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# POTENTIAL OF POSITRON ANNIHILATION LIFETIME SPECTROSCOPY IN MICROSTRUCTURAL STUDY OF NUCLEAR REACTOR STEELS

**Vladimír Kršjak**

The positron annihilation lifetime spectroscopy (PALS) can be very suitable complement to many experiments on the RPV steels. The most interesting examples of such applications published in the scientific papers in last decade as well as our recent experiments are summarized in this paper.

K e y w o r d s: science, reactor steels, positron annihilation, radiation damage

## 1 INTRODUCTION

If we consider positron annihilation from the general point of view, we could say that the positron annihilation spectroscopy (PAS) is nowadays well recognized as

- A powerful tool of microstructure investigations of condensed matter,
- A unique spectroscopic technique for the study of vacancy type defects,
- A technique capable to register very low concentration of defects in crystal lattice,
- One of suitable techniques for defects study in the near surface region [1 - 4].

The utility of positron annihilation studies of reactor pressure vessel steels (RPV) relies on the fact that the characteristics of the annihilation process depend almost entirely on the initial state of the positron-many-electron system. Since energetic positrons are rapidly thermalized after entering condensed matter, the characteristics of the annihilation process in most cases depend on the initial state of the many-electrons system where positrons annihilate. The energies, moment, and time of the gamma rays emitted during the annihilation may be measured with high precision using the modern detector systems. Therefore, studying the positron annihilation process characteristics can identify the state of electrons in metallic substances.

Besides the positron lifetime measurement, momentum distribution of the annihilation gamma rays can be measured using the method of angular correlation of the two collinear annihilation gamma rays, and energy distribution can be determined using the method of Doppler broadening.

Hence, the lifetime method yields information regarding the electron density in the region of electron-positron overlap through the determination of positron decay rates from various states in the metal. The method of angular correlation and Doppler broadening (momentum techniques) gives information regarding the electron moment in this overlap region through the shape of the angular ($\Theta$) and energy ($E$) distributions of the annihilating $\gamma$-rays. These shapes are usually characterized by one or more "line shape parameters". The most commonly used ones are the normalized peak-counts determined in an interval around 0 (in angular correlation) or $E = 0$ (in Doppler broadening). These measure in various, but qualitatively equal, ways the change of annihilations with electrons of low momentum relative to those with high momentum. These changes in the annihilation characteristics yield information whether the positron annihilates in a defect-free region of the crystal or from a defect site.

## 2 PALS APPLICATIONS ON RPV STEELS

Since mid 1980-s PAS has been extensively used in the study of RPV steels [5-19]. The positron annihilation technique can give essential information about the deterioration of the mechanical properties of RPV steels during their irradiation, which is known as neutron embrittlement. The lifetime of positrons trapped at radiation-induced vacancies, vacancy-impurity pairs, dislocations, micro-voids, *etc* is longer than that of free positrons in perfect region of the same material. As a result of the presence of open-volume defects, the average positron lifetime observed in structural materials is found to increase with radiation damage.

The embrittlement of steel is a very complicated process depending on many factors (thermal and radiation treatment, chemical compositions, ageing, *etc*). Properties of the RPV steels and influence of thermal and neutron treatment on these properties are routinely investigated by macroscopic methods such as Charpy V-notch and tensile tests. A number of semi-empirical laws, based

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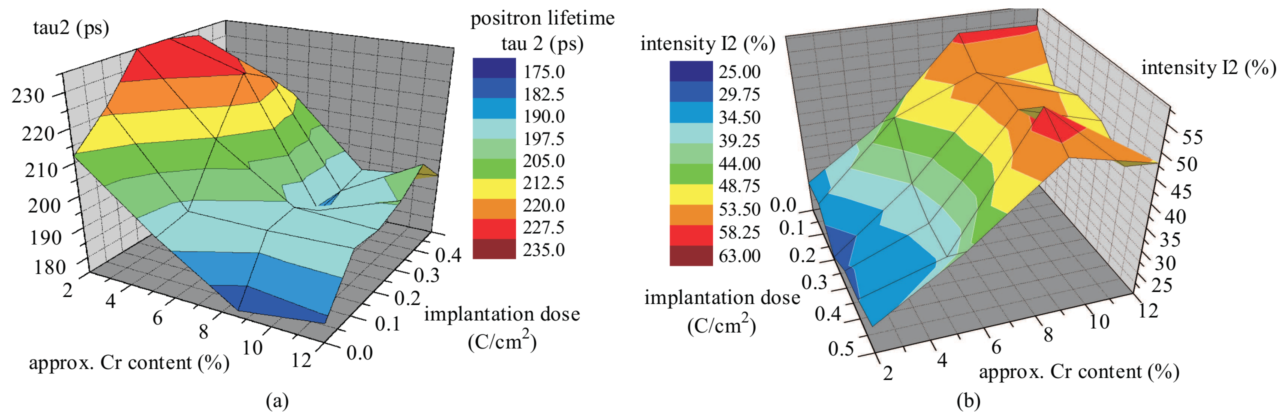


**Fig. 1.** Positron annihilation in defects (component 2): (a) – positron lifetime, (b)–intensity of the annihilation

on macroscopic data have been established, but unfortunately, these laws are not completely consistent with all data and do not provide the desired accuracy. Therefore, many additional test methods [20-22] have been developed to unravel the complex microscopic mechanisms responsible for RPV steel embrittlement.

RPV embrittlement poses one of limiting factors in the lifetime of vessels of today's nuclear power plant (NPP) This problem is very serious in Eastern (Russian) types of nuclear reactors (VVER). It is due to the narrower gap between the outside surface of the core barrel and the inside surface of the RPV than in Western RPV's. The neutron flux and consequently neutron fluency on the RPV wall is generally higher for VVER-440 type reactors than for other equivalent types. This influence of neutron flux (even neutrons of energy over 0.5 MeV) on RPV embrittlement is much more pronounced than other contributions, *eg* from a coolant temperature or from the operational pressure in the primary circuit.

According to previous reports [23-25] it seems to be generally accepted that even in the Western types of RPV steels containing more than 0.1 wt% of Cu, the Cu- and P-rich precipitates play a dominant role in thermal and neutron embrittlement. In case of Eastern-type RPV steels, comprehensive PAS studies [26-28] have suggested that carbide formation is an observable additional microstructural mechanism.

The positron annihilation lifetime spectroscopy can be very suitable complement to many experiments on the RPV steels. The most interesting examples of such applications published in the scientific papers in last decade as well as our recent experiments in the area of nuclear structural materials are summarized in following.

### 2.1 Differences in reactor pressure steels according to chemical composition

The direct influence of the chemical composition on the positron annihilation in low-alloy steels cannot be debated. However, the alloying elements significantly affect the steel properties during the fabrication process (thermal treatment, rolling) and final microstructure contains various density of vacancy type defects. With appropriate knowledge of the studied material and the specimen history we can gather additional important information from the PAS technique. Nevertheless, this information is more relevant, the bigger is set of the studied materials which simplifies the interpretation of the spectra. One of the largest works in this field was published by Slugeň *et al* in 1999 [18] where 5 western and 3 Russian RPV steels have been studied using PAS, MS (Mössbauer spectroscopy) and TEM techniques.

This approach is even more suitable for the investigation of the model alloys, where well defined chemical composition and fabrication process is available. Following, for instance the effect of individual alloying element in the steel can be studied [29].

### 2.2 Irradiation effects observation

The positron annihilation can register an increase of vacancy type defects due to irradiation [5, 12, 15], however, the loads during RPV operating lifetime 1020 n/m2 are too small to register large changes in positron annihilation parameters. Nevertheless, even this low density of point defects in the RPV steel can be register with PAS [30].

For more significant damage rate acquirement, the neutron flux can be with advantage simulated with ion implantation [29, 31, 32]. The implanted layer is usually the near surface region and can be studied with corresponding depth sensitive technique. Although the conventional 22Na positron source lifetime technique, can be often used for the qualitative characteristics of the nature of this kind of microstructural damage. In Fig. 1 can be seen four different Fe-Cr binary alloys (with different Cr content) implanted by 250 keV He ions and measured with conventional PALS technique. It is clear that character of the implantation-induced defects is changing with the implantation dose (a) with almost constant intensity of annihilation (b). This corresponds with the choice of approx. 9 wt% of Cr content in ferritic/martensitic steels as optimal because of the maximum resistance the DBTT shift after irradiation of this material. It seems also that the after specific dose about 0.3C (local damage 50 DPA) the crystal lattice is so damaged that new Frenkel pairs are not created anymore and instead these defects recombine (Fig. 1a).

## 2.3 Optimal annealing temperature determination

PAS techniques are very sensitive on annealing. Many works were focused on registration of optimal annealing procedures. One of the most appreciate picture in the "positron community" from industrial point of view was PAS confirmation of optimal annealing temperature of 15Kh2MFA RPV steel (see Fig.2). This picture shows the temperature region of annealing and informs that annealing at 500 causes creation of next large amount of small defects connected to a huge, probably the carbide precipitation [30].

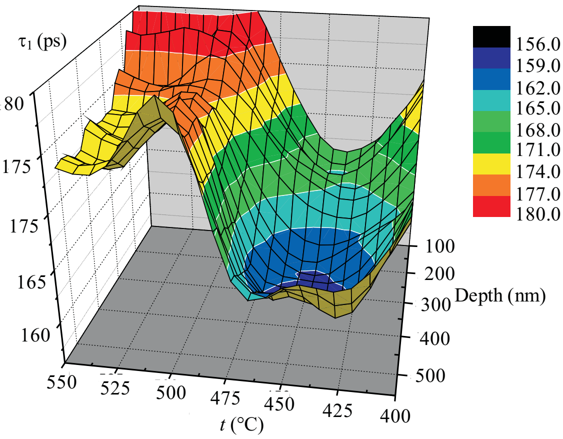


**Fig. 2.** The 3D presentation of positron lifetime results of irradiated (neutron fluence 1.25x1024 m-2) and annealed Sv-10KhMFT steel (WWER-440 weld metal)

## 2.4 Depth profiling

Depth profiled positron lifetime measurement is unique ability of the slow positron beam facilities. The investigation of the vacancy type defects in near surface region cannot be measured in non-destructive way by any other experimental technique.

This kind of investigation of defects up to one micrometer below the surface has been performed on the non-implanted Fe11.62%Cr sample and three Fe11.62%Cr samples with different damage levels using positron implantation energies between 1 keV and 18 keV. The depth profiles were measured with the Pulsed Low Energy Positron System (PLEPS) [33] at the high intensity positron source NEPOMUC [34] at the research reactor FRMII at TUM.

Figure 3 shows the positron mean lifetime as a function of helium implantation dose and mean positron implantation depth. The positron mean lifetime (MLT) is increasing with the implantation dose, thus indicating the creation of defects due to implantation. The increase of the MLT close to the surface (< 200 nm below the surface) is probably due to positrons annihilating in surface oxide layer. At higher depths the course of the MLT depth profile corresponds to the expected zone of maximum damage.

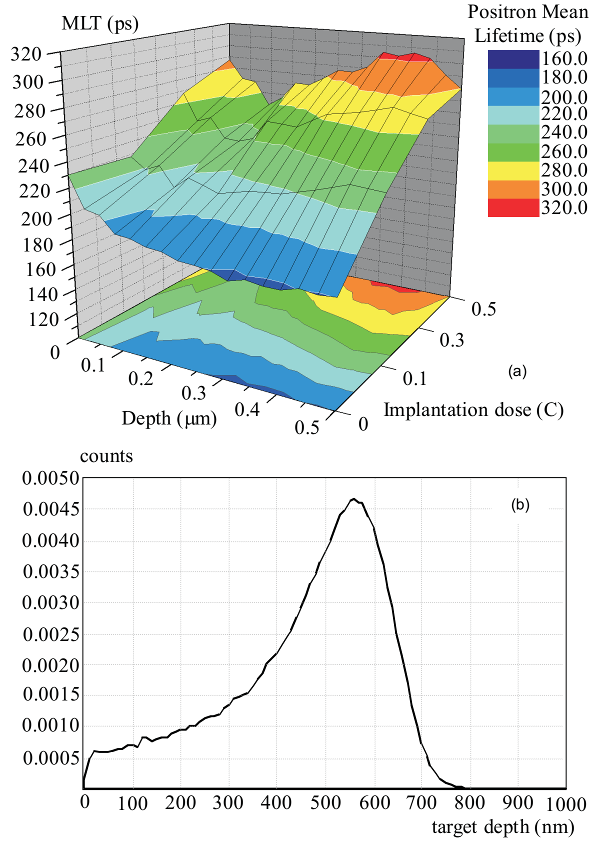


**Fig. 3.** (a) – Positron mean lifetime in different treated Fe11.62%Cr alloy, (b) – Implantation profile of He ions

When the original calculated depth profile of the implantation (Fig.3(b)) is considered, one can see good relation to the experimental PLEPS results. The experimental confirmation of the defects distribution with the scanning electron microscopy (SEM) is described in the next chapter.

## 2.5 Possibility of unique PAS correlation with other non-destructive techniques

Our recent experiments show, that positron lifetime measurements can be with advantage correlated with different experimental non-destructive techniques. Besides the more complex information about the material microstructure, there is also great advantage in the increasing of reliability of the data interpretation which is a difficult process when the complex materials like steels are considered. The TEM confirmation of PAS-predicted defects in different alloys and steels have been published in scientific papers last two decades [18, 20, 29, 35]. However our recent measurements of the X-ray diffraction and SEM imaging show that there is conjunction with

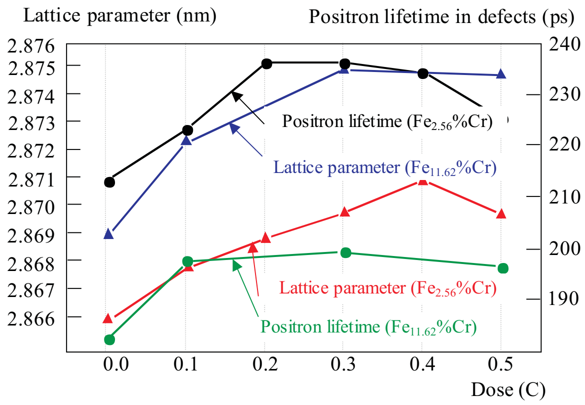


**Fig. 4.** Lattice parameter *vs* positron lifetime in defects in helium implanted Fe-Cr alloys (XRD error bars are negligible; error of the positron MLT is ±5 ps)

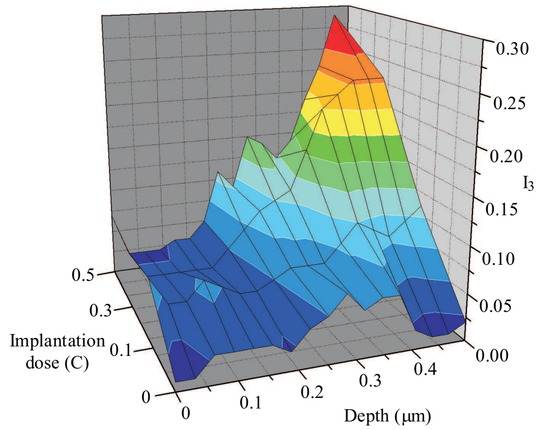


**Fig. 5.** Intensity of annihilation in large defects (voids) measured in Fe11.62%Cr alloy

the positron lifetime measurements as well. Also measurements of He-implanted Fe-Cr alloys using the conventional and slow positron lifetime measurements have been evaluated with the common goals.

As can be seen on Fig. 4 the positron mean lifetime shows very similar behavior as the lattice parameter measured by XRD, with the increasing implantation dose. However, there are clearly more issues that play a role here (chemical composition, individual microstructure of as-received materials *etc*).

The possibility of depth profiling of the PLEPS measurements enables the correlation of the data with the calculated defects distribution based on the radiation-matter interaction. As can be seen on the Fig. 5, the intensity of annihilation in large voids (I3) corresponds very well with the simulation of the collision events distribution (Fig. 3(b)). This result presumes presence of large vacancy agglomerates (¿50 vacancies; ¿1nm) in the depth of damage peak (500 - 700 nm). This later experimentally confirmed using the SEM technique. Small-angle cut through the implanted layer ensured additional magnification and finally the microstructure was observed as can it be seen in the Fig. 6. The SEM images confirm that the most damage is accumulated in the mean stopping depth of the He ions. This is the cause of blistering effect, which can be seen in the SEM image.

In the case of He-implanted Fe-Cr alloys study, the conventional PALS technique as well as PLEPS measure-

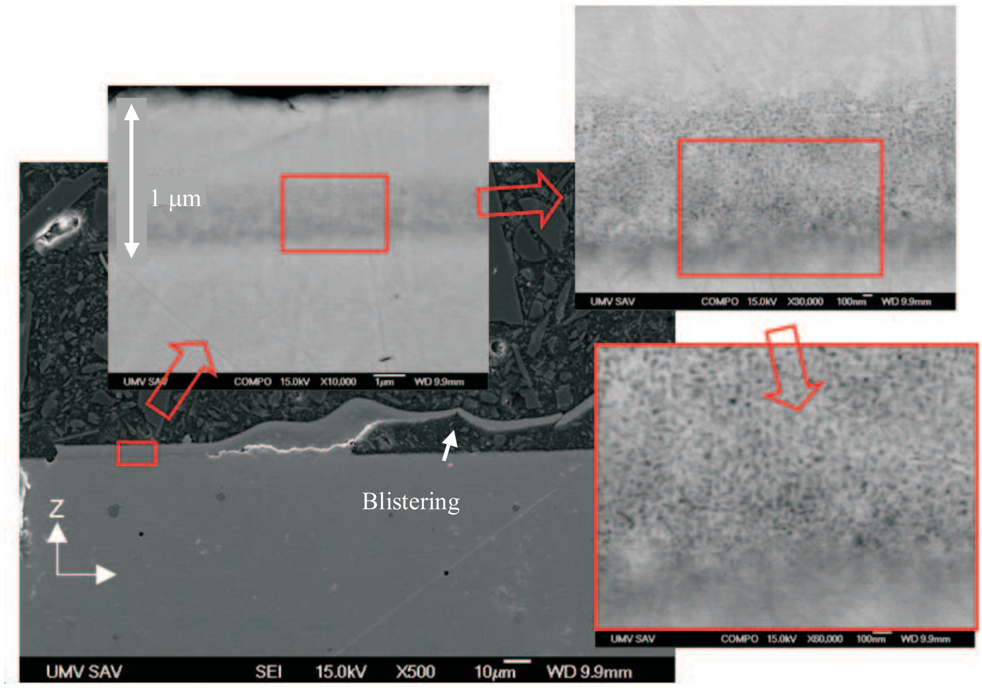


**Fig. 6.** Backscattered electron images of the Fe-Cr alloy implanted with He (0.3C)

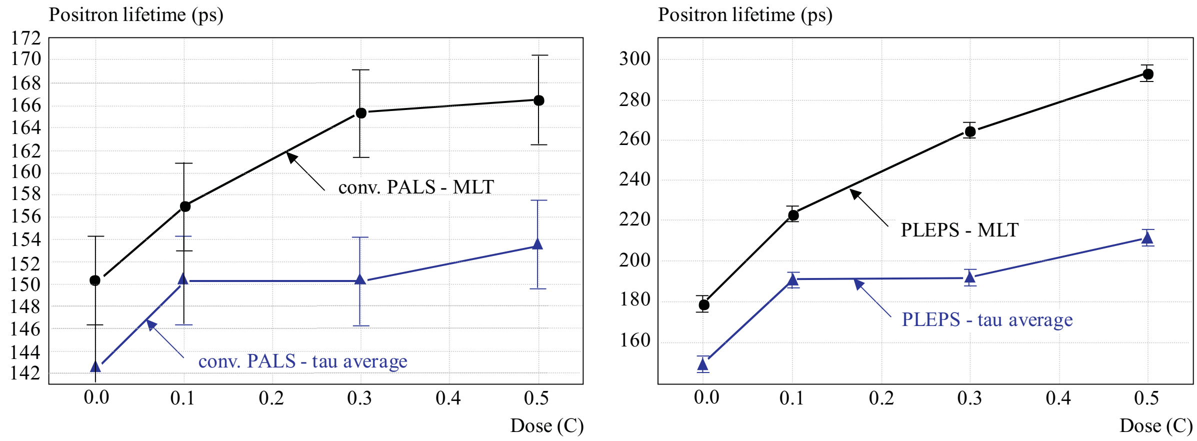


**Fig. 7.** (a) – positron mean lifetime and AV from the conventional PALS technique, (b) – the PLEPS measurements of the Fe11.62%Cr alloy implanted with He ions (PLEPS data corresponds to arithmetic mean of values from 200 - 600 nm of depth)

ments shows significant changes in the studied parameters as an effect of helium implantation. The increasing lifetime due to helium implantation was observed in the MLT as well as the AV parameter. This behavior was more significant in the PLEPS measurements (due to more suitable depth sensitivity) than in conventional PALS. Nevertheless, the shape is almost identical for both techniques, see Fig. 7(a) and (b).

### 2.6 Positron microscope

Even the concept of positron microscope will probably not be able to effectively substitute the electron microscope in high resolution applications, there are important advantages and potentialities of this technique. Besides the image of the microstructural defect, the positron microscope can reliably characterize the type of this defects and therefore provide the more qualitative characteristics of the material. The first lifetime measurements obtained by scanning positron microscope (SPM) has been published by group from Universität der Bundeswehr [36]. They discuss the measurements of GaAs sample with a small surface scratch by electron microscope and SPM. The image of the same specimen from both techniques is compared in Fig. 8.

## 3 CONCLUSIONS

Positron annihilation spectroscopy - lifetime measurements is a unique tool for for probing the electronic and atomic structure of the condensed matter. The behaviour of vacancy type defects in the microstructure of radiation damaged steels as the key factor of the RPV integrity can be very well determined with this technique. In fact there is no any other technique capable to investigate atomic defects of low concentration and in non-destructive way. The PALS measurements together with conventional destructive tests can reliably characterize the microstructure of model alloys after various radiation and thermal treatment and help not only to evaluate the state of existing RPV steels but also to support the research of new developing materials.

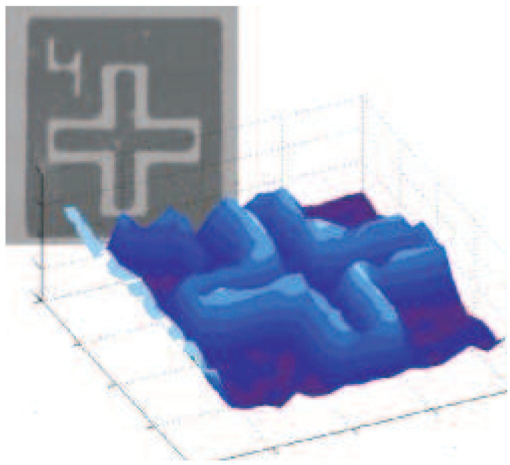

**Fig. 8.** An electron microscope image (top) contains less information on a test pattern of surface defects than that of the new scanning positron microscope (bottom)

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