Non-destructive research methods applied on materials for the new generation of nuclear reactors

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Abstract. The paper is aimed on non-destructive experimental techniques applied on materials for the new generation of nuclear reactors (GEN IV). With the development of these reactors, also materials have to be developed in order to guarantee high standard properties needed for construction. These properties are high temperature resistance, radiation resistance and resistance to other negative effects. Nevertheless the changes in their mechanical properties should be only minimal. Materials, that fulfil these requirements, are analysed in this work. The ferritic-martensitic (FM) steels and ODS steels are studied in details. Microstructural defects, which can occur in structural materials and can be also accumulated during irradiation due to neutron flux or alpha, beta and gamma radiation, were analysed using different spectroscopic methods as positron annihilation spectroscopy and Barkhausen noise, which were applied for measurements of three different FM steels (T91, P91 and E97) as well as one ODS steel (ODS Eurofer).

1. Introduction
GEN IV is an assemble of six theoretical concepts of fission reactors. Comparative advantages include reduced capital cost, enhanced nuclear safety, minimal generation of nuclear waste and reduction of the risk of proliferation. In comparison with present nuclear power plant technology the claimed benefits of the GEN IV reactors are [1]:

- Reduced lifetime of nuclear waste from $10^5$ to $10^2$.
- 100–300 times more energy yield from the same amount of nuclear fuel.
- The ability to use MOX and thorium fuel in a closed fuel cycle.

High temperatures, irradiation stress and consequential change in the mechanical properties of the components of these reactors, urge scientists over the world to examine and develop construction materials that would resist these severe conditions with minimal changes in their mechanical properties. Our research is aimed on FM and ODS steels. These materials are actually foreseen also for the use in the area of thermonuclear fusion in ITER construction [2].

2. Irradiation damage
Fusion neutrons, which originate from the Deuterium-Tritium (D–T) fusion reaction, interact with the walls, blanket and other structures that surround the plasma. Neutron damage weakens the construction materials and induces secondary radioactivity, which will require conscientious salvage procedures or deposition of the used materials as waste. The radioactivity in fusion power plants will
only concern construction materials, because the product of the fusion reaction is only harmless helium. When a neutron collides with an atom of the primary wall or blanket, it can shoot out the atom from its ordinary position in the crystal lattice. The displaced atom can occupy the space between other atoms in their ordinary positions as interstitial and in his former position in the lattice a vacancy appears. This atom may have enough energy to distort the positions of other atoms before he finds his place in the lattice. Therefore the distortion usually occurs in a form of a cascade. The damage is quantified by dpa (displacements per atom) during the durability of the construction. On the primary wall, where the neutron flux is most intensive we expect that the damage state will be in order of hundred dpa [2]. With this kind of high dpa damage the material strength decreases and some parts of the wall will have to be repeatedly replaced during the functioning. The neutron can interact with the atom in the crystal lattice and leave a transmuted atom or atoms. The new atom is generally radioactive and is the primary source of radioactivity. The neutron can also shoot out a proton or alpha particle from the stricken atom. These reactions can be inscribed as (n,p) and (n,α) reactions. The reactions (n,α) are essential to neutrons with energies of 14 MeV that occur from the D–T fusion reaction. Protons and alpha particles from these reactions capture electrons and produce hydrogen and helium atoms in the crystal lattice. Individual atoms then form into clusters and produce bubbles of gas that decrease the strength of the construction. The process of irradiation damage is sufficiently studied in fission reactors and their understanding provides a solid base for studying and evaluating similar problems in the future fusion power plants.

3. Reduced activation materials

One of the most important advantages of fusion in comparison with fission is the absence of highly radioactive waste in the fusion fuel cycle. The construction will be irradiated due to neutrons and be therefore radioactive, but with a cautious selection of construction materials we can provide low activation so the radioactivity of a fusion power plant may decrease on a very low level in a period of less than 100 years. The blanket, primary wall and the system of diverters (in case of magnetic confinement) will have a definite durability and will have to be maintained with the help of robots. Important source of activation are also small impurities in the construction materials, so the testing of quality and purity of materials will be essential in the effort to achieve low activation. One of the construction materials that may be considered as low activated (LA) are known as FM steels, which are frequently used where we can encounter high temperatures for example in fission reactors. Iron and chromium, the main components of FM steels, have relatively good activation properties. The main source of activation comes from minor impurities, for example niobium. Therefore LA materials commute niobium for vanadium or titan or we can commute molybdenum for wolfram [2].

4. FM steels

FM steels consist of a feritic matrix and particles of martensit. The 9–12% Cr FM steels are being recently considered as main candidate materials for the first wall components of future fusion reactors. These materials can be used in components that have to resist high temperatures up to 550 °C. After exceeding 18% of chromium the material inclines to fracturing. Recent developments are aimed on the chemical composition to achieve low activation, fast decay properties after irradiation (RAFM steels) and also on the reduction of the shift of the ductile-brittle transition temperature after irradiation.

4.1. T91

T91 presents valuable advantages related to their excellent elevated temperature strength, creep behavior and improved resistance to thermal fatigue, and good transfer and low expansion coefficient compared to austenitic steels. This material has been identified as the ultimate candidate for DEMO in the frame of the international fusion program. Despite very limited data within MYRRHA (Multipurpose hybrid research reactor for high-tech applications) operating conditions on neither the effect of irradiation, liquid metal and their synergy, T91 is the prime candidate material to be in contact with the LBE (Lead-Bismuth-Eutectic) and to contain the fuel in MYRRHA. This material seems to be the
The best compromise between fabricability and resistance to severe irradiation conditions in the operating temperature range from 200 to 450 °C [3].

### Table 1. Chemical composition of material T91 [3]

| element | C   | Si  | Mn  | P  | S  | Cr  | Ni  | Nb  | N   | V   | Mo  |
|---------|-----|-----|-----|----|----|-----|-----|-----|-----|-----|-----|
| intensity (%) | 0.11 | 0.4 | 0.4 | -  | -  | 8.3 | 0.13| 0.08| 0.02| 0.2 | 0.95|

4.2. P91

P91, is widely used in the construction of power plants and other sectors involving temperatures higher than 500 °C. The heat resistant martensitic steel P91 is being utilized in conventional power plants for piping systems with operating temperatures of 560 – 600°C and high pressures in the range of 270 bars. The design of welded structures requires resistance to high temperatures and pressures for up to 30 years. Therefore, the selection of the correct welding consumables is of great importance to prevent premature equipment failure. Although the creep strength is the prime consideration for elevated temperature applications, notch toughness is also important. P91 steel weld fusion zone toughness depends on factors such as welding process, chemical composition, and flux composition. Niobium and vanadium are the main alloying elements that significantly influence the toughness as well as creep strength.

### Table 2. Chemical composition of material P91 [4]

| element | C   | Si  | Mn  | P  | S  | Cr  | Ni  | Nb  | N   | V   | Mo  | Al  |
|---------|-----|-----|-----|----|----|-----|-----|-----|-----|-----|-----|-----|
| intensity (%) | 0.1 | 0.45| 0.5 | 0.019| 0.002| 9.12| 0.05| 0.06| 0.04| 0.21| 0.96| 0.004|

4.3. Eurofer97 (E97)

E97 has been developed as a possible first wall and breeder blanket structural material for fusion applications. E97 is a representative of RAFM steels, manufactured on an industrial scale. Further reduction of impurities and therefore achieving lower activation is possible. The properties of E97 meet the requirements of the test blanket module of ITER. The mechanical and compatibility properties are adequate, also in the neutron flux exposure environment expected in ITER. The potential for applying E97 in the blanket for a demonstration fusion power plant is there, but experimental verification that simulates the near plasma environment remains essential. E97 has a tempered martensitic microstructure which permits operation at relatively high temperatures (500 °C), offers good dimensional stability under high neutron doses and exhibits higher swelling resistance in comparison to austenitic steels. Future developments are aimed on obtaining a fine microstructure with small prior austenite grains and a low number of inclusions which should provide an optimum combination of toughness, strength and high temperature properties.

### Table 3. Chemical composition of material E97 [6]

| element | Fe  | W   | Mn  | P   | S   | Ta  | C   | V   | N2  | Cr  |
|---------|-----|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| intensity (%) | 88.805| 1.1 | 0.4 | <0.005| <0.005| 0.12| 0.11| 0.2 | 0.03| 9.0 |

5. ODS Eurofer

Oxide dispersion strengthened (ODS) steels are considered as leading candidates of structural materials for advanced nuclear systems such as GEN IV fission power plants and DEMO fusion reactor because of their excellent creep strength [7], corrosion [8] and radiation resistance [9]. To apply ODS steel in these advanced nuclear systems with huge and complex structures, suitable bonding and welding techniques need to be developed. These techniques must provide such a process that the microstructures with very fine grain and homogeneous distribution of nano-scaled oxide
particles are not remarkably changed by the joining processing [10]. Whereas the temperature window of FM steels is limited to 550 – 650 °C by degradation of mechanical strength, ODS steels have the potential to increase this limit considerably up to 1000 °C. The main problem in the development of ODS steels is the inefficient production of material by the means of powder metallurgy and thermal isostatic pressing. ODS Eurofer is strengthened by particles of Y₂O₃. E97 mainly differs from ODS Eurofer in properties that relate with the formation of defects. One of the first applications of ODS Eurofer might be in the dual coolant lead lithium (DCLL) BB concept. ODS Eurofer layers will be plated onto the first wall made of E97 steel. The feasibility of joining ODS Eurofer and E97 by diffusion welding was recently demonstrated [11].

6. Positron annihilation lifetime spectroscopy (PALS)
Positron (e⁺) is the antiparticle of electron (e⁻). The principal of PALS is the ability of a positron emitted from the source due to β⁺ decay thermalize and diffuse into the material, until it gets trapped by a defect and encounters an electron with which it annihilates due to electromagnetic interactions. The annihilation results in two annihilation photons γ with energies of 511 keV. When the energetic positrons (Eₖin ≈ 0.1 – 1 MeV) enter the condensed medium, they rapidly lose their energy. At highest positron energies, the positron excites core electrons when it collides with the host atoms, so the main mechanism of energy losses is ionization. At lower energies the electron-hole excitations is more present. When the positron energy has declined to a fraction of eV, the scattering off phonons dominates. Nevertheless positrons reach thermal equilibrium with the medium, due to phonon emission and absorption. The initial kinetic energy of positrons during thermalization drops below 0.1 eV. Despite the rapid loss of energy the positron thermalization time tₖ is only a few picoseconds [12].

During diffusion, the positron eventually annihilates with an environmental electron. In homogeneous defect-free media all positrons annihilate with the same rate λb which is a characteristic of the given material. In a medium with defects the positron may become trapped in open-volume defects (monovacancies, larger vacancy clusters, dislocations etc.) because they represent a potential well for positrons. The transition from the delocalized state to the localized one is called positron trapping. Thermal detrapping is practically impossible from such deep wells and positrons remain trapped until annihilation. There may occur phonon-assisted detrapping if a positron trap is shallow enough (E_b < 0.1 eV). The lifetime of the trapped positrons (LT2) is longer than the lifetime of positrons which annihilate within the material (LT1) due to the lowered density of electrons near the defect. Positron can form a quasi-stable state with a host electron called positronium (Ps) which is an analogue of the hydrogen atom [12]. The time interval between the injection of positrons into a sample and the instant of annihilation is known as the positron lifetime spectrum and has a statistical distribution. In homogeneous defect-free materials, the lifetime spectrum is a simple decaying exponential. When the material contains defects, the lifetime spectrum becomes a superimposition of decaying exponentials. The principal of PALS is based on injecting positrons into a sample and measuring their lifetime spectrum. Our positron source is a β⁺ radioactive isotope ²²Na (figure 1).

![Figure 1. ²²Na decay scheme](image1.png)

![Figure 2. Basic scheme of PALS equipment setup in PAS laboratory at STU Bratislava](image2.png)
The advantage of this isotope is the emission of a photon with the energy of 1.274 MeV in coincidence with the positron which gives the signal that indicates the birth of the positron (START signal). The signal of annihilation is either one of the two annihilation photons with energies of 0.511 MeV (STOP signal). Typical measuring equipment contains two detectors (scintillator + photomultiplier), two energy analyzers (SCA), time to amplitude converter (TAC) and a multichannel analyzer (MCA) that records particular events. Simple scheme of this setup is shown in figure 2.

7. Barkhausen noise
Ferromagnetic materials consist of domains. These domain are magnetized along a certain crystallographic direction. Domains are separated from one another by domain walls. AC magnetic fields cause domain walls to move back and forth. In order for a domain wall to move, the adjacent domain of the wall has to increase in size while the domain on the opposite side of the wall shrinks. As a result the overall magnetization of the sample changes. If we place a coil of conducting wire near the sample while the domain wall moves, the resulting change in magnetization will induce an electrical pulse in the coil. Barkhausen effect proves that the magnetization process, which is represented by the hysteresis curve, is not continuous, but is made up of small, abrupt steps caused when the magnetic domains move under an applied magnetic field. When the electrical pulses produced by all domain movements are summed, a noise-like signal called Barkhausen noise is generated. Due to magnetoelastic interaction, in materials with positive magnetic anisotropy, compressive stresses will decrease the intensity of Barkhausen noise while tensile stresses increase it. This fact can be exploited so that by measuring the intensity of Barkhausen noise the amount of residual stress can be determined. The measurement also describes the direction of principal stresses. The micro-structure of the sample is another important material characteristic affecting Barkhausen noise. The noise intensity continuously decreases in microstructures characterized by increasing hardness. Barkhausen noise measurements provide information on the microstructural condition of the material.

8. Measurements
The source of positrons contains particles that annihilate positrons, therefore we have to calibrate the source before every use to estimate the amount of positrons annihilating in the source. The calibration is performed by measuring the spectrum of an almost defect-free reference material, for which we know the characteristic lifetimes of the positrons, in our case pure silicon. After the measurement of silicon we measure the spectrum of the investigated material and consecutively compare the lifetimes of positrons with theoretical values of known defect types.

Meanings of components in table 4: LT1 – lifetime of positrons in bulk, LT2 – lifetime of positrons in defects, I1 – Intensity of LT1, I2 – Intensity of LT2, MLT – mean lifetime of positrons, FV – Fit’s variance.

From figure 3 and table 4 we can see, that every material was described by two LT’s, which means that all materials contain defects in their microstructure. From theoretical values we can deduce that materials T91 and P91 contain divacancies and E97 monovacancies with dislocations.

Figure 5 represents the intensities of defects in particular samples. The intensities are as follows: in the material E97 74%, in P91 43% and in T91 52%. The next non-destructive method we used to study materials was the method of Barkhausen noise. The output noise, produced by the rotating domain walls, can be described by an envelope. The bigger the envelope the less amount of residual stress the material exhibits, because the domains move more freely due to lack of defects. The measurement confirms the accuracy of the previous results. The material P91 exhibits minimal residual stress and thus contains least defects. After the material P91 follow materials T91, E97 and ODS Eurofer. Due to the lack of time, material ODS Eurofer was not measured by PALS so we cannot confirm the accuracy of results for this material.
Table 4. Measurements of materials T91, P91 and E97

| Sample  | T91     | P91     | E97     |
|---------|---------|---------|---------|
| LT1 (ps)| 110 ± 7 | 117 ± 7 | 83 ± 15 |
| I1 (%)  | 48 ± 5  | 57.2 ± 1| 24.4 ± 5|
| LT2 (ps)| 199 ± 9 | 207 ± 2 | 178 ± 8 |
| I2 (%)  | 51.5 ± 5| 42.5 ± 1| 73.8 ± 5|
| FV      | 1,07    | 1,02    | 1,01    |
| MLT (ps)| 160     | 159     | 161     |

9. Summary

Materials P91, T91 and E97 were studied by PALS and the method of Barkhausen noise. The material P91 showed the most resistance to the formation of defects. Unfortunately, this material can resist temperatures only in the range of 560-600 °C, whereas ODS steels can resist temperatures exceeding 1000 °C. All the materials are prime candidates for components for the GEN IV, ITER, DEMO and other applications. Nevertheless, we have to keep improving and developing construction materials, so we can provide efficient contribution to the safe design of advanced nuclear facilities.
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