

HIGH SENSITIVE DEUTERIUM MEASUREMENTS

Alexandru Răzvan PETRE¹, Doru Gheorghe PĂCEŞILĂ¹

The paper presents results of the Accelerator Mass Spectrometry (AMS) depth profile measurements of deuterium concentrations in protection tiles of fusion facilities. Simulations of deuterium trapping in sandwich like deposition layers were performed in the laboratory by use of the pulsed laser deposition method. The obtained results are relevant to the fusion discharges in Tokamak reactor.

Keywords: Deuterium, simulations of material deposition, AMS, depth profiling.

1. Introduction

An old dream of humanity was to produce a never exhausting energy source like that of our Sun. It was rapidly recognized that such an enormous energy is produced by the fusion reactions of hydrogen atoms and that the necessary conditions to obtain such a regenerable energy source are high temperature and huge pressure (approx. 10^{16} N/m²) preserved in the inner part of the Sun. Unfortunately, similar pressure values, capable to produce the fusion of atoms, can not be attained on Earth. So far, the best-achieved magnetic configuration to perform a controlled fusion reaction is the Tokamak system [1]. Today, to finalize the construction of the first International Thermonuclear Experimental Reactor (ITER) in Cadarache-France [2, 3] a high and very special interest for the research of fusion facilities exists in all their functioning details. ITER will be the first nuclear reactor based on fusion.

The Accelerator Mass Spectrometry (AMS) is a high sensitive analyzing method capable of measuring particles produced in the fusion reactions, or those accelerated in the Tokamak plasma, and subsequently bombard the protection tiles of the fusion reactor vessel. The AMS method is well known for a wide range of applications in many other domains, from environmental physics [4-7] to medicine [8-12], geology [13-16] and many more [17-22]. The AMS experiments have been designed to measure the amount of fuel (D and T) retained on the new tiles [23]. Such experiments could localize the plasma disruption phenomenon and determined the plasma confinement stability during the discharges in the Tokamak. The properties of wolfram coating of carbon tiles (practiced against fuel retention) were tested.

¹ National Institute for R&D in Physics and Nuclear Engineering, Horia Hulubei, Romania, e-mail: alexpetre@nipne.ro

This paper describes, in the second section, a short introduction to the Tokamak physics. The third section describes the obtained results of AMS analyzing experiments performed in the frame of the project described in this paper.

2. The Tokamak concept and the basic of fusion experiments

The name Tokamak comes from the Russian words that mean “Toroidal Chamber with Magnetic Coils” and it performs the confinement of plasma (T, D, H-gas) in order to produce the fusion of the light nuclei. For this purpose, a big magnet called transformer, is electrically charged by increasing the current flow through its primary windings. Then, after pre-filling the vacuum vessel of the Tokamak with H, D or/and T gas, some of the atoms will be rapidly ionized. Finally, ramping down the current it will discharge the transformer and the self-induction effect will produce a large electric field gradient around the initial vertical magnetic field (produced by the transformer). The few charged particles will be accelerated and by collisions with other particles will produce further ions, creating the high-ionized plasma in the Tokamak torus. The confinement of plasma is obtained in the following way: the plasma current produces its own magnetic field (poloidal) which is “vector composed” with the toroidal magnetic field (produced by the toroidal coils) and the result will be a magnet field with twisted lines, forming the magnetic cage that squeezes the ions together, producing the necessary confinement for fusion.

In order to maintain a fusion reaction, the Lawson Criterion has to be fulfilled. As the British scientist D. J. Lawson formulated it in 1955 [24] this criterion requires that the following plasma conditions need to be simultaneously achieved: plasma temperature (T): 100-200 million Kelvin, energy confinement time (τ_E): 4-6 seconds; central density in plasma (n_e): $1-2 \times 10^{20}$ particles m^{-3} (approx. $1/1000$ gram m^{-3} , that is one millionth of the density of air).

A significant part of the energy is lost by radiation. On one hand, this energy loss has to be recovered by additional heating and on the other hand, one has to protect the walls from the very hot plasma core. It is a twofold problem that requires a good confinement and the supplementary heating of the plasma.

About 20 years ago, the revolutionary Divertor concept [25] was introduced. Since then, the magnetic field lines at the plasma edge were modified, so that the magnetic field lines are diverted downwards into a dedicated region at the bottom of the Tokamak vacuum vessel. There, the plasma exhaust ends up in collisions with the special target plates called Divertor plates. In 1951, the initial idea was to use a divertor for isolating the point of first contact between the plasma and the vessel wall and to allow the removal of the helium ash (Spitzer 1951 [26]). The compensation of the energy loss also requires higher injected energy and is

provided by three auxiliary plasma-heating systems at the Axial Symmetric Divertor Experiment (ASDEX-Upgrade) [27].

The Neutral Beam Injection (NBI) is based on the injection of powerful beams of neutral ions into the preheated plasma [28, 29]. When introduced into the plasma, they lose electrons due to multiple collisions, get ionized and, as a consequence, are captured by the “magnetic cage” of the Tokamak. After a series of subsequent ion-ion, ion-electron and electron-electron collisions, the group velocity of the new ion beam is transferred into an increased mean velocity of the entire plasma.

Finally, in order to withstand the extreme heat current fluxes, the Tokamak has been using carbon fiber composite (CFC) tiles. The non-metal material proved to be perfectly resistant to heat loads, but unfortunately, it showed a strong affinity for hydrogen. In order to avoid the fuel retention at JET a new lining was introduced, called the “ITER-Like Wall, based on beryllium and tungsten coated carbon tiles. The plasma interaction with the inner wall causes erosion and formation of deposited/co-deposited layers, mixed materials and spatially non-uniform fuel retention. As we will show in this paper, AMS has contributed to such a research.

3. Measurements and results

Useful applications for the Tokamak are the determination of the amount of different fuel elements implanted into and diffusing inside the Plasma Facing Components (PFC) of the reactor vessel. To investigate such processes depth profiling of the element concentration (DP) by use of AMS was applied in previous researches [30]. The samples for the AMS analyze were cut in small pieces from the large protection tiles of the Tokamak vessel (not easy to access). An useful and easy to handle alternative way, was to use small “spy” samples made of pyrolytic carbon placed in-between the protection tiles of the vessel.

LTS samples are more efficient for the AMS. They do not depend on the structure or on the material type of the tiles (can be used between the Be and W tiles), they have an uniform and compact internal structure and a flat mirror-like surface providing unperturbed information of the energy and concentration of the implanted particles.

The AMS depth profiling (AMS-DP) is performed by sputtering the sample material in the ion source by the aid of the Cs beam, followed by the measurement of element concentration in dependence of time. The time scale can be later converted to depth value if one performs, at the end of the in-beam experiment, an optic profilometry that determines the total crater depth. Practically, each AMS facility can be adapted for depth profiling [31 - 33]. The elimination of the rim effect is the most difficult part of the procedure.

The applied solutions to remove the crater edge effects are to move the target or the target holder in front of the Cs^+ - sputter beam in the ion source. When the Cs^+ ion beam is sputtering on the crater walls, the data acquisition system ignores the secondary ions. In comparison to a static ion bombardment, both the target movement and the beam sweeping produce a tremendous increase of the measurement time. After careful experimental investigations, a mathematical unfolding procedure was promoted [31, 34] that corrects the DP for the perturbing contribution produced by the secondary ions that are sputtered from the sidewalls of the produced crater

The mathematical unfolding procedure uses the crater dimensions provided by the optic profilometry. In this way all DP spectra are corrected, and the speed of depth profiling was highly increased and can also be varied, according to the required depth resolution, between 5 and 250 $\mu\text{m}/\text{h}$. As a consequence, several applications of AMS depth profiling became possible for the fusion experiments [35 – 42].

Depth profiling of material concentrations also requires standard samples that must correspond, in structure, to the material under investigation. Thus, the host materials should have the same atomic matrix and the calibration of standards is not always a simple task (e.g. T/ C, T/Si, T/W, T/Be, D/C etc). The measured data are expressed in units of ion concentration, in atoms/ cm^3 and can be easily converted using the optic profilometry to the bulk inventories expressed in atom/ cm^2 . The overall errors in our measurements were in the range of 30%.

In the Tokamak testing reactors, deuterium is present as main component of the gas fuel, but it is also introduced in the reaction vessel as an accelerated neutral atomic beam produced by the NBI systems to increase the energy of the plasma. It can also participate to charge exchange reactions with the materials of the PFC. For all these reasons, deuterium will exist in the fusion reactor vessel with different energies.

Fig. 1 presents the deuterium DP distributions measured from the LTS placed between protection tiles of the internal wall, of the ASDEX-Upgrade Tokamak [43]. The positions of the LTS were placed far from the NBI systems. The concentration oscillations in the DP distributions were caused by depositions of different materials that occur regularly in the Tokamak vessel.

Frequently, the deposited hydrogen isotope is captured under or in-between of deposited layers of other materials. A sandwich-like structure will be formed and, correspondingly, deuterium will be measured at larger depth values that it could not penetrate by its own thermal energy.

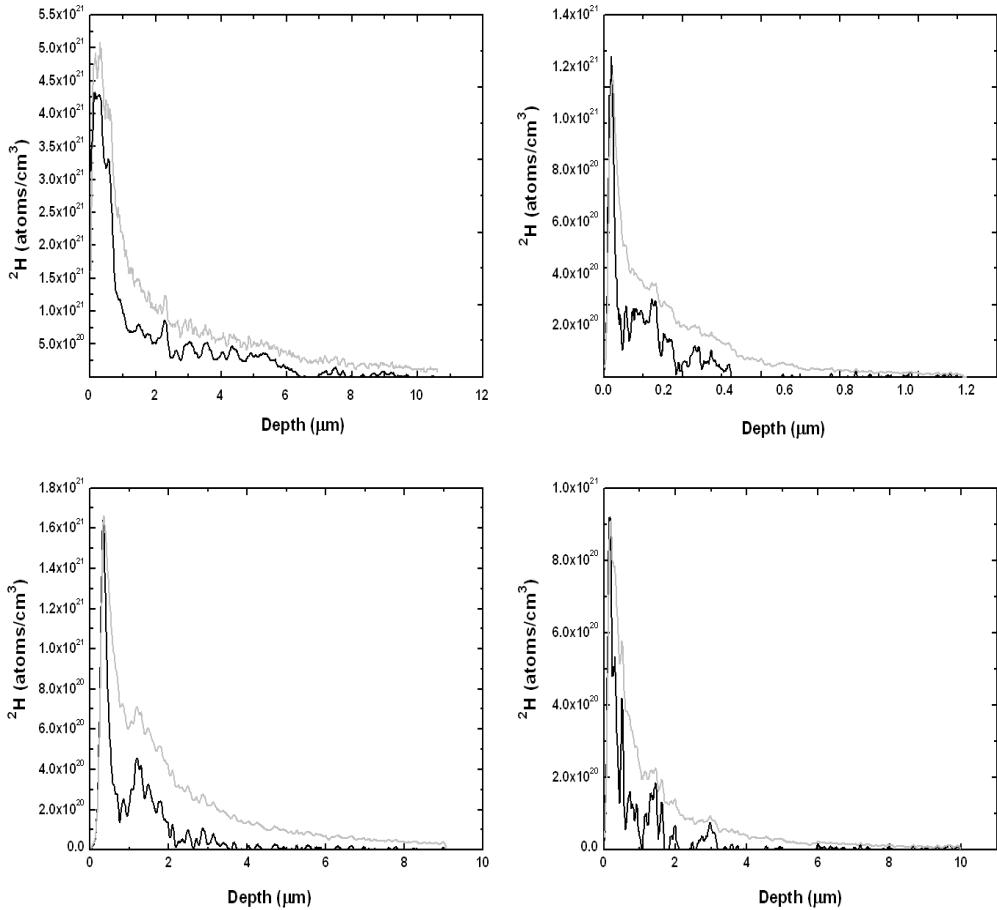


Fig. 1. Deuterium AMS depth distributions measured from LTS. The upper curves represent the initial measured AMS concentration depth distributions and the lower curves represent the values corrected for the crater rim effects.

A complete toroidal distribution measured for the accumulated deuterium inventory in the inner vessel wall of the ASDEX-Upgrade is shown in Fig. 2. The initial AMS measured values expressed in atom/cm³ were multiplied with the depth interval to provide the integrated values (atoms/cm²) of the accumulated deuterium. Our measurements were calibrated with standard samples of precise D/C concentrations and were corrected for the crater rim effect.

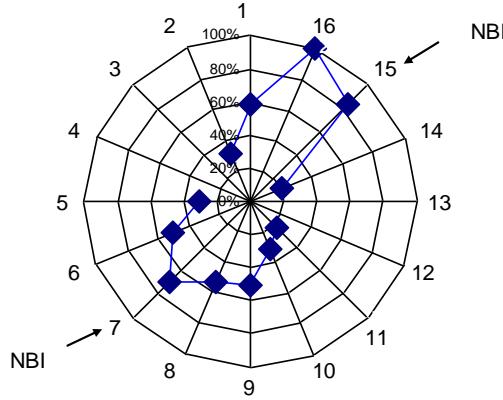


Fig. 2. Integrated toroidal distribution of deuterium. The depth profiles were integrated up to a depth of 2.5 μm . The measurements were performed on LTS placed on the inner wall of the vessel in between the protection tiles.

The two large peaks around sector no.7 (injection energy 60 keV) and sector no.15 (injection energy 140 keV) show that part of the neutral deuterium beam is not interacting or is not stopped by the confined plasma. Deuterium neutrals fly through the plasma and are stopped in the protection tiles of the inner wall. The density of the plasma is low and the beam energy is sufficient high to reach the plasma center. However, in a small extend, it also escapes on the opposite side. Therefore, for total energy absorption, AMS can optimize the conditions.

Many simulations and tests are performed only by the use of deuterium since no radioactivity will be involved. An important issue studied with deuterium is erosion and the subsequent deposition occurring on the protection tiles.

In our experiments, an important issue was to perform experimental simulations in the laboratory for fuel retention on different materials during continuous deposition of other materials. Such a scenario is characteristic for the processes developed under the envelope of a fusion reactor vessel. Material displacements, produced by heat erosions in certain parts of the vessel, are transported along the magnetic field lines and redeposit elsewhere.

The samples had sandwiches like structures and were produced by laser ablation, using the Radio Frequency assisted Pulsed Laser Deposition (RF - PLD) method [44]. For comparison, one graphite sample was deposited with a thin layer (100nm) of tungsten (W) and was then exposed to the RF- PLD of deuterium. The second graphite sample was exposed directly to the deuterium deposition. Fig. 3 presents the results of our AMS measurements of the deuterium concentration depth profile in the two samples. The spectra were overlapped to show the insignificant effect of the tungsten coating.

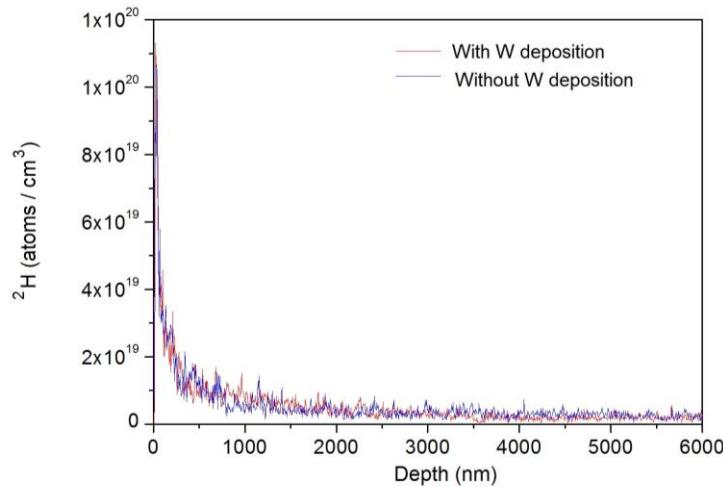


Fig. 3. Overlapped spectra of AMS depth profiles of deuterium in the two samples employed.

This experimental simulation has shown again that thin depositions of tungsten layers cannot stop the retention of the fuel deposition and penetration into the protection tile of a fusion reactor.

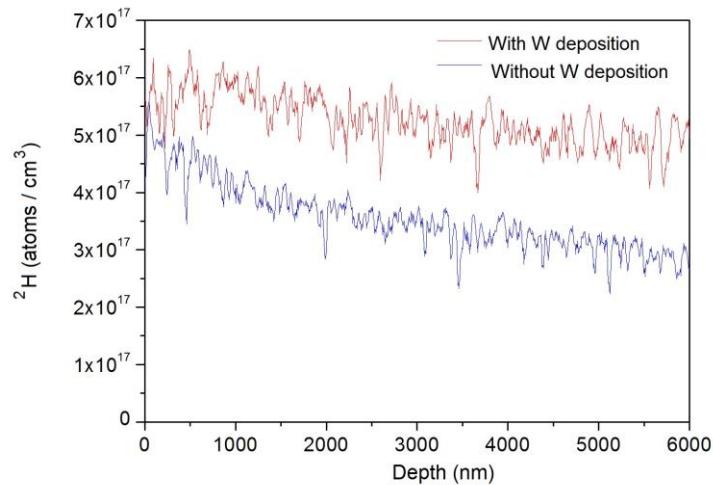


Fig. 4. Absorption of deuterium gas at room temperature in two graphite samples: one with a W deposition layer and the other with any deposition layer.

In fusion reactors, it has been observed that deposition followed by diffusion substantially contributes to the fuel retention in the protection tile of the reactor vessel. Thus, we performed a simulation of the two graphite samples (with and without W deposited layer) that were exposed to deuterium gas at room temperature, with a gas density of 5.72×10^{17} atoms/cm³. Fig. 4 shows the depth

profiles of deuterium concentrations by gas absorption in samples with and without tungsten deposition.

Obviously, the absorption is higher for the sample deposited with the tungsten layer. The result is in accordance with the simulation presented above, indicating again that the high permeability of a thin tungsten layer deposited over the uneven surface of the graphite substrate. This initial solution adopted by the fusion experiments was changed using solid blocks of tungsten to build the protection tiles in regions of high material depositions and erosion rates.

3. Conclusions

More than twenty years ago, it was established that AMS is an analyzing method able to perform sensitive measurements of tritium. At that time most applications were performed for medicine. More recently, this method has been motivated by the increasing interest for the construction of fusion reactors and AMS was applied to the analysis of the hydrogen isotopes retained in materials, by measuring the depth profile of their concentrations.

This paper presents AMS results of deuterium retention detection in tiles delivered for fusion reactors. The depth profile measurements provided information of the neural beam injection efficiency used in fusion reactors for supplementary energy input and compensation. Experimental simulation has demonstrated that a thin tungsten layer deposited on the surface of a graphite substrate is highly permeable and cannot be used in fusion reactors.

R E F E R E N C E S

- [1]. *B. D. Bondarenko*, Phys. Usp. 44 (2001) 844.
- [2]. www.iter.org
- [3]. http://www.tercad.org/images/actualites/2014/CARLOS%20_VINON%20JULY%202014%20V2.pdf
- [4]. *C. Stan-Sion, M. Enachescu, A.R. Petre*, AMS analyses of I-129 from the Fukushima Daiichi nuclear accident in the Pacific Ocean waters of the Coast La Jolla - San Diego, USA Environ. Sci.: Processes Impacts 17(2015) 932-938.
- [5]. *L.J. Hainsworth, A.C. Mignerey, G.R. Helz, P. Sharma, P.W. Kubik*, Modern chlorine-36 deposition in southern Maryland, U.S.A, Nucl Instr Meth B 92(1-4) (1994) 345-349.
- [6]. *H.-A. Synal, J. Beer, G. Bonani, M. Suter, W. Wölfl*, Atmospheric transport of bomb-produced ^{36}Cl , Nucl Instr Meth B 52 (3-4) (1990) 483-488.
- [7]. *C. Stan-Sion, M. Enachescu*, Applications of AMS in Bucharest for detecting nuclear pollution, Romanian Journal of Physics (2014) 59 (9-10) 930-940.
- [8]. *J.S. Vogel*, Accelerator mass spectrometry for quantitative in vivo tracing, Biotechniques, Suppl (2005) 25-29.
- [9]. *J.S. Vogel, A.H. Love*, Quantitating isotopic molecular labels with accelerator mass spectrometry, Methods Enzymol 402 (2005) 402-422.
- [10]. *J. S. Vogel, J. Mc Aninch, Stewart P.H.T. Freeman*, Elements in biological AMS, Nucl Instr Meth B 123, (1-4) (1997) 241-244.

- [11]. *Mihaela Enachescu*, Accelerator mass spectroscopy measurements of the deuterium inventory in the Tore Supra toroidal limiter, *U.P.B. Sci. Bull., Series A*, 79(4) (2017) 337-348.
- [12]. *Karen Brown, Karen H. Dingley, Kenneth W. Turteltaub*, Accelerator Mass Spectrometry for Biomedical Research, *Methods in Enzymology* 402 (2005) 423-443.
- [13]. *K. Nishizumi, C.P. Kohl, J.R. Arnold, J.R. Dorn, J. Klein, D. Fink, R. Middleton, D. Lal*, Role of in situ cosmogenic nuclides ^9Be and ^{26}Al in the study of diverse geomorphic processes, *Earth Surf. Processes and Landforms* 18 (1993) 407-425.
- [14]. *C. Stan-Sion, M. Enachescu*, AMS with light nuclei at small accelerators, *AIP Conference Proceedings*, Volume 1852, Issue 1, id.060004, 10.1063/1.4984868.
- [15]. *J.L. Repka, R.S. Anderson, R.C. Finkel*, Cosmogenic dating of fluvial terraces, Fremont River, Utah, *Earth Planet. Sci. Lett.* 152(1-4) (1997) 59-73.
- [16]. *B. Heisinger, M. Niedermayer, F.J. Hartmann, G. Korschinek, E. Nolte, G. Morteani, S. Neumaier, C. Petitjean, P. Kubik, A. Synal, S. Ivy-Ochs*, In-situ production of radionuclides at great depths, *Nucl Instr Meth B* 123 (1-4) (1997) 341-346.
- [17]. *C. Stan-Sion, M. Enachescu, A.R. Petre, C.I. Calinescu*, AMS measurements of trace levels of boron in graphite, *Anal. Methods*, 6 (2014) 5808-5811.
- [18]. *G. Korschinek, A. Bergmaier, T. Faestermann, U.C. Gerstmann, K. Knie, G. Rugel, A. Wallner, I. Dillmann, G. Dollinger, Ch. Lierse von Gostomski, K. Kossert, M. Maiti, M. Poutivtsev, A. Remmert*, A new value for the half-life of ^{10}Be by Heavy-Ion Elastic Recoil Detection and liquid scintillation counting, *Nucl Instr Meth B* 268(2) 2010 187-191.
- [19]. *C. Bronk Ramsey*, Development of the radiocarbon calibration program, *Radiocarbon* Vol 43 (2A) (2001) 355-363.
- [20]. *C. Stan-Sion, A. Letourneau, H. Reithmeier, V. Lazarev, M. Enachescu, E. Nolte*, AMS measurement of the neutron capture cross-section $^{209}\text{Bi}(\text{n},\gamma)$ ^{210}mBi , *Nucl Instr Meth B*, 259 (2007) 739-744.
- [21]. *S. M. Kristiansen, K. Dalsgaard, M.K. Holst, B. Aaby, J. Heinemeier*, Dating of prehistoric burial mounds by ^{14}C analysis of soil organic matter fractions, *Radiocarbon* 45(1) (2003) 101-112.
- [22]. *S. Oliver, S. Bajo, L.K. Fifield, H.W. Gaeggeler, T. Papina, P.H. Santschi, U. Schrotter, M. Schwikowski, L. Wacker*, Plutonium from global fall out recorded in an ice core from the Belucha Glacier, Siberian Altai, *Environmental Science and Technology* 38 (2004) 6507-6512.
- [23]. *C. Stan-Sion and N. Bekris and G. Kizane and M. Enachescu and J. Likonen and M. Halitovs and A. Petre and JET contributors*, Tritium retention measurements by accelerator mass spectrometry and full combustion of W-coated and uncoated CFC tiles from the JET divertor, *Nuclear Fusion* 56(4) (2016) 046015.
- [24]. *J. D. Lawson*, (Technical report). Atomic Energy Research Establishment, Harwell, Berkshire, U. K. (December 1955).
- [25]. *G. Janeschitz, K. Borrass, G. Federici, Y. Igitkhanov, A. Kukushkin, H.D. Pacher, G.W. Pacher, M. Sugihara*, *The ITER divertor concept, Journal of Nuclear Materials*, 220–222 (1995) 73-88.
- [26]. *L. Spitzer*, US Atomic Energy Commission Report NYO-993 (PM-S-1) (1951).
- [27]. *C. Stan-Sion, J. Roth, K. Krieger, M. Enăchescu, K. Ertl, V. Lazarev, H. Reithmeier, E. Nolte*, AMS – Sensitive tool used as nuclear safeguard and to diagnose fusion experiments, *Nucl Instr Meth B* 259(1) (2007) 694-701.
- [28]. *P-H. Rebut, D. Boucher, D.J. Gambier, B.E. Keen, M.L. Watkins*, The ITER challenge, *Fusion Engineering and Design*, 22(1-2) (1993) 7-18.
- [29]. *J.-H. Feist, B. Heinemann, J. Sielanko and E. Speth*, Magnetic shielding considerations for the ITER neutral beam injectors, In *Fusion Technology 1994*, Elsevier, Oxford, 1995, Pages 633-636, ISBN 9780444822208, <https://doi.org/10.1016/B978-0-444-82220-8.50126-6>.

- [30]. *M. Dogaru, M.A. Calin, C. Stan-Sion*, Tritium measurements by AMS and applications, *J Radioanal Nucl Chem* 288 (2011): 491-498, doi.org/10.1007/s10967-011-0981-6.
- [31]. *C.Stan-Sion, M. Enachescu*, AMS method for depth profiling of trace elements concentration in materials - Construction and applications, *Nucl. Instr. and Meth. B*, 361 (2015) 250-256.
- [32]. *C. Stan-Sion, M. Enachescu, O. Constantinescu, M. Dogaru*, A decade of experiments and recent upgrading at the AMS facility in Bucharest, *Nucl Instr Meth B* 268(7-8) (2010) 863-866.
- [33]. *J. Likonen, E. Alves, A. Baron-Wiechec, S. Brezinsek, J. P. Coad, A. Hakola, K. Heinola, S. Koivuranta, G. F. Matthews, P. Petersson, M. Rubel, C. Stan-Sion, A. Widdowson, JET-EFDA Contributors*, First results and surface analysis strategy for plasma-facing components after JET operation with the ITER-like wall, *Physica Scripta* T159 (2014) 014016.
- [34]. *M. Enachescu, V. Lazarev, C. Stan-Sion*, Unfolding procedure for AMS depth profiling, *J. Phys. D: Appl. Phys.* 39 (2006) 2876-2880.
- [35]. *E. Thompson, D. Stork, H. P. L. de Esch, JET Team*, The use of neutral beam heating to produce high performance fusion plasmas, including the injection of tritium beams into the Joint European Torus (JET), *Phys. Fluids B5* (1993) 2468, <https://doi.org/10.1063/1.860977>.
- [36]. *E. Surrey, D. Ceric, D.J. Godden, B. Crowley*, Properties of the JET 130kV/60A positive ion neutral injector, *Fusion Engineering, Proc. 19th SOFE*, Atlantic City (2002), doi: 10.1109/FUSION.2002.1027643.
- [37]. *G.Y. Sun, M. Friedrich, R. Grotzschel, W. Bürger, R. Behrisch, C. García-Rosales*, Quantitative AMS depth profiling of the hydrogen isotopes collected in graphite divertor and wall tiles of the tokamak ASDEX-Upgrade, *Journal of Nuclear Materials*, 246(1) (1997) 9-16.
- [38]. *M. Friedrich, W. Pilz, G. Sun, R. Behrisch, C. García-Rosales, N. Bekris, R.D. Penzhorn*, Tritium depth profiling in carbon samples from fusion experiments, *Nucl Instr Meth B* 172 (1-4) (2000) 655-658.
- [39]. *W. Pilz, M. Friedrich, K.-H. Heinig, B. Schmidt, J. von Borany*, AMS depth profiling of humidity in silica, *Nucl Instr Meth B* 219-220 (2004) 459-462.
- [40]. *M. Friedrich, W. Pilz, G. Sun, R.-D. Penzhorn, N. Bekris, R. Behrisch, C. Garcia-Rosales*, Tritium depth profiling by AMS in carbon samples from fusion experiments, *Phys. Scr. T* 94 (2001) 98.
- [41]. *M Friedrich, W Pilz, N Bekris, M Glugla, M Kiisk, V Liechtenstein*, A small and compact AMS facility for tritium depth profiling, *Nucl Instr Meth B* 223-224 (2004) 21-25.
- [42]. *C. Stan-Sion, R. Behrisch, J.P. Coad, U. Kreißig, F. Kubo, V. Lazarev, S. Lindig, M. Mayer, E. Nolte, A. Peacock, L. Rohrer, J. Roth*, Hydrogen isotope depth profiling in carbon samples from the erosion dominated inner vessel walls of JET, *Journal of Nuclear Materials*, Volumes 290-293 (2001) 491-495.
- [43]. *C. Stan-Sion, A.R. Petre*, AMS measurements of fuel retention on the surface of materials used in fusion experiments, *U.P.B. Sci. Bull., Series A*, 76(3) (2014) 265-274.
- [44]. *A.R. Petre, D. Colceag, F. Stokker-Cheregi, M. Enachescu, C. Stan-Sion, M. Dogaru*, Carbon retention of deuterium when exposed to deuterium gas during tungsten deposition, *16th International Conference on Plasma Physics and Applications*, June 20-25, 2013, Magurele, Bucharest, Romania.