Investigation of materials for fusion power reactors

A. Bouhaddane, V. Slugeň, S. Sojak, J. Veterníková, M. Petriska and I. Bartošová

Department of Nuclear Physics and technology: Faculty of Electrical Engineering and Information Technology, Slovak University of Technology, Ilkovičova 3, 812 19 Bratislava 1, Slovakia

E-mail: amine.bouhaddane@centrum.sk

Abstract. The possibility of application of nuclear-physical methods to observe radiation damage to structural materials of nuclear facilities is nowadays a very actual topic. The radiation damage to materials of advanced nuclear facilities, caused by extreme radiation stress, is a process, which significantly limits their operational life as well as their safety. In the centre of our interest is the study of the radiation degradation and activation of the metals and alloys for the new nuclear facilities (Generation IV fission reactors, fusion reactors ITER and DEMO). The observation of the microstructure changes in the reactor steels is based on experimental investigation using the method of positron annihilation spectroscopy (PAS). The experimental part of the work contains measurements focused on model reactor alloys and ODS steels. There were 12 model reactor steels and 3 ODS steels. We were investigating the influence of chemical composition on the production of defects in crystal lattice. With application of the LT 9 program, the spectra of specimen have been evaluated and the most convenient samples have been determined.

1. Introduction

Rising energy needs of human society require the creation of modern electric sources. These are for example the Generation IV fission reactors and thermonuclear reactors. Running these modern facilities is a difficult issue in terms of requirements for materials. Increased radiation damage to materials significantly limits operational life as well as the safety of reactors. It is necessary to search and explore more resistant substances that find their application in these facilities. This paper is focused on the fusion reactors and materials to use for their construction. The detection of the effect of chemical composition of construction materials on their structure and hence the resistance to radiation stress plays an important role. This work will refer the possibility of analysis using positron annihilation spectroscopy and program LifeTime 9. Proper interpretation of the results obtained on the basis of broad knowledge related to the topic is essential.

2. Radiation damage

2.1. Interaction of radiation with matter

During operational lifetime of nuclear facilities there is radiation damage. The radiations of concern here include charged particles such as electrons (beta particles), protons, alphas and fission fragment ions, and the neutral radiations including photons (gamma and X rays) and neutrons.
Table 1 compares some key characteristics of the radiation, including charge, mass and range in air. For a kinetic energy of 1 MeV, the electron acts as a relativistic particle (0.94c). For the same energy, the heavier particles are slower, stopped easier and deposit their entire energy over a shorter distance [1].

| Characteristic | Radiation (E_k = 1MeV) | Alpha (α) | Proton (p) | Beta Electron (e) | Photon (γ or X ray) | Neutron (n) |
|----------------|-------------------------|-----------|------------|------------------|---------------------|-------------|
| Symbol         |                         | ^4_2α or He ^2+ | ^1_p or H^1+ | ^0_1e or β       | ^0_0γ           | ^1_0n       |
| Charge         |                         | 2         | 1          | -1               | 0                  | 0           |
| Ionization     |                         | Direct    | Direct     | Direct           | Indirect           | Indirect    |
| Mass (u)       |                         | 4.001506  | 1.007276   | 0.0005486        | -                  | 1.008665    |
| Velocity (m/s) |                         | 6.94E+05  | 1.38E+07   | 2.82E+08         | 3.00E+00          | 1.38E+07    |
| Speed of light [% of c] |               | 2.30      | 4.60       | 94.10            | 100                | 4.60        |
| Range in air [m] |                       | 5.6x10^-3 | 1.81x10^-2 | 3.19x10^-2      | 820 *              | 392.5 *     |

* range based on a 99.9% reduction

2.2. Radiation effects in materials

Due to influence of irradiation on structural lattice and its interaction there are these processes [1]:

- **Impurity production**, that is, transmutation of nuclei into other nuclei which themselves may be radioactive; this mechanism is caused by neutrons through fission and activation (capture). Impurities can also be deposited from the creation of hydrogen or helium when a proton or an alpha particle, respectively, becomes neutralized in the material of passage.
- **Atom displacement** from their normal position in the structure of the material; displacement atoms may leave lattice vacancies and lodge in interstitial locations or cause interchange of dissimilar atoms in the lattice structure.
- **Ionization**, that is, the removal of electrons from atoms in the material and the formation of ion pairs in the path of the charged particles
- **Large energy release** in a small volume, which can result in thermal heating of the material. This may be especially important in those cases where the material is a radiation shield [1].

2.3. Types of defects in materials

Few, if any, crystals are perfect in that all unit cells consist of the ideal arrangement of atoms or molecules and all cells line up in a three dimensional space with no distortion. Some cells may have one or more atoms less whereas others may have one or more atoms than the ideal unit cell. The imperfections in crystal are called crystal defects. Defects may be classified into four categories depending on their dimension [2]:

- **0D, Point defects**: atoms missing or in irregular places in the lattice vacancies, interstitials, impurities),
- **1D, Linear defects**: groups of atoms in irregular positions (e.g. screw and edge dislocations),
- **2D, Planar defects**: the interfaces between homogeneous regions of the material (grain boundaries, external surfaces),
- **3D, Volume defects**: extended defects (voids, Stacking Fault Tetrahedra, pores, cracks).

3. Structural materials of fusion reactors

Nuclear fusion is a process by which multiple atomic nuclei join together to form a single heavier nucleus. It normally takes place on the surface of sun, when Hydrogen nuclei collide and create heavier Helium atoms while releasing tremendous amounts of energy in the process. To achieve such a process you need to get Hydrogen atoms close to each other to overcome repulsive Coulomb force. It is possible by accumulating energy. A fusion reaction requires nonetheless temperatures of ~150x10^6 K to take place - ten times higher that the H-H reaction occurring at the Sun's core. At extreme
temperatures, electrons are separated from nuclei and a gas becomes plasma - a hot, electrically charged gas. In a star as in a fusion device, plasmas provide the environment in which light elements can fuse and yield energy. Currently there are few facilities which experiments with fusion reaction (JET) and new facility ITER (International Thermonuclear Experimental Reactor) which is in building process. The fusion reaction will be achieved in a TOKAMAK (Toroidal chamber with magnetic coils) device that uses magnetic fields to contain and control the hot plasma [3].

In fusion reactors, the plasma facing (first wall, divertor) and breeding blanket components will be exposed to plasma particles and electromagnetic radiation and will suffer from irradiation by an intense flux of 14.1 MeV neutrons. The high-energy fusion neutrons will produce atomic displacement cascades and nuclear transmutation reactions within the irradiated materials. From the point of view of materials science, atomic displacement cascades induce the formation of point defects (i.e. vacancies, interstitial atoms, vacancy and interstitial clusters) and segregation of alloying elements, while nuclear transmutation reactions produce helium and/or hydrogen gas atoms. The final microstructure of the irradiated materials results from a balance between environmental conditions, especially radiation damage and temperature, and stress/strain histories [4].

Extreme conditions of a fusion reaction lead into high demands on structural materials. What we expect from these materials is high resistance to radiation damage, so they can be in operation for a long period and a low level of activation. It means that it will be possible to handle them in a shorter period after removing from a power plant. For this purpose, new materials were designed.

3.1. RAFM steels

The development of ferritic/martensitic steels for applications in fusion devices emanates from the limitations of the austenitic stainless steels and the promising high dose experience with ferritic/martensitic steel fuel cladding in liquid metal cooled fast reactors. The austenitic steels suffer from helium embrittlement at elevated temperatures and swell to a degree not acceptable for fusion reactor components. The ferritic/martensitic steels exhibit superior performance to austenitic steels in both swelling and helium embrittlement resistance. At a temperature of about 575 K the swelling rate of ferritic/martensitic steels is about 1 vol% after 100 dpa, while it is about 1 vol% after 10 dpa for typical austenitic steels. Ferritic/martensitic steels also exhibit a better surface heat capability than austenitic steels (5.4 kW m⁻¹ at 675 K, i.e. about three times that of austenitic steels) [5], favourable cost, availability and service experience, and their good compatibility with aqueous, gaseous and liquid metal coolants permits a range of design options. At the same time ferritic/martensitic steels with alloying elements such as chromium, some tungsten, vanadium and tantalum activate little compared to conventional austenitic stainless steels with nickel and molybdenum. In addition, manganese-stabilized austenitic stainless steels are not attractive due to high decay heat (safety) concerns. In Europe, Japan and Russia it has been demonstrated that it is now feasible to produce RAFM steels on an industrial scale with sufficiently low impurity levels [6–11]. The promise remains to produce low activation varieties that allow recycling within a century [12, 13]. Main RAFM steels that are being investigated include the Chinese low activation martensitic (CLAM) steel, the European EUROFER 97 alloy, the Japanese F82H and JLF-1 alloys, and the Russian RUSFER-EK-181 alloy. Their composition lies in the following range: Fe-(7.5–12)Cr-(1.1–2)W-(0.15–0.25)V, in weight percent.

The major issues remaining with the RAFM steels are:

- Limited strength at high temperatures. RAFM steels show a drop in tensile strength at about 825K and a strong reduction in creep strength at \( T = 875 \) K. In addition, softening occurs during cyclic loading, which may lead to maximum allowable loads much smaller than the limits predicted by the current design rules.
- Irradiation-induced hardening and embrittlement effects at temperatures \( T < 675 \) K.
- The production of high amounts of helium and hydrogen under fusion neutron irradiation conditions.
• Fabrication techniques, in particular joining, as RAFM steels are more difficult to weld than austenitic stainless steels.
• Limited database at high irradiation doses [4].

3.2. ODS steels
Oxide dispersion strengthened (ODS) ferritic alloys (e.g. MA957, MA956) developed by the International Nickel Company (INCO) are also considered for application in future nuclear facilities. The oxide dispersion, which is in case of MA957 & MA956 yttrium, is intending to provide high temperature strength. The alloys are produced by “mechanical alloying”, which is a powder metallurgy process which distributes the alloy elements homogeneously on a very fine scale [14]. Chemical composition of yttrium strengthened ODS alloys with addition of titanium and molybdenum in order to improve strength, ductility and oxidation resistance are shown in Table 3. They belong to main candidate materials for first wall (plasma facing and structural material) and breeding blanket components.

4. Measurement
Practical part is aimed on investigation of influence of chosen elements and impurities on mechanical behaviour and sensitivity to radiation damage of model reactor steels used in present reactors (VVER-1000) and of ODS steels. All primary material preparation of reactor steels was coordinated by the scientific technological company OOO "Nucleon-M" in Moscow, Russia in 2004.

4.1. Chemical composition of samples
The nominal base compositions of the 12 model steels are derived from typical Russian and Western RPV materials for:
1. WWER-1000 (15Cr2NiMoV steel grade) and
2. PWR (ASTM A533-B steel grade).
This choice is explained by striving to reveal the possible distinction of different compositions in their sensitivity to the deleterious element components. The studying material includes mainly various Ni and Mn combinations and nearly the same range of Si content (see table 2).

| Sample | C  | Si  | Mn  | Cr  | Ni  | Mo  | V   | Cu  | S   | P   |
|--------|----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| Ap631  | 0.11 | 0.28 | 0.43 | 2.22 | <0.02 | 0.71 | 0.10 | 0.09 | 0.008 | 0.010 |
| Bp632  | 0.11 | 0.26 | 0.38 | 2.19 | 0.99 | 0.70 | 0.10 | 0.10 | 0.008 | 0.010 |
| Cp633  | 0.12 | 0.24 | 0.38 | 2.13 | 2.00 | 0.69 | 0.10 | 0.10 | 0.008 | 0.010 |
| Dp634  | 0.11 | 0.23 | 0.83 | 2.13 | 2.00 | 0.68 | 0.10 | 0.09 | 0.008 | 0.009 |
| Ep641  | 0.12 | 0.33 | 0.77 | 2.16 | 1.02 | 0.70 | 0.10 | 0.10 | 0.008 | 0.009 |
| Fp642  | 0.12 | 0.33 | 1.37 | 2.15 | 1.02 | 0.70 | 0.10 | 0.10 | 0.008 | 0.010 |
| Gp643  | 0.11 | 0.32 | 1.36 | 2.06 | 1.99 | 0.69 | 0.10 | 0.10 | 0.008 | 0.009 |
| Hp644  | 0.12 | 0.51 | 1.31 | 2.07 | 2.00 | 0.69 | 0.10 | 0.10 | 0.008 | 0.010 |
| Kp301  | 0.17 | 0.35 | 0.78 | 0.10 | 0.58 | 0.64 | -   | 0.07 | 0.005 | 0.009 |
| Lp302  | 0.18 | 0.35 | 0.77 | 0.08 | 0.96 | 0.63 | -   | 0.05 | 0.005 | 0.010 |
| Mp303  | 0.16 | 0.37 | 0.74 | 0.09 | 1.90 | 0.61 | -   | 0.05 | 0.005 | 0.010 |
| Np304  | 0.16 | 0.33 | 1.27 | 0.07 | 1.97 | 0.63 | -   | 0.06 | 0.005 | 0.010 |
Table 3. Chemical composition of ODS steel MA957 (mass %) [15].

| Sample | C   | Si  | Mn  | Ni  | Cr  | Mo  | Ti  | Al  | Y₂O₃* |
|--------|-----|-----|-----|-----|-----|-----|-----|-----|-------|
| MA957  | 0.03| 0.04| 0.09| 0.13| 13.7| 0.30| 0.98| 0.03| 0.28  |
| MA956  | 0.03| 0.05| 0.06| 0.11| 21.7| <0.05| 0.33| 5.77| 0.38  |

* Estimated from yttrium content assuming it is contained in the form of Y₂O₃

These limits reflect the common range of Ni and Mn variations in WWER and PWR RPV materials. Potentially, silicon plays an active role in the radiation damage of ferritic steels but its content in RPV materials varies within rather a narrow range and that is why this element was concluded to be of secondary importance. For this reason the most part of the prepared materials should contain the nominal 0.35 % of Si excluding one model steel of WWER type. In the latter case the Si content is increased up to about 0.6 % simultaneously with the maximum 2 % of Ni and 0.5 % of Mn, respectively. The impurity elements (as Cu, P, S) content should also reach the required level close to the upper concentration limit typical for WWER-1000 RPV materials [16].

4.2. Investigation method and evaluation program

Positron annihilation lifetime spectroscopy (PALS) was chosen for this measurement, because of its sensitivity on vacancy defects. Obtained annihilation spectra were processed by program LifeTime 9 [17]. From LT9 we gain output data like positron lifetime, intensity of positrons or mean lifetime. Positron lifetimes were divided into: \( \tau_1 \) (positrons annihilating in bulk according to standard trapping model), \( \tau_2 \) (positrons annihilating in defects) and \( \tau_3 \) (in-flight annihilation). Similar partition is valid for intensities of positrons (\( I_1, I_2 \) and \( I_3 \)). For the ODS steels investigation, 2-component analysis was chosen. We neglected \( \tau_3 \) and \( I_3 \) (which refers to in-flight annihilation), because their values were very low and brought an error into evaluation.

5. Results

5.1. Model reactor steels

Figure 1 and 2 show results for model reactors steels. The analysis is based mainly on values \( \tau_2 \) and \( I_2 \). In samples Ap631-Dp634, content of Ni was rising and so \( \tau_2 \) rose too (while amount of the rest elements was not changing significantly). From lower \( I_2 \) values results presence of larger clusters of defects.

Samples Ep641-Hp644 have apparently lower values of \( \tau_2 \). It is probably caused by the highest amount of Ni and Mn, which are known for decorating defects from the inside and so they seem to be smaller.

The highest \( I_2 \) values in this measurement reveal us, that defects in these samples are distributed homogeneously.

Remaining samples Kp301- Np304 are slightly different in comparison to previous 2 groups, because of zero V content and slightly increased C content. Characteristic for them is significant drop of Cr content (≤ 0.1). Evaluation was the most complicated due to multiple changes in composition (share of many elements was changing at the same time). Reduction of Cr caused increased \( \tau_2 \) values. By Np304 we expected lower value of \( \tau_2 \) than by Mp303, but it did not happen, probably due to obliteration of marking into the sample.

5.2. ODS steels

Figure 3 represents results for ODS steels. The evaluation is based on \( \tau_1, \tau_2, I_1 \) and \( I_3 \) values. In case of \( \tau_1 \) we cannot talk about annihilation only in bulk, because values are moving around 202 ps. The increase of \( \tau_1 \) is caused by strengthening oxides (Y₂O₃) and partly by 2-component analysis. According to \( I_2 \), contribution of large defects was very low. Variances of intensities were less than 1%.
6. Conclusion
This paper was aimed on investigation of radiation damage to structural materials of nuclear facilities with increased concern over materials for fusion power reactors. The experimental part included 2 measurements – model reactor steels and ODS steels. Results indicate the influence of alloying elements on defects occurrence. Higher content of Cr decreases amount of defects so the steel has better mechanical and thermal properties. By samples with higher amount of Ni and Mn, defects seem to be smaller, but it is probably caused by the ability of Ni and Mn to decorate interior of defects.

Second group of model reactor steels was evaluated as the best according to PALS (based on \( \tau_2 \) comparison). By ODS steels, almost no defects were observed. Positron annihilation spectroscopy is a powerful tool of investigation vacancy type defects. It is necessary to do more measurements to confirm the influence of alloying elements on defects appearance and hence on properties of steels.
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