INVESTIGATION OF HIGHLY ACTIVATED MATERIALS BY CONVENTIONAL POSITRON ANNIHILATION SPECTROSCOPY

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1. Abstract

Positron annihilation process is helpful tool for material microstructure study. Many useful methods for detecting vacancies and their clusters in various alloys use its principle. Experimental procedure of positron source preparation (radioactive ^{22}Na) for these methods is difficult task. The unusual small sample dimensions of studied irradiated Eurofer 97 alloy form difficult conditions for its manufacturing. The unique sodium positron source proper preparation is confirmed by positron annihilation lifetime spectroscopy (PALS) and coincidence Doppler broadening spectroscopy (CDBS) in Paul Scherrer Institute in Switzerland.

2. Introduction

Combination of used positron annihilation techniques is able to describe complex characterization of various alloys microstructure. Positron lifetime spectroscopy determines the amount and size of vacancies and their agglomerates in lattice. On the other hand, the energy/momentum distribution techniques as Doppler broadening spectroscopy or angular correlation of annihilation radiation (ACAR) describe quantity of vacancies and their clusters, their concentration, microstructural and chemical changes. The main part of this article consists of description of unusual small positron source preparation for coincidence Doppler broadening spectroscopy, which uses two detectors engagement for better background reduction. For as prepared source verification was used non irradiated and irradiated ferritic/martensitic alloy Eurofer 97, which is perspective material for using in fission and fusion facilities (first wall of DEMO (DEMOnstration Power Plant) fusion reactor [1]).

3. Positron source preparation

As was written above the conventional sources for positrons study couldn't be used in consideration of small sample dimensions. It was necessary to prepare adequate positron source with small active dimensions and sufficient activity. The sodium salt in aqueous solution with 3.7MBq total activity was chosen as source of positron for that purpose. Even sources activities as low as <1MBq [2] are sufficient for conventional positron lifetime spectroscopy and Doppler broadening spectroscopy. Our decision was to have stronger one for higher count rate and therefore shorter time of measurements. On the contrary, the positron beams and angular correlation techniques require stronger sources about 4GBq [3].

Decay scheme of radioactive isotope is shown in Fig.1, whose characteristic decay reaction is $^{22}\text{Na} \rightarrow {}^{22}\text{Ne} + \beta^{+} + \nu_e + \gamma$.

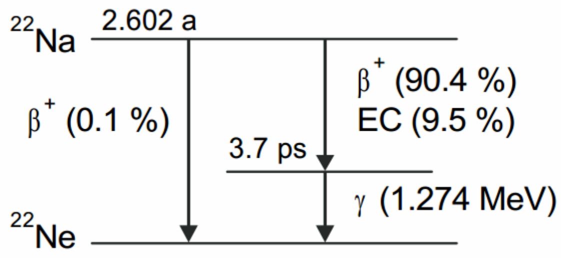

Fig.1: *Decay scheme of the radioactive isotope 22Na, used for positron source preparation. Electron capture (EC) is process with lower probability as well as direct transition to the Ne ground state. Half-life of isotope is 2.6 years [2].*

The kapton tape with thickness 20μm, with silicon adhesive layer with thickness 43μm was the first bearing layer for positron source. This layer, with punched out opening (2.3mm radius), serve for fixing of top components. The punch out small ring from kapton foil (7.5μm thickness, 3mm radius) was glued on first bearing layer. Twelve drops of sodium salt solution were located in the centre of ring. The process of 0.5ml solution drops evaporating took adequately long time to prevent small salt crystals growing. During the whole process of preparation it is necessary to reduce possibilities to break the thin kapton foil, in order to prevent possible contamination of surrounding. In the end of the process the final upper kapton foil covered dry-out sodium solution and bearing layer. The detailed description of positron sodium source preparation scheme is in Fig.2.

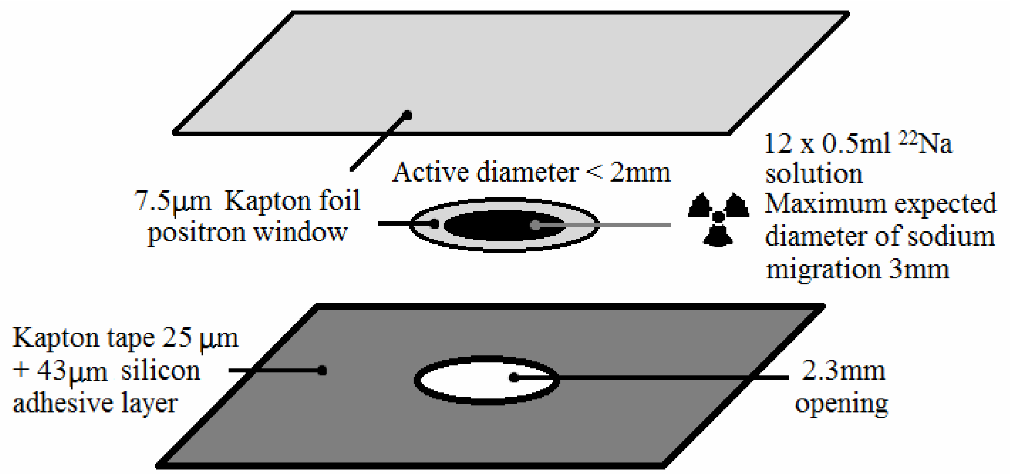

Fig.2: *Positron source layers detail description with specific dimensions and 3.5MBq final activity. It consists from 12x0.5ml drops of ^{22}Na solution.*

The prepared positron source with total activity 3.5MBq was used for CDBS measurements in samples-source sandwich arrangement. Dose rate of such prepared source was 100μSv/h in 10cm distance. Total time of positron source preparation was 12 hours. It was necessary to follow radiation safety rules during manufacturing (strong led shielding, dosimeters, masks, gloves, glasses, coats, etc.) in order to minimize the exposure of person and minimize the environmental contamination risk.

The small positron fraction annihilate in positron source itself. It depends of foil thickness and atomic number of sample. This fraction is between 2-15% [2]. Amount of annihilated positrons in our source is 14.8% (measured by positron lifetime spectroscopy). The high activity, fraction of positrons annihilated in source itself and its unique small dimensions satisfied condition of well prepared source.

4. Materials specification

For a demonstration of successful source preparation and determination of experiments conditions, small tensile samples of Eurofer 97 materials were investigated. The specimen was made from a 25mm thick plate of Eurofer 97 steel (Fe-Cr(8.93), Ni(0.22), Mo(0.0015), Mn(0.47), Ti(0.009), V(0.20), Nb(0.002), W(1.07), Ta(0.14), Cu(0.003), C(0.12), Si(0.06), P(<0.005), S(0.004), B(<0.001), N(0.018)(wt.%)), which was normalized at 980°C for 27 min and tempered at 760°C for 1.5 h and followed by air cooling. The mean grain size is about 16±2μm.

Samples were irradiated in 2000 in frame of STIP-II programme at the Swiss spallation neutron source (SINQ) in Paul Scherrer Institute with total irradiation time 16 months. More than 2000 samples of 40 different materials, including Eurofer 97, were irradiated [4]. Depending of rod position within the target, displacement damage and irradiation temperature were ranging 100-400°C and 7 – 22DPA. The Eurofer97 sample investigated in the presented experiment was designated as IJ15. The calculated irradiation parameters are:DPA 7.4; appm(He)=565; appm(H)=2240; irradiation temperature range 119-147°C. With weight 0.05189±0.00020g the dose rate of IJ15 in 2cm distance was 310μSv/h.

5. CDBS measurements

CDBS is method for positron annihilation momentum distribution determination in measured materials. This can be used for study of electron distribution in solids and for defects investigation. The two quanta of 511keV (511keV corresponds to momentum $p_L=0x10^{-3}m_0c$) energies are emitted after annihilation of positron and electron in material. Energy values around this specific energy correspond to annihilation with valence electrons. And just S parameter describes this effect and therefore it determines open volume defects (vacancies and vacancy clusters).

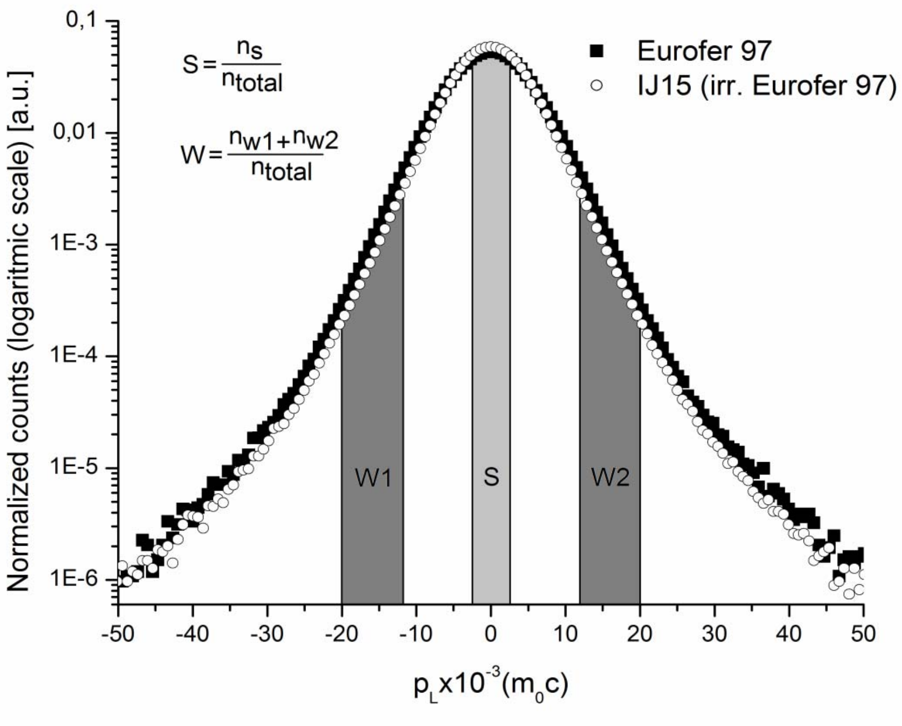

Fig.3: *One-dimensional momentum distribution spectra from CDBS measurements of Eurofer 97 and IJ15 (irradiated Eurofer 97) with S and W parameter calculations.*

Range for S parameter determination was chosen from -2.5 to $2.5 \times 10^{-3} m_0 c$ of momentum. Energy values more distance from 511keV describe annihilation with core electrons and W parameter describes its characterization with the meaning of microstructural changes in lattice. The momentum values for W determination are from -20 to $-12 \times 10^{-3} m_0 c$ and from 12 to $20 \times 10^{-3} m_0 c$.

The measured one-dimensional momentum distribution spectra of Eurofer 97 and IJ15 are shown in Fig.3. Total numbers of each measurement counts were over 160mil in less than 2 days. The count rate was over 1100 counts per second. Resolution of CDBS measurement was 0.173keV. FWHM for Eurofer 97 spectrum after Gauss fitting is $4.966 \pm 0.01725 \times 10^{-3} m_0 c$ and for IJ15 it is $4.493 \pm 0.01499 \times 10^{-3} m_0 c$. The final values of S and W parameters are possible determine from equations in Fig.3. They are: S(Eurofer 97)=0.452, W(Eurofer 97)=0.0362, S(IJ15)=0.491 and W(IJ15)=0.0259. The changes after irradiation of material are possible to determine using these values. Higher value of S(IJ15) shows defects creation. In the other hand lower W parameter describes minor annihilation on core electrons and therefore microstructural changes in material caused irradiation process conditions (He bubbles creation, high temperature, etc.).

6. Results and discussion

The strong sodium positron source (total activity 3.5MBq) with very small active dimensions was made. Reason of that was to use it for measurement of positrons annihilation methods of highly irradiated small samples (Eurofer 97, irradiated in SINQ during STIP II procedure) with dimensions 2.5x2.5x0.5mm. The total time of CDBS measurements was less than two days with above 160mil total counts number, with sufficient count rate.

All components dimensions of source are small (kapton foil, kapton tape with adhesive layer) for reason of positron annihilation reduction in source itself (14.8%). Prepared source can be simply used with two identical samples in sandwich arrangement. It is necessary to preserve sensitivity of manipulation with it. By tearing of source the radioactive contamination of samples can occur. Measured one dimensional momentum distribution spectra of CDBS proved correct source preparation. The result values from calculated S and W parameters satisfy condition of its determination.

Acknowledgement

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