $E_0$ is hitting thick layered sample under incident angle $\alpha$, scattered particles at an angle $\psi$ (see Figure 6). Scattering particles angle is $\theta = \pi - (\alpha + \psi)$. If we divide the sample into a series of thin layers and we can ask, how many particles, that are generated in a given layer, enter the detector. The number of detected particles from the selected layer is donated by target yield $dY$ and depends on the cross-section $\frac{d\sigma_i}{d\Omega}$ of the selected nuclear reaction in the core layer selected, the solid angle $\Delta \Omega$ of the detector, the particles flux $j_p$ and the chemical composition of the individual layers (density, atomic mass and mass elements in layers). Since the treated layer consists of several types of atoms (carbon and deuterium), measured yield of each element in the layers separately[7]:

$$dY = \frac{d\sigma_i(E(E_0, z))}{d\Omega} \Delta \Omega j_0 \frac{\rho A_i}{M_i} C_i(z) Adz$$

(10)

where we denoted by $C_i$ concentration of selected element in the layers. And $i$ runs through all the elements in the layer at which we are interested in. If you now introduce $Q_{iHe} = j_0 \frac{\rho N_A}{M_i} A$ in considering that $dz = dx \cos \alpha$ we can write yield of thin layer as:

$$dY = Q_{iHe} \frac{C_i(x)}{\cos \alpha} \frac{d\sigma_i(E(E_0, z))}{d\Omega} d\Omega_{det} dx$$

(11)

where $d\Omega_{det}$ is the fraction of the solid angle of the detector, which overlooks the particles flying from the sample at an angle $\approx \psi$.

Figure 6: Scattering geometry of the experiment. Incident projectile energy is $E_0$, the energy just before the nuclear reaction $E'$, energy just after nuclear reaction $E''$, and the output energy from target is $E$. With $\alpha$ denoted the angle of the incident particle beam, $\psi$ is out going angle and $\theta$ is scattering angle of a particle. $\delta \Omega$ is a small part of the solid angle, which dislodged particles are scattered at an angle $\theta$[7]

Total yield per unit of time in selected sample under investigation is computed by integrating expression (11)
over the entire thickness of the layers and the entire solid angle of the detector. At the end this is resulting in

\[ Y_i(E_0, L) = \frac{Q^3He}{\cos \alpha} \Delta \Omega_{det} \int_0^L C_i(X) \frac{d\sigma_i(E(E_0, z))}{d\Omega} dx \]  

(12)

It is important to point out that in the derivation of the proton yield we assumed to have a homogeneous mixture of deuterium and carrier material (CFC or W) that is without crystal structure. The reason for this lies in the fact that programs which are used for analysis of the measured data, in their Monte Carlo calculation do not take the crystal structure as parameter [11] [12].

8 Experimental measurements

In the experiment, the beam of incident helium ion is focused on the given sample and we are measuring the energy distribution of the proton yield as a function of proton energy. This relationship is obtained by dividing \( \frac{dY_p}{dE_p}(E_p) \) by energy dependence scatter cross section, we can express from the equation (11)[7]:

\[ \frac{1}{Y_0} \frac{dY_D}{dE_p}(E_p) \frac{1}{d\sigma(E_p)} = C_D(E_p) \]  

(13)

Since we are interested in concentration as a function of position \( x \) in the layer and not from the energies \( E_p \), we must overwrite (13) in the appropriate dependency.

Therefore, we must take into account the proper stopping power to make a connection between the energy \( E_p \) and the position of the particle in the layer. Since the stopping power is dependent on the concentration of all the elements in the layer, it is not easy problem. Therefore, the concentration profile is determined by an iterative procedure.

To be able to deduce the concentration of deuterium from the measured \( Y_p \), we must deconvolute expression (12). We may do this by a suitable program (i.e. simNRA[12]) for simulated concentration profiles, to seek out appropriate energy dependence yields and trying to adjust them to the measured points.

Figure 7: The energy distribution yields \( \frac{dY_p}{dE_p} \) as a function of the kinetic energy of protons detected at different energies of incident \(^3\)He ions for \( d = 10 \ \mu m \) thick layer of carbon containing 7% of deuterium, which is evenly distributed over the layers[7].
9 Some measurements and results from Jožef Stefan Institute

Measurements are executed in a vacuum chamber in one of the experimental lines in the Jožef Stefan Institute tandem accelerator.

Measurements are carried out as follows: on each sample, 5 measurements at different energies of the He beam are executed: 4.2 MeV, 3.3 MeV, 2.5 MeV, 1.5 MeV and 0.74 MeV. We measure the energy distribution of protons emitted at each of 5 incident energies. In this case we obtain spectrum seen on figure 8 for one energy.

Figure 8: The spectrum of fast protons, obtained by $^3$He beam at energy of 1.5 Mev, measured at Jožef Stefan Institute.

The resulting spectra are then processed with the program simNRA. The calculated yields of a model build target are compared with the results of measurements and a depth distribution of deuterium is determined. The result is seen on figure 9.
10 Upgrade of classical NRA at Jožef Stefan Institute

In the laboratories of the Jožef Stefan Institute (JSI) beside of common broadbeam NRA, a so-called μNRA is developed. In this method, the beam of $^3$He ions is focused through quadrupole lenses on the approximate size of $4 \times 4 \mu m^2$, which at energy of 4.5MeV penetrate in to tungsten target approximately $4 \mu m$. This enables a good lateral resolution, which provides information on the lateral distribution of deuterium in the walls of fusion reactors. Due to experimental set-up, namely Al foil in front of the detector and measuring at one beam energy we loses all information on the depth distribution of deuterium.

During the measurements, in total four detectors are use simultaneously: an NRA PIPS detector, an RBS PIPS detector, an HPGe X-ray detector[13] for 3-helium-induced X-ray emission (3HIXE) and additionally a PIPS detector for beam dose normalization that detects ions scattered from the beam chopper[13], shown on Figure 10. Each detected event in the set of detectors is record and save in a list mode together with information on the beam position at the moment of the event. On the basis of these measurements were obtained the following results shown in Figure 9. Where shown part of elemental inventory of one element of limiter from Tore Supra tokamak.

Conclusion

In the seminar I tried to demonstrate the use of nuclear reaction $D(^3He,p)\alpha$ for determining the concentration of deuterium in the sample. At the same time I presented importance of this method for the determination of deuterium deposition in the walls of fusion reactors. Allowing us to better predict the operation of a new generation of fusion reactors, for example ITER.

I demonstrated the use of traditional methods and its upgrade mNRA. And present some measurement results to Jožef Stefan Institute.
Figure 10: NRA detection set-up for deuterium analysis with a $^3$He focused beam at the JSI microprobe. The three detection systems, NRA PIPS detector, RBS PIPS detector and HPGe X-ray detector for 3-helium-induced X-ray emission (3HIXE) were all positioned at an angle of $135^\circ$ with respect to the beam direction[14].

Figure 11: Qualitative mapping of the castellation gap of the limiter tile in the erosion zone (Tore Supra DITS experiment, Gap F26T9 Q3L1) with micro-NRA (D) and 3HIXE (Fe, Cr, Ti, Cu). To cover the total depth of the gap of 6 mm, four frames of $1840 \times 1840 \mu m^2$ size were measured, each consecutively shifted $1500 \mu m$ along the gap depth axis, and merged into a single gap image [14].

References


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