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NEW METHOD TO DETECT LOW TRITIUM CONCENTRATION IN METALS

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A new method for tritium concentration measurement in plate matrix samples by AMS method is proposed. Using the relative sensitivity factor from Secondary Ion Mass Spectrometry, the depth profiles of tritium concentration in steel and copper plates were obtained in different conditions with good results in agreement to other method measurements

Keywords: AMS analysis, tritium concentration, profilometry

1. Introduction

The design, exploitation and decommissioning of tritium handling facilities implies a good understanding of the isotope’s influence on the fundamental properties of the materials used in these facilities. As metals are the main component of nuclear equipment, an increased interest for the tritium concentration evaluation in these materials is noticed.

Liquid Scintillation Counting (LSC) is the most popular tritium concentration measuring method in water samples with low tritium levels [1-3]. Tritium is a low energy beta emitter, so measuring its concentration in samples that are not soluble or dissolvable in the scintillation liquid cocktail is quite difficult and requires preliminary separation of radio-chemical species [4, 5].

Other methods used for the determination of tritium concentration in solids, as well as the depth profiles are Secondary Ion Mass Spectrometry (SIMS) and Accelerator Mass Spectrometry (AMS). These techniques use sputtering processes to produce secondary ions from the sample. These ions are then identified and separated by electric and magnetic filters and finally measured one by one inside a particle detector [6-9].

The difficulties for SIMS and AMS techniques rise from the quantifying of electric signal produced by analyzed ion signal. As this signal’s size is seriously affected by the matrix effect and electronic affinity/ionizing potential of rejected particles, SIMS and AMS require the use of standards obtained by doping the analyzed particle in the same host matrix. Standards achievement demands a doping method able to ensure the uniformity and time stability of the

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concentration of the doped element in host matrix. The most common methods combine layers deposition and implants. Depositions present a thickness limitation to several hundreds of microns of the standard samples. Otherwise, for larger thicknesses, exfoliations of the deposited layer will occur. For the implants, the concentration of the implanted element is almost uniform only in a relatively narrow region. Afterwards, a depth dependence of the implanted element beam energy is observed. To obtain a uniform surface concentration just below the surface and inside the standard sample, the implants are placed over a deposited layer made from the same material as the host matrix. In the end, this layer is removed. The energy of ionic implant is selected so the implant's maximum (whose value is known from previous calculation) is placed on the interface between the deposited layer and the substrate (the host matrix). When the deposited layer is removed, the surface regions and those situated in the immediate vicinity of the surface have rather constant concentrations of interest element.

The ionic implant dose might be changed to obtain the desired surface concentration. Previous studies revealed that the surface roughness after removing the substrate was not significantly changed [10]. All the methods described here have a major inconvenience: the dopant concentration is evenly distributed inside a very thin layer i.e. the standard sample thickness is very small. Thus, they can be used for a very small number of measurements.

Wilson et al [11] used the depth distribution of 50 elements implanted in aluminum, titan, nickel and wolfram and gold to realize the necessary samples for quantitative SIMS measurements. The results pointed a 15%-20% between experimental and calculated data for metals with high Z values (wolfram and gold) but they were not satisfying due to the reduced depth possible for elemental implant [11]. To determine the fuel retention in safety systems of tokamak, standard samples were prepared with very precise tritium and deuterium concentrations in graphite plates. They did not rely on the implant method or the successive deposition of graphite layers with deuterium or tritium. They were based on the hydrogen atoms diffusion into graphite and on the stable bonds they make in the carbon matrix [12]. The deuterium and tritium carbon standards, manufactured in IFIN-HH, Romania, have been successfully used in various studies conducted by both AMS and SIMS [13-17].

In this paper we developed a method to evaluate the very low concentration of tritium in metals, without using special standard for each metal matrix. The method was applied to steel and copper plates which were subjected to extremely low tritium concentrations in liquid and gaseous environment. The tritium concentration in the tritiated steel and copper samples was determined using the AMS facility in IFIN-HH, Romania [18, 19].

2. Relative sensitivity factor

By sputtering process, a single ion having low energy (few keV) hits the sample producing a collision cascade near the sample surface. The particles with high energy, intercept the surface and are rejected outside the sample. During this process, they might either loose or gain electrons by collision processes and mainly by electron exchange on the matrix surface. This phenomenon depends on electron affinity and ionization potential of the rejected particles. The sputtered particles form a secondary beam which can be analyzed by AMS or SIMS system.

In Secondary Ion Mass Spectrometry (SIMS), the relative sensitivity factor (RSF) is used as a conversion factor for the secondary isotope beam intensity to isotope concentration [20], is defined according to the following equation:

$$C_M^i = RSF_M(i)\frac{I_M^i}{I_M^m} \tag{1}$$

where C_M^i is concentration of impurity isotope *i*, I_M^i is the ionic current of the impurity isotope from matrix *M* and I_M^m is the isotope majority from the matrix *M*. C_M^i and $RSF_M(i)$ have the same concentration unit (atoms/cm^3). The evaluation of unknown concentration of impurity isotope from a matrix *M*, require the measurement of secondary ion current of impurity beam and those of isotope majority of the matrix if $RSF_M(i)$ is known. $RSF_M(i)$ depends on the physical nature of isotope majority from the matrix and on the ionization potential or electron affinity for secondary positive or negative ions [21]. In the literature, there are theoretical and empirical formula for RSF [21-23].

The problem is easy when reference samples are available and their parameters are precisely known. Unfortunately, this can’t be obtained for each element and many authors are trying to find another solution. Previous studies refer mostly on silicon element or semiconductors *RSF*, being the materials in which useful implants were made in electronics [21].

In classical AMS analysis the sample material is transformed, by chemical processes in homogeneous powder. The same chemical procedures are used for reference samples so the RSF evaluation is precise and easy. If AMS is used to determine the depth concentration (profilometry), the sample remains in its raw state. Making standards with the same solid matrix is difficult and RSF can’t be determined.

The determination of the tritium concentration in a matrix *M*, in the absence of tritium standards in the given matrix, was performed using the tritium concentration in graphite, different RSF values of some elements in Si [21] and the recurrence relation between RSFs of various elements in different matrices [23]. Thus, for tritium, Eq. (1) becomes:

$$C_M^T = RSF_M(T)\frac{I_M^T}{I_M^m} \tag{2}$$

For tritium standard in graphite (*C*), Eq. (2) can be written as:

$$C_C^T = RSF_C(T)\frac{I_C^T}{I_C^{^{12}C}} \tag{3}$$

where ^{12}C is the main isotope in graphite.

Recurrence relation between RSFs of various elements in different matrices is [23]:

$$RSF_M(y) = \frac{RSF_Z(y)}{RSF_Z(m)} \cdot C_M^m \tag{4}$$

where: RSF_M and RSF_Z are the relative sensitivity of each matrix, while *m* and *y* are the isotopes. C_M^m is the atomic concentration of *m* isotope in *M* matrix.

Using *Si* as *Z* matrix and Tritium (*T*) as *y* isotope, Eq. 4 becomes:

$$RSF_M(T) = \frac{RSF_{Si}(T)}{RSF_{Si}(m)} \cdot C_M^m \tag{5}$$

The $RSF_C(T)$ for standard sample can be calculated from Eq. 5 considering graphite (*C*) as *M* matrix and ^{12}C as main isotope:

$$RSF_C(T) = \frac{RSF_{Si}(T)}{RSF_{Si}(^{12}C)} \cdot C_C^{^{12}C} \tag{6}$$

By replacing $RSF_{Si}(T)$ from Eq. (6) in Eq. (5) we obtain:

$$RSF_M(T) = \frac{RSF_C(T) \cdot RSF_{Si}(^{12}C)}{RSF_{Si}(m)} \cdot \frac{C_M^m}{C_C^{^{12}C}} \tag{7}$$

Using Eq. 2 şi Eq. 3 in Eq. 7 we get :

$$C_M^T = \frac{RSF_{Si}(^{12}C)}{RSF_{Si}(m)} \cdot \frac{I_M^T}{I_M^m} \cdot \frac{I_C^{^{12}C}}{I_C^T} \cdot \frac{C_M^m}{C_C^{^{12}C}} \cdot C_C^T \tag{8}$$

The $RSF_{Si}(^{12}C)$ and $RSF_{Si}(m)$ factors from Eq. 8 are given in [20]. I_M^T and I_M^m are the tritium and *m* isotope ionic currents measured in the sample whose matrix is *M*. I_C^T $I_C^{^{12}C}$ are the ionic currents of tritium and ^{12}C isotope, measured in standard sample and with the known tritium concentration C_C^T .

For a better understanding of standard sample parameters, we can replace I_C^T with I_{st}^T, $I_C^{^{12}C}$ with $I_{st}^{^{12}C}$ and C_C^T with C_{st}. Thus, Eq. 7 becomes:

$$C_M^T = \frac{RSF_{Si}(^{12}C)}{RSF_{Si}(m)} \cdot \frac{I_M^T}{I_M^m} \cdot \frac{I_{st}^{^{12}C}}{I_{st}^T} \cdot \frac{C_M^m}{C_C^{^{12}C}} \cdot C_{st} \tag{9}$$

Two M matrixes were experimentally used: steel, with ^{56}Fe as majority isotope (m) and copper with majority isotope ^{63}Cu.

3. Experimental

3.1 Samples

Five stainless steel plates and five copper plates having the thickness of 1 mm were cut to fit the AMS sample holder (7x7 mm) and were exposed to the action of tritiated water and elemental tritium (T_2). The same size plates were cut from non-tritiated samples and were analyzed to establish the tritium background level in AMS facility.

Exposure to tritiated water

For tritiated water exposure, the samples were placed in 4 mL high density polyethylene bottles (HDPE) provided with a low-density polyethylene (LDPE) screw cap. Each bottle was filled with 2 mL of tritiated water with a radioactive concentration of 7.682 MBq/mL (4.3×10^{15} ^3H atoms/mL), closed and sealed with M parafilm. The samples were stored in ventilated enclosure at room temperature. Exposure times were 10 and 20 days, respectively. After exposure, the tritiated water was removed from bottles by means of 2 mL polypropylene syringes.

The samples were decontaminated by immersion in ethyl alcohol p.a. (5 mL) and stored for 24 hours to remove labile tritium from the surface and dried in the oven for 8 hours at 90 ± 2 ^oC.

Exposure to elementary tritium

For exposure to elementary tritium, the samples were placed in glass tubes ended with capillary coupled to the Tritium Vacuum and Manipulation Device via a SN10 normalized cone. The vials were previously degassed for two hours at room temperature. Elementary tritium was generated by a U Bed (made in Russia) and transferred in exposure vials by a mercury piston pump. The elementary tritium pressure, read at the manometer, was 150 Torr. The radioactive concentration of elemental tritium was 17.5 GBq / mL (9.8×10^{18} ^3H atoms/mL).

The vials were sealed by flame welding and stored in a ventilated fume hood at room temperature. Exposure times were 3, 6, and 11 days.

After exposure, elemental tritium was recovered with the Tritium Vacuum and Handling Device. The samples were decontaminated by degassing at low pressure (< 10^{-1} Torr) for 2 hours at room temperature by immersion in ethyl alcohol l p.a. (5 mL) and stored for 24 hours to remove unstable tritium from the surface and dried in the oven for 16 hours at 120 ^{o}C + 5 ^{o}C.

3.2 Techniques

The AMS installation from IFIN-HH was changed to determine the depth profile of the trace elements in the materials [24]. This improvement was taking into account that the $^{133}Cs^{+}$ sputtering beam penetrate target sample producing a small crater. Then, the ions will be extracted from a higher depth and the data acquisition system will measure the time variation of the element concentration inside the material. Finally, the crater depth will be measured by optic profilometry and thus the time scale will be converted to the depth scale.

A serious problem that occurs in depth profiling measurements is the crater edge effect. It gives false information about the concentration corresponding to the depth *h*. It consists in the fact that the excavated material at time *t* belongs not only to the corresponding depth *h*, but also to a part of the upper layers on the edge due to the crater's width with the depth. The effect would not occur if the crater is cylindrical. To correct this effect, a simple mathematical procedure has been developed in [25].

4. Results

Tritiated samples were analyzed by AMS facility at IFIN-HH Romania and the tritium concentration was evaluated from Eq. 9. The standard tritium in graphite sample concentration was $1.2\text{x}10^{10}$ atoms /cm^3.

Before and after each measurement, the $I_{st}^{^{12}C} / I_{st}^{T}$ ratio for standard sample was determined. The $I_{st}^{^{12}C}$ and I_{M}^{m} currents were determined in a Faraday cup placed at the exit of AMS facility, in front of particle detector. Tritium currents I_{M}^{T} and I_{st}^{T} were measured in particle detection system and expressed in atoms/s. For each measurement cycle the standard sample and the background tritium level in the facility was determined by measuring the steel and copper untritiated samples. The background level value was subtracted from each analyzed sample values.

For the depth profilometry measurements by AMS, an Itutoyo SJ 301 profilometer was used to determine the dimensions of the sputtering crater.

Steel

The steel plate, from which the samples were cut, was previously analyzed by X-ray fluorescence to determine each components concentration. The result showed the following composition: 91.2 % *Fe*, 7.9 % *Cr*, 0.4 % *Ni*, 0.3 % *Cu* and 0.2% *Mn*. For steel the equation (8) become:

$$C_{stell}^{T} = \frac{RSF_{Si}(^{12}C)}{RSF_{Si}(^{56}Fe)} \cdot \frac{I_{steel}^{T}}{I_{steel}^{^{56}Fe}} \cdot \frac{I_{st}^{^{12}C}}{I_{st}^{T}} \cdot \frac{C_{steel}^{^{56}Fe}}{C_{C}^{^{12}C}} \cdot C_{st} \tag{10}$$

The ^{56}Fe isotope was considered as the majority one in this matrix. The steel mass density is 7.93 g/cm^3 and the ^{56}Fe naturally abundance is 91.72%. By these values, the atomic concentration of ^{56}Fe from the steel sample is $C_{steel}^{^{56}Fe} = 7.1\times10^{22}$ atoms/cm^3. The ^{12}C isotope concentration in graphite is $C_{C}^{^{12}C} = 0.5\times10^{22}$ atoms/cm^3. Iron and carbon RSFs values in silicon are $RSF_{Si}(^{56}Fe) = 4.8\times10^{22}$ atoms/cm^3 and $RSF_{Si}(^{12}C) = 4.9\times10^{24}$ atoms/cm^3 respectively [22].

Experimental results for tritium concentration profilometry by AMS in steel tritiated samples in THO are presented in Fig. 1 and for T_2 analysis in Fig. 2.

As it can be noticed from the plots, the tritium atoms retained amount is one magnitude order higher in steel samples immersed in gas than those immersed in water. In gas environment, the tritium isotopes penetrate 6 microns more than HTO where the penetration depth is 200 nm, as it can be seen from other papers too.

During the measurements of the tritiated sample for 11 days in gas, tritium concentration instabilities were observed occasionally exceeding the data acquisition system power. This can be explained by the surface concentration of Tritium in the sample that might exceed 1.2×10^{12} atoms /cm^3.

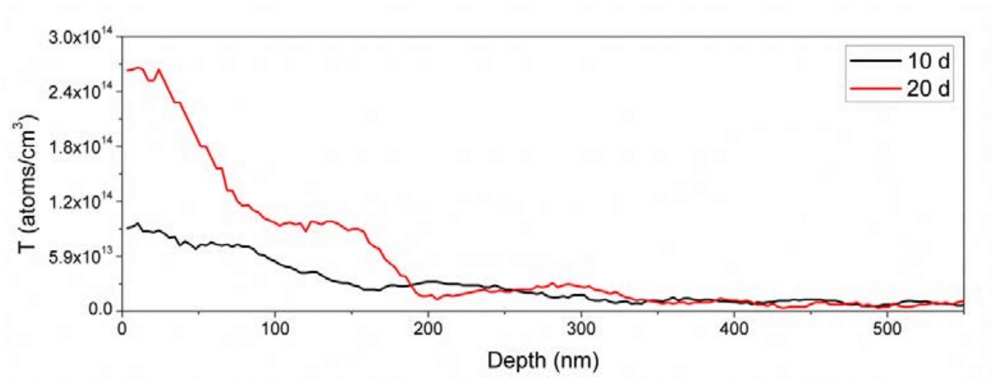

Fig. 1: Tritium concentration profilometry for steel in THO (water) for 10 days, and 20 days respectively

It was also noticed that the penetration rate in water environment was three orders of magnitude lower than for gaseous tritium which is also in good agreement with other reports in the literature.

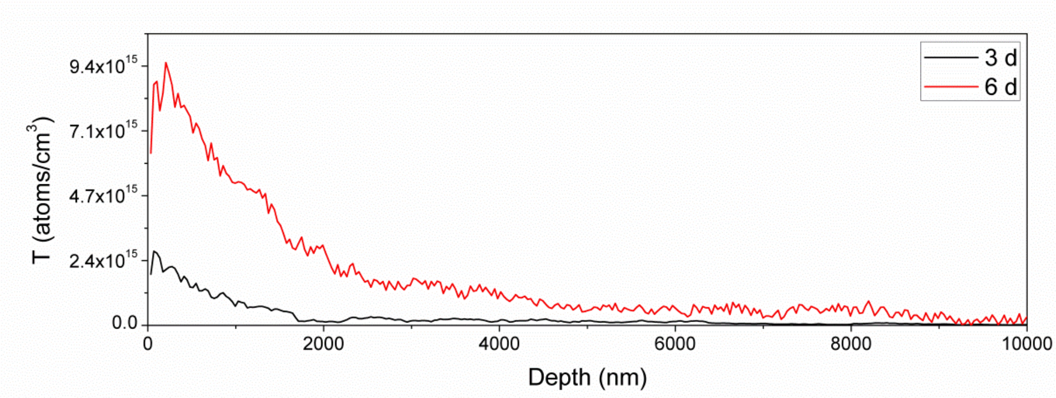

Fig. 2: Tritium concentration profilometry for steel in T_2 (gas) for 3 days, and 6 days respectively.

Copper

The copper plate from which the samples were cut was also analyzed by X-rays fluorescence to determine the compound ratio. The result showed a 99.8% purity of the sample. Eq. 9 become in this case:

$$C_{Cu}^{T} = \frac{RSF_{Si}(^{12}C)}{RSF_{Si}(^{63}Cu)} \cdot \frac{I_{Cu}^{T}}{I_{Cu}^{^{63}Cu}} \cdot \frac{I_{st}^{^{12}C}}{I_{st}^{T}} \cdot \frac{C_{Cu}^{^{63}Cu}}{C_{C}^{^{12}C}} \cdot C_{st} \tag{11}$$

In copper samples the isotope ^{63}Cu is the majority one. The mass density of copper is 8.94 g/cm^3 and its abundance is 69.15 %.

With these values, the atomic concentration of a ^{63}Cu in the sample is $C_{Cu}^{^{63}Cu} = 5.9\times10^{22}$ atoms/cm^3. The ^{12}C isotope concentration in graphite is

$C_C^{^{12}C} = 0.5\times10^{22}$ atoms/cm^3. The RSFs values for carbon and copper in silicon are $RSF_{Si}(^{12}C) = 4.8\times10^{22}$ atoms/cm^3 and $RSF_{Si}(^{63}Cu) = 4.2\times10^{23}$ atoms/cm^3 respectively [22].

Experimental results for tritium concentration profilometry by AMS in tritiated samples in THO are presented in Fig. 3 and for those T_2 (gas environment) are presented in Fig. 4.

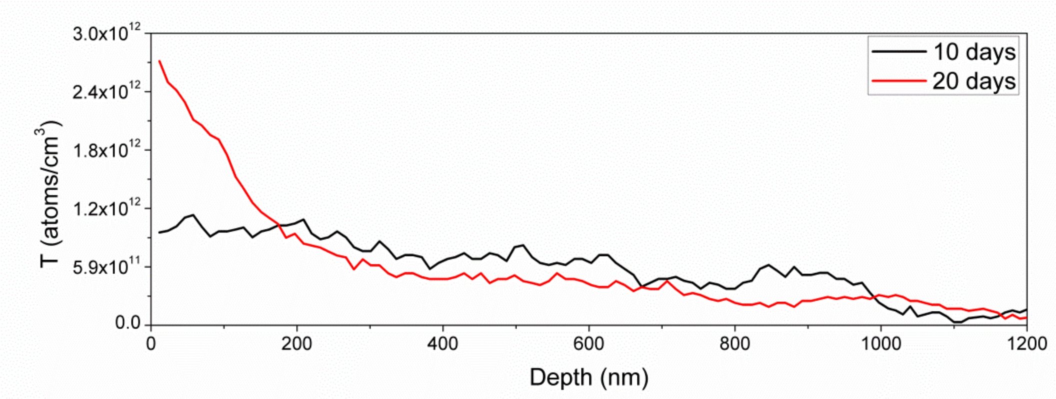

Fig. 3: Tritium concentration profilometry for copper in THO (water) for 10 days, and 20 days respectively.

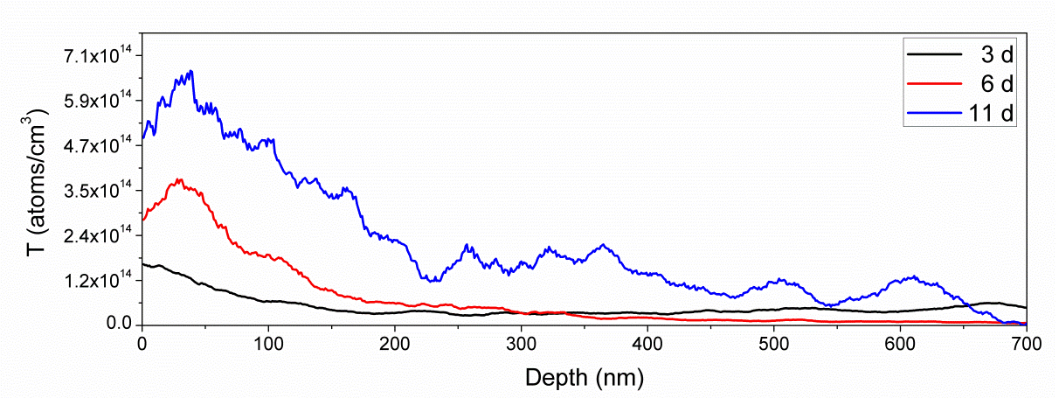

Fig. 4: Tritium concentration profilometry for copper in T_2 (gas) for 3 days, and 6 days respectively.

As it can be noticed from Fig. 3 and Fig. 4, tritium absorption is two orders of magnitude higher in coppers samples immersed in gas compared to the ones immersed in tritiated water. It was also observed that in water, the tritium isotopes penetrate deeper with 1 micron than in gas where the deepest penetration level is 200 – 300 nm. This can be justified by the oxidation process on the sample surface. The experimental plot of the most exposed sample (11 days) present some irregularities due to the disorder degree of the lattice due to the absorbed tritium. For the other samples, this phenomenon is less observed. Tritium absorption in copper was rarely determined by researchers, many papers reporting that tritium is only absorbed in copper in a thin oxide substrate. Even so, there are some papers that contradict this assumption [29].

5. Conclusions

AMS measurements are relative, so standard samples are necessary. Standard preparation is hard to achieve in plate form matrix. To avoid this inconvenience, the relative sensitivity factor was used, just as in SIMS method, to calibrate or to obtain quantitative data. Using this method, the depth profiles of tritium concentration in steel and copper plates were obtained in different conditions. The results are in good agreement with other results reported in the literature.

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