JOINT INSTITUTE FOR NUCLEAR RESEARCH
Dzhelepov Laboratory of Nuclear Problems

FINAL REPORT
ON STAGE 1 OF THE INTERNATIONAL STUDENT PRACTICE

"Studies of structural changes in heavy ion irradiated materials"

Supervisor:
Dr K. Siemek

Student(s):
Mahmoud Abdelazeem Mohammed Hussien
& Fatma Mahmoud Hamed Obiedallah
Faculty of science - Assiut University - Egypt.

Participation period:
01-24 March

Dubna, 2022

Abstract

The study of the penetrating hydrogen in the thermonuclear materials with defects provoked by temperature gradient and radiation caused by 14 MeV neutrons, 3.5 MeV He, and hydrogen products after the fusion is of major importance. The vanadium, with its specific protective properties, is a possible candidate for fusion material. Using SRIM/TRIM analysis, we estimated the number of collisions, the loss of ionic energy, the depth of penetration of irradiated ions, and the recoil energy when irradiating vanadium with 167 MeV Xe. The range of ionic deposition is about 10 micrometers at its maximum, and this is in good sync with the energy losses and target depth. The recoil energy can be compared with the recoil ionization, where the profile is analogous. The main energy recoil losses are again expected to be in the order of 10 micrometers. By the positron spectroscopy, change of the S parameter depending on the irradiation dose of vanadium with 167 MeV Xe and the energy of the positrons was analyzed. By increasing the radiation dose from the reference point to 10^{12} ions/cm^2 the S-parameter increased dramatically from 0.484 to about 0.500 and after this point, the S-parameter starts to decrease again until reaches 0.496.

Introduction

In the future fusion reactor, a blanket is expected to be placed at high temperature between 700 and 800 °C, see Fig (1). For this reason, in order to achieve long-duration and stable electricity generation it is necessary to use advanced materials as vanadium alloys which can withstand high doses at elevated temperatures and can be used in manufacturing the blanket which is in a location where it almost touches the plasma, and as if to enfold the plasma, the blanket is placed on the inner surface of the vacuum chamber. The blanket, by absorbing the high-speed particles generated by the fusion reaction inside the plasma, releases heat and a higher temperature is achieved. It is possible to develop a vanadium alloy successfully and improve its properties, i.e., ductility (property that prevents breakage by extending under tension). These properties were already improved substantially, which were able to overcome the problem of breakage at the time of machining and after welding.

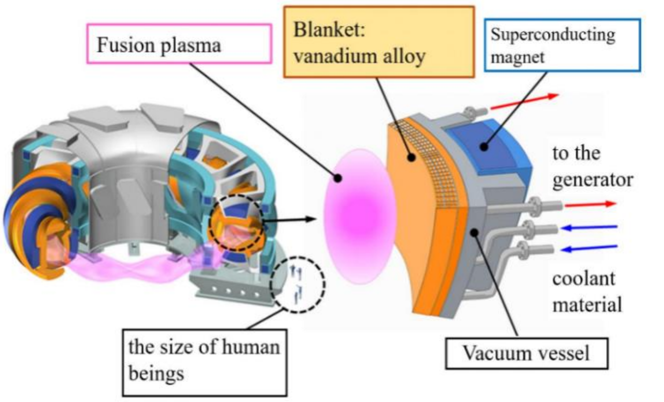

Fig. 1. *The blanket is colored orange, and the combined weight of the reactor exceeds 1000 tons. The blanket collects the high-speed particles that have emerged from the plasma in the fusion reaction and converts that kinetic energy into heat (Dr. Takuya Nagasaka).*[1]

In the presence of fusion reactions, several degradation processes must be considered that will affect the evolution of structural defects. In the first place, the defects that will be caused by the products entering the reactor walls after fusion must be taken into account. 3.5 MeV ionized helium, together with 14 MeV neutrons, is the major product of the fusion reaction. While helium mainly affects the surface, at a depth of several microns, neutrons due to lack of charge create many different defects, and its clusters, deep inside in the structure of the materials. The damages caused by neutrons can be replaced by irradiation with heavy-ions. We should also note that after the primary attack of hydrogen, helium, and neutrons, there will be a second cycle of defects, from those atoms of the base material that are hit by the products listed so far. The kinematics of this process shows that this will lead to the deposition of the repulsed atoms on the surface of the blankets, which in turn would lead to the possibility of plasma contamination and termination of the operating cycle. Each defect formation in a lattice requires certain amount of energy. This is called (defect) formation energy. In particle irradiation an incoming particle collide with atoms in the material passing its kinetic energy in the collisions. When the passed energy for atoms is larger than the material characteristic threshold displacement energy, lattice point defects, such as vacancies and interstitial atoms, can be created due to irradiation. An atom in the lattice struck by an irradiation particle is called primary knock-on atom (PKA). After the struck, the PKA is propagating in the lattice with an energy given in the struck minus the displacement energy. If the transferred energy is sufficiently high, the atom can also displace other atoms in the lattice forming a collision cascade. Finally, when the excited knock-on atoms have lost their energy, they are terminated as interstitial atoms in the lattice.

If the lattice contains a lot of mono-vacancies homogeneously or heterogeneously, they start to cluster forming dislocation loops, voids and stacking faults. Increasingly, these lead to macroscopic effects such as swelling, hardening, embrittlement and direct failure. In the case of especially proton, neutron or alpha particle irradiation, in addition to radiation damage, the target material may form radioactive isotopes via nuclide reactions. As a consequence, materials close to the core of a fission or fusion reactor are activated and need to be handled as a radioactive waste after decommissioning.

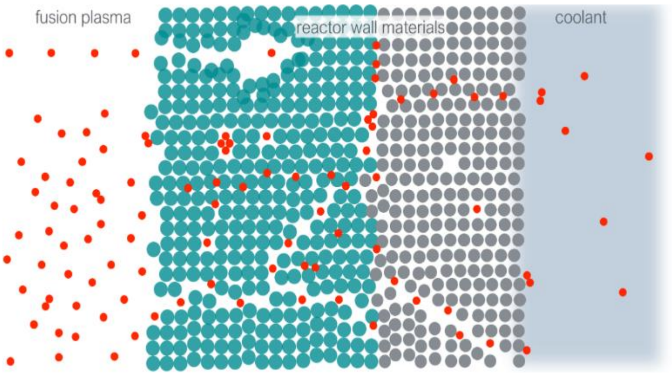

Fig. 2. *An illustration of the atomic level processes of hydrogen permeation from the fusion plasma through the reactor wall materials into the reactor coolant. (Image: K.Heinola/IAEA)* [2,3]

Hydrogen is the lightest and smallest of all the elements. It is also ubiquitous. Its presence within the structure of metals cannot be avoided, partly due to its high mobility. The interactions between hydrogen and metals play an essential role in the structure-property relationships of engineering alloys. Hydrogen atom's most striking effect on materials is a sudden and often unpredictable decrease in ductility, toughness, and generally in the material's resistance to fatigue-crack propagation known as hydrogen embrittlement. hydrogen embrittlement happens when a very small amount of hydrogen goes inside a piece of the material as apear in Fig (2), as hydrogen is very light, very small, very fast, and the most abundant element in the universe. When we think of metal, we think of a big block, but we actually think of small bricks and every brick may contain different elements which consist of the alloy (crystal lattice). On the macroscale when the hydrogen goes in, the lattice will make it more brittle.

Generally, the role of lattice point defects in nuclear materials is very important in determining their properties, such as radiation tolerance, diffusion of atoms in the material and mechanical endurance.

The most commonly microscopy techniques used in nuclear material research are positron annhilation spectroscpy (PAS), transmission electron microscopy (TEM), X-ray diffraction spectroscopy (XRD), Electron paramagnetic resonance (EPR) and Neutron scattering spectroscopy. Other powerful characterization family is the particle beam methods, such as secondary ion mass spectrometry (SIMS), ion beam analysis techniques (IBA) and electron beams. It should be noted that usually a single method is not sufficient for getting the whole defects in nuclear materials experimental picture, and other methods are required for completeness. In this report we have used positron annihilation spectroscopie for vacancy-type defect characterization in nuclear materials.

Through Monte Carlo simulations using SRIM/TRIM The theoretical defect and ion distributions can be provided by commonly available programs such as SRIM/TRIM. This program includes the basics interaction of ions with matter as appear in fig (3) and allow us to prepere materials with well defined ions distributions below the Surface. For example it is very important in semiconductor where the doping atoms have to got the same concentractions in whole samples region. In such cases a multiimplanations process have to be carried. The multiimplantion is the proces of implanation with different ions energies. Its clear that increase of ions energies allow to increse the depth on which the ions are implanted.

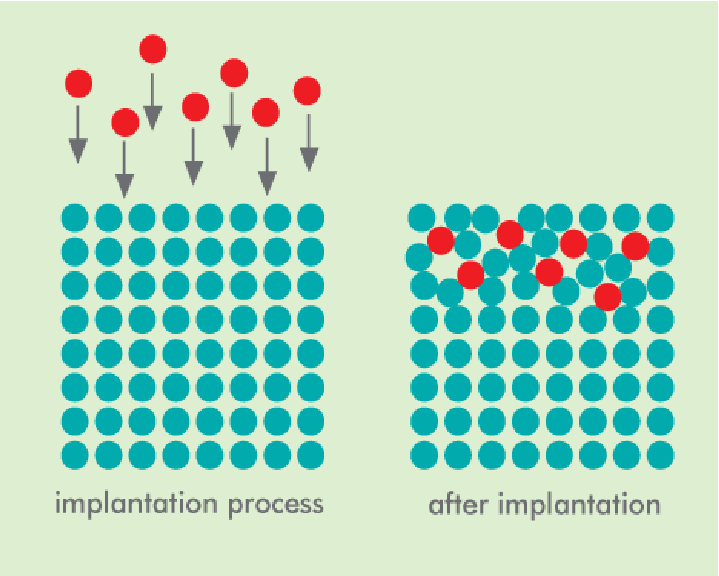

Fig. 3. *interaction of implantaion ions with matter .*

During interaction with matter energetic ions lose energies. High Energy for example 100 MeV ion mostly lose energies on ionization, end not collide with target atoms until it reach the smaller energies. The most collisions and vacacnies creations occurs for small energies of ions. It cause different defects and ions distributions after implanation. At first we look how the Energy of ions is deposited in materials for ion with Energy 5 MeV and 25 kev.

The main goals of the project

The objectives of the project were to learn possibilities of positron and introduction with other complementary methods used in the field of studies of nuclear materials. These possibilities can be abbreviated in determination of defect concentration, evaluation of defect concentration profile and detection of the kind/size of defects. For these purposes, damage to the pure vanadium after irradiation with heavy high-energy 167 MeV Xeions was considered. Heavy ions irradiation was used in replacement of neutrons irradiation occurring in fission reactors. An SRIM (Stopping and Range of Ions in Matter) / TRIM (Transport of ions in matter) analysis was performed to assess the depth of penetration of the irradiating ions and to study the ion energy losses process. Using Doppler broadening spectroscopy, we study how the annihilation parameters change depending on the positron energy. In addition, model calculations of positron annihilation were performed for titanium carbide, which is also a suitable candidate for fusion material.

Scope of work

In this work, a narrow range of gamma spectra (about 511 KeV) is considered, with the help of which the full range of different types of defects in materials can be studied. Combining the possibilities of positron spectroscopy with the analysis of radiation materials, we can enter a challenging field in terms of future research, namely - fusion materials.

Methods

Positron annihilation spectroscopy (PAS) is a suitable tool for studying changes in the material structure. It is a sensitive method for the detection of open-volume defects, such as vacancies, vacancy clusters, dislocations, etc. Positrons offer unique information about the studied material based on their inherent properties. Positrons are light positively charged particles and they annihilate with electrons.

A positron has a mass of an electron, allowing it to penetrate material deeper and cause less damage in the studied samples in comparison to heavier particles. PAS techniques allow one to find defects with the size ranging from 0.1 nm with the concentration up to 10^7 ppm. PAS makes it possible to detect defects in a wide range of depths from single nanometers up to millimeters as shown in Fig (3).

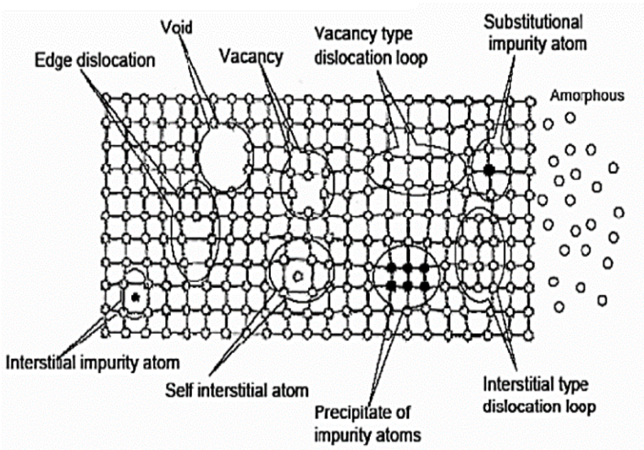

Fig. 3. *Defects in the lattice structure of materials* [4, 5].

PAS has three main techniques angular correlation of gamma quanta, Doppler broadening spectroscopy (DPS) and positron annhilation lifetime spectroscopy (PALS).

The two most frequently used techniques to detect and to investigate the properties of open volume defects and their interaction with the medium are the lifetime and the Doppler broadening techniques. If fast positrons from a radioactive nuclide are used, the mean properties of the material under investigation are measured in a layer several tens of micron thick . Otherwise, if a slow positron beam is utilized, a depth scanning from the surface to a few microns is possible. The momentum of a thermalized positrons is a lot smaller than the momentum of electrons in a crystalline lattice. Therefore, the momentum seen in the Doppler broadening of the annihilation photons is that of an electron at the annihilation location.

When a positron annihilates with an electron, two γ photons are created with the energies of $E = 511\ KeV \pm \Delta E$, where ΔE corresponds to the Doppler shift caused by the momentum of the annihilating electron-positron pair. These shifts are recorded experimentally with a high-purity germanium detector of a high energy resolution, and the shifts cause the characteristic broadening of the 511 keV annihilation peak.

The shape of the Doppler-broadened 511 keV annihilation line is generally characterized by dividing it into two types of regions, as in Fig (5). The central region describes the count rate of the low momentum electrons, and the wing regions, taken from the peak edges, describe the count rate of the high momentum electrons. The peak shape is conveniently described by using the S and W parameters. The S parameter is defined as the counts in the central region divided by the total number of counts in the annihilation peak and the W parameter is defined accordingly as counts in the wing regions divided by the total number of counts in the annihilation peak. The S and W parameters are very sensitive to changes in the electron momentum distribution at the annihilation site. So, increasing the S parameter is evidence of increasing the number of defects.

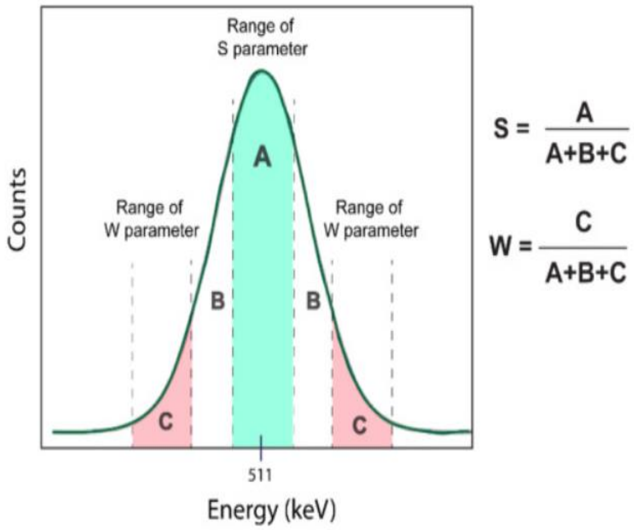

Fig. 5. *Definition of defect parameters S and W extracted from the 511 keV annihilation peak. A typical value for the windows are: from 510.2 to 511.8 keV for region A. For the wings C, from 507.8 to 509.3 keV and from 512.7 to 514.8 keV.*[6].

Variable energy positron beam at the Laboratory of Nuclear Problems has recently been applied in PAS investigations. It offers the possibility of measurements with slow positrons in the energy range between 50 eV and 35 keV. Positrons emitted from ^{22}Na isotope with energy up to 0.545 MeV are moderated to a few electron volts on the frozen Neon. On the exit of the moderator, both fast and slow positrons appear. Next, they are separated by the combination of the longitudinal magnetic field and two sections of the transverse magnetic field of opposite directions placed one by one. As a result, the slow positrons have a 'slalom-like' trajectory when they come to the aperture diaphragm. Only low-energy positrons go to the sample chamber. The holder with samples hung on a vertically moving rod is under potential, which can be changed in the range between 0 and –35 kV. In this way, positrons can be accelerated to expected energies [7,8]. Fig (6) shows a diagram of a Positron Injector with LEPTA entrance.

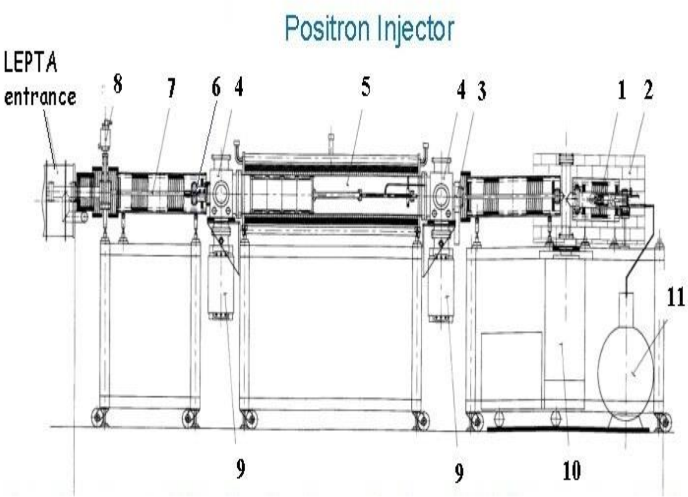

Fig. 6. *Positron Injector with LEPTA entrance: 1-positron source ^{22}Na, 2-radioactive protection shield, 3-vacuum valve, 4-vacuum chamber for pumping out and diagnostic tools, 5-positron trap, 6-vacuum isolator, 7- positron vacuum channel, 8-vacuum "shutter" (fast valve), 9- ion pump, 10-turbopump, 11-LHe vessel 19* [8].

In the current work, we conducted a series of experiments, which explored the possibilities provided by positron spectroscopy for the study of structural changes in condensed materials and defects in them. At the same time, several approaches to simulate the irradiation process were implemented, which provided an opportunity to fully characterize the irradiation process and the resulting positron annihilation dependencies.

Results and Discussion:

Using SRIM/TRIM analysis, we estimated the number of collisions, the loss of ionic energy, the depth of penetration of irradiated ions, and the recoil energy when irradiating vanadium with 167 MeV Xe.

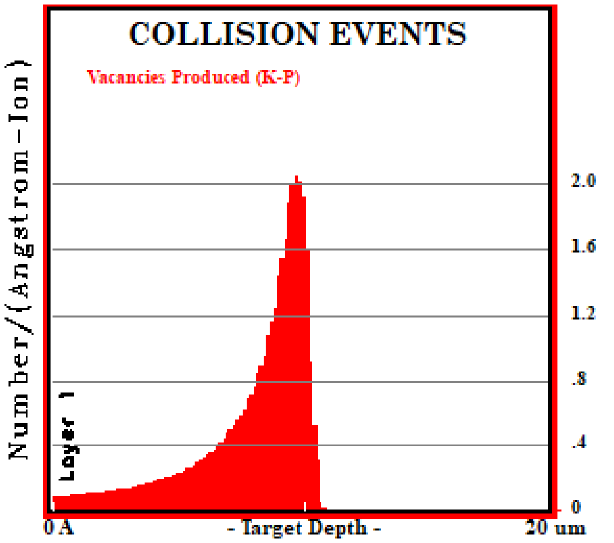

Fig. 7. *Relation between the target depth on the X-axis versus the number of vacancies in the Y- axis.*

As shown in Fig (7), when the ions collide with the target atoms, they give them energy and the maximum energy occurs at the place where the ions stop, here the number of vacancies will be maximum but we will focus our study on the first 1 micrometre (surface) target depth which has a very small number of vacancies.

Generally, as shown in Fig (8) when the ions enter the target material, they lost energy through the ionization process and when they reached equilibrium with the atoms; they start to collide with the atoms randomly so the energy of the ions decreases by increasing the target depth.

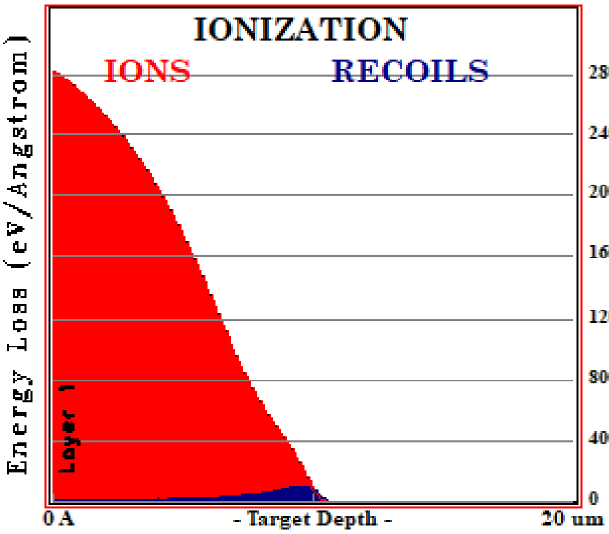

Fig. 8. *Relation between the target depth on the X- axis versus the energy loss (eV/Å).*

Fig (9) shows that the range of ionic deposition is about 10 micrometres at its maximum. This, in turn, is in good sync with the graphs of energy losses and target depth. The following figure shows the recoil energy. A comparison can be made with the recoil ionization of Fig. (**8**), where the profile is analogous.

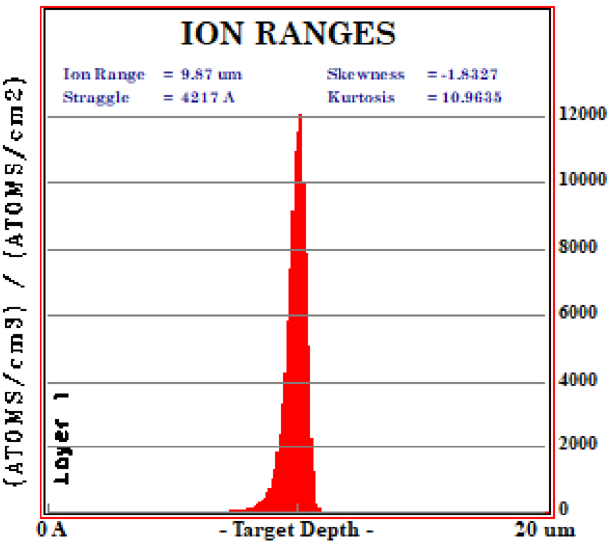

Fig. 9. *Relation between the target depth on the X- axis versus the ion ranges.*

As can be seen in Fig (10), the main energy recoil losses are again expected to be in the order of 10 micrometres.

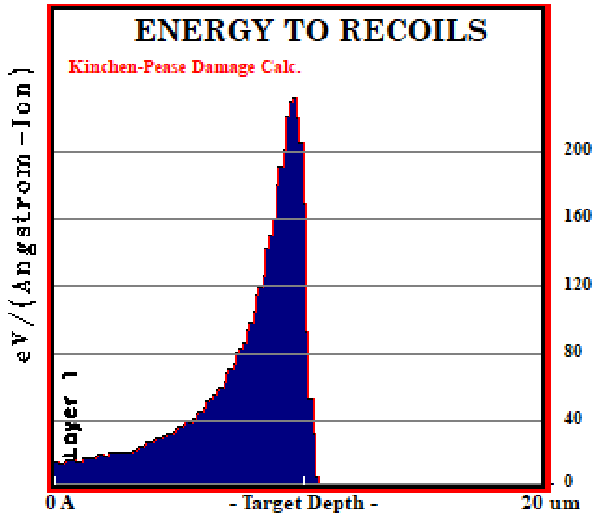

Fig. 10. *Relation between the target depth on the X- axis versus the energy to recoils.*

Using the methods of positron spectroscopy, the change of the S parameter depending on the irradiation dose of vanadium with 167 MeV Xe and the energy of the positrons was analyzed.

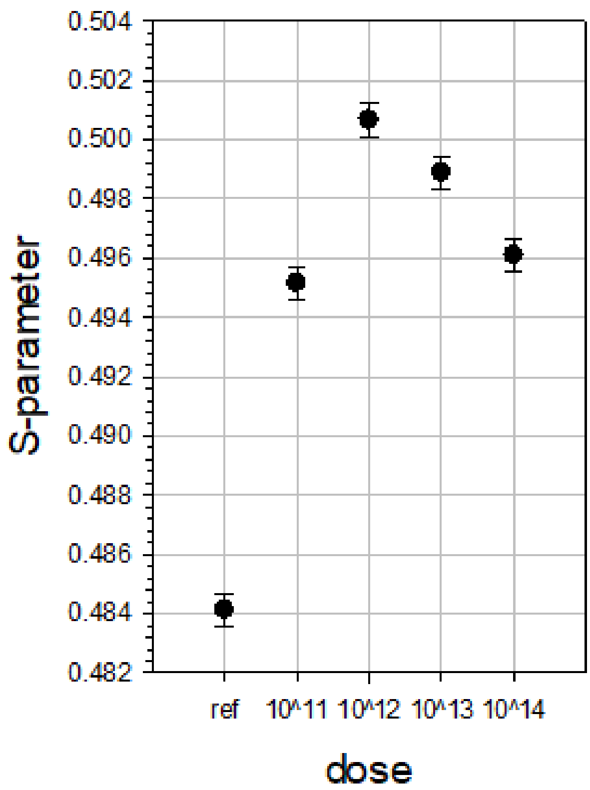

Fig. 11. *The dependency of the S parameter in the function of the irradiation dose of 167 MeV Xe ions obtained using conventional Doppler broadening spectroscopy*.

In Fig (11) we present the results of conventional Doppler broadening spectroscopy measurements. In the conventional method, positrons are reaching average implantation depth equal to 37 µm. It is 3 times much higher than the predicted implantation range in Fig (9).

As shown in Fig (11) by increasing the radiation dose from the reference point to 10^{12}, the S-parameter increased dramatically from 0.484 to about 0.500 and after this point, the S-parameter starts to decrease again until reach 0.496.

This result can be explained as follows, the higher the irradiation dose, the greater the number of vacancies, which increases the value of the S parameter however after the dose increased more than 10^{12}, some vacancies are filled by the implanted ions so it decreases resulting in decreasing in the S-parameter.

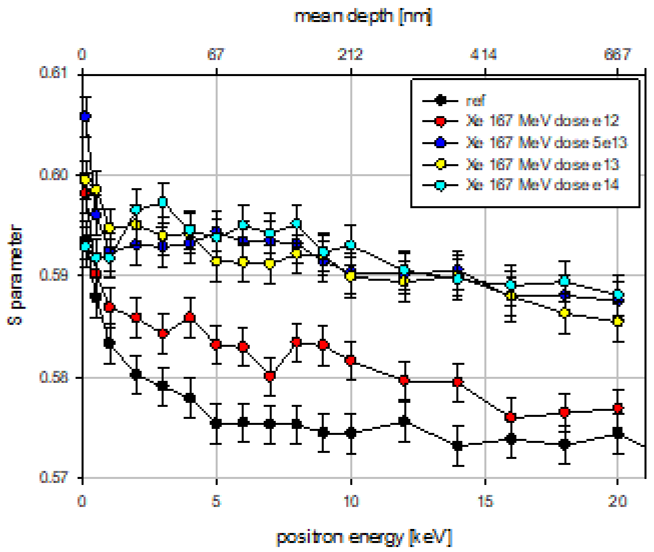

Fig. 12. *The dependency of S parameter in function of the implanted positron energy obtained using variable energy positron beam*.

As discussed in Fig (11) The S-parameter increases with increasing the dose. At the upper axis, we calculate the mean depth (from 0 to 667 nm) of positrons in the material, which corresponds to the energy of the implanted positrons. The reference sample (black curve) has the lowest S parameter and hence the lowest number of vacancies. However, the other three samples have almost the same S parameter and the Xe 167 MeV dose 10^{12} ion/cm^2 (red curve) has the intermediate S parameter this is clear in Fig (12). It means that the number of vacancies for the positrons is big enough to find a defect, so all the positrons annihilate in these defects and all the defects are similar. We calculated the mean depth of the position and then compared it with the results. One can observe that at the reference sample, the positron can go through a 67 nm thick layer by diffusion to the surface, but through the other three samples it can only go for about 10 nm. So, the positron diffusion length decreased, and therefore also the defect concentration increased.

Conclusions

The result from conventional Doppler broadening spectroscopy can be explained as follows, the higher the irradiation dose, the greater the number of vacancies, which increases the value of the S parameter, however after the dose increased more than 10^{12} some vacancies were filled by the implanted ions so it decreases resulting in decreasing in the S-parameter.

By the positron beam, positrons are reaching average implantation depth equal to 37 µm. It is three times higher than the predicted implantation range.

The S-parameter increases with increasing the dose, but the defects from all doses are similar.

At the reference sample, the positron can go through 67 nm layer by diffusion process but through the other three samples, it can only go for about 10 nm. The positron diffusion length decreased respectively therefore also the defect concentration increased.

Acknowledgments

In the preparation of this work, we received significant support from the colleague from the laboratory of neutron physics JINR, Dr. E. Popov. At the same time, we thank our colleagues from JINR M. Mirzyaev and D. Neov for their willingness to include us in future experiments in order to continue the work on this study.

Also, we want to thank our Egyptian colleagues and our Egyptian supervisors, Prof. Walaa and Prof. Medhat for their responsibility.

Finally, we are very grateful to our families and our friends especially Mr. Faisal Sayed for his efforts.

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