THEORETICAL AND EXPERIMENTAL CHARACTERISATION OF ODS STEELS

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Received 22 April 2014; accepted 10 May 2014

1. Introduction

Oxide dispersion strengthened (ODS) steels are considered as leading candidates of structural materials for advanced nuclear systems, including 4th generation of nuclear reactors (GEN IV) and DEMO fusion reactor due to their excellent creep strength, corrosion [1] and radiation resistance [2]. The foremost consideration in the successful development and deployment of GEN IV reactor systems is the performance and reliability issues involving structural materials for both in-core and out-of-core applications. The structural materials need to endure much higher temperatures, higher neutron doses and extremely corrosive environment, which are beyond the experience of current nuclear power plants [3]. ODS steels are considered as primary structural materials for Gas-cooled Fast Reactor (GFR) and Sodium-cooled Fast Reactor (SFR). ODS steels are suitable as cladding materials mainly in SFR systems with MOX fuel and Supercritical Water-cooled Reactor (thermal/fast).

Application of ODS steels in these advanced nuclear systems with huge and complex structures as well as suitable bonding and welding techniques need further development. These techniques must provide such a process that the microstructures with very fine grains and homogeneous distribution of nano-scaled oxide particles are not remarkably changed by the joining processing [4]. Investigating the microstructure of steels and the effect of radiation on their properties is of interest to ensure high nuclear safety of power plants and nuclear facilities.

2. Experimental details

In this paper we examine four different ODS steels received from the Korean Atomic Energy Research Institute (KAERI). These samples differ only in Tantalum and Boron content as can be seen in Tab. 1. Samples were measured using Positron Annihilation Lifetime Spectroscopy (PALS) before irradiation. PALS has a long and successful history of measuring defects in materials. Conventional methods exploit positron sources, such as ^{22}Na, or positron beams where accelerators and beam optics are used to deliver a beam of positrons to a sample. The fraction of transmitted positrons through a uniform material can by calculated using Eq. (1).

$$P(z) = \exp(-\alpha z) \tag{1}$$

Where α is the absorption coefficient and z is distance. Using Mourino et al. [5] empirical expression of the absorption coefficient in Eq.(2) and approximate density for ODS steels about 7g/cm^3 we assess that 98% of all positrons annihilate in 0.2mm of our steels.

$$\alpha = 26.8Z^{0.15}\rho \tag{2}$$

The effective atomic number Z of the sample was obtained using Eq.(3) where f_n is the fraction of the total number of electrons associated with each element, and Z_n is the atomic number of each element.

$$Z = \sqrt[2.94]{f_{Cr}.(Z_{Cr})^{2.94} + f_W.(Z_W)^{2.94} + f_V.(Z_V)^{2.94} + f_{Ti}.(Z_{Ti})^{2.94} + ... + f_{Fe}.(Z_{Fe})^{2.94}} \tag{3}$$

Tab.1. *Chemical composition of KAERI samples. All samples contain 0.3 wt% strengthening particles of yttria.*

Wt.%	**Cr**	**W**	**V**	**Ti**	**Ta**	**Y2O3**	**B**
A0102	12	1.1	0.2	0.3	0.14	0.3	-
B0102	12	1.1	0.2	0.3	0.14	0.3	0.002
C0103	12	1.1	0.2	0.3	-	0.3	-
D0102	12	1.1	0.2	0.3	-	0.3	0.002

Positron lifetime measurements were carried out using ^{22}Na, with activity about 20μCi, deposited on a kapton foil. The source was sandwiched between two identical samples of steel. The count rate was approximately 40 counts per second.

Positron lifetime data were evaluated using PATFIT-88 software. A detailed description of the resolution function required for spectrum analysis used three Gaussian functions with intensities 80%, 10%, 10%, and appropriate relative shifts. All spectra contained at least 1.3x10^6 counts. The lifetime spectrum is analyzed as a sum of exponential decay components, $n(t)=\Sigma_i I_i \exp(-t/LT_i)$, convoluted with the Gaussians functions describing the spectrometer timing resolution using POSITRONFIT. Decay components due to annihilation in NaCl (≈430ps) and kapton foil (≈382ps) were subtracted in the procedure. The value of positron lifetime in bulk material LT_b can be obtained from Eq.(4) and Eq.(5) serves to calculate the average lifetime LT_{avg}.

$$LT_b = \left(\frac{I_1}{LT_1} + \frac{I_2}{LT_2} \right)^{-1} \tag{4}$$

$$LT_{avg} = \Sigma_i I_i . LT_i \tag{5}$$

The spectra show that KAERI samples are not defect free (Tab.2.). If the average lifetime LT_{avg} is greater than the bulk lattice lifetime characteristics of the material LT_b then it indicates that vacancy-type defects are present [6]. The analysis reveals that defects with a positron lifetime from 233-241ps exist in all samples. The lifetime is equivalent to that of vacancy clusters with an average size of about 3-4 defects per cluster. Also the intensity of this component was found to be high (60-70%). The rate of positron trapping to a vacancy, κ_d, is proportional to the concentration of these defects C_d, where the constant of proportionality is the defect specific trapping coefficient μ_d. The two-state defect trapping model gives Eq.(6) [7]. The calculated positron trapping rates and concentration of defects using Eq.(6) are listed below in Tab.3.

$$\kappa_d = \mu_d . C_d = \frac{LT_{avg} - LT_b}{LT_b\left(LT_2 - LT_{avg}\right)} \qquad (6)$$

Tab.2. *Obtained PATFIT-88 results for KAERI steels, where LT_1-reduced positron lifetime in bulk material, I_1- intensity of positrons with LT_1, LT_2- lifetime of positrons in defects, I_2- intensity of positrons with LT_2, ΔLT_1, ΔLT_2, ΔI_1, ΔI_2- deviations in lifetimes and intensities, LT_b- positron lifetime in bulk using Eq.(4), LT_{avg}-average lifetime from Eq.(5).*

PATFIT	**LT_1**	**ΔLT_1**	**I_1**	**ΔI_1**	**LT_2**	**ΔLT_2**	**I_2**	**ΔI_2**	**LT_b**	**LT_{avg}**
KAERI-A	66.7	13.7	30.7	1.89	233.8	5.8	69.4	1.89	132.3	182.6
KAERI-B	56	9.6	32	1.22	237	4.8	68	1.22	116.4	179
KAERI-C	58	6	39	0.9	241.2	3.4	60.7	0.9	107.6	169
KAERI-D	84.5	9.6	32	3.6	233.2	6.8	67.7	3.6	148.6	185

Tab.3. *Calculated positron trapping rates κ_d and defect concentration C_d for KAERI samples using Eq.(6).*

	κ_d [s^{-1}]	**C_d [ppm]**
KAERI-A	7.43645 E+9	2.25
KAERI-B	9.27366 E+9	2.81
KAERI-C	7.94893 E+9	2.41
KAERI-D	5.10875 E+9	1.55

The specific trapping coefficient for a single vacancy in pure Fe is μ_d=1.1x$10^{15}$$s^{-1}$, and analogously for a cluster of 3-vacancies it is μ_{3v}=3*1.1x10^{15} [8].

The effect of yttria particles on positron lifetime can be proven by comparing LT_{avg} values of conventional ferritic/martensitic steels (LT_{avg} ≈130ps) without additions of yttria oxides and ODS steels (LT_{avg} ≈170-180ps). The relatively low content of yttria oxides plays a role due to significant positron affinity of yttrium (-5.31) [9]. The experimentally reported positron lifetime in Y_2O_3 bulk is 239ps [10]. Our measured lifetimes in defects are in the range of 233-241ps which corresponds well to positron lifetime of Y_2O_3 bulk. It is plausible to assume that yttria oxides serve as trapping sites for positrons due to their high positron affinity and positrons annihilate in their vicinity rather than in bulk material of the steel [11]. This is supported also by PALS measurements which show high intensities of positrons annihilating in defects. Yttria particles distort the environment of the lattice, creating defects to which positrons are attracted by yttria affinity and positrons annihilate at these sites.

We assume also different type of defects in the material. However, the lifetime of the next component must be at least ≈40% longer/shorter than the previous one to allow them to be separated. Therefore, the value of the shorter component would have to be smaller than 140-145ps for us to be able to detect. These values are however close to lifetime of positrons in Fe (107ps) and are therefore merged into one positron lifetime. The calculated lifetime of bulk LT_b indicates the following: samples A, B and D (LT_{avg} =132.3, 116.4 and 148.6ps) probably exhibit another defect type, which could not be separated due to similar lifetime of bulk or does not satisfy the ±40% rule. Dislocations with theoretical lifetimes of 160-180ps could raise the bulk values and could not be detected. It is also rational to assume dislocation existence in these materials because dislocations are responsible for ductility in steels which is a desired attribute.

Sample C is the only sample with LT_b of 107.6ps which is in excellent agreement with lifetime of Fe matrix.

Samples are currently in the Washington State University reactor (WSUR). WSUR is a TRIGA type reactor and is the only reactor with mixed 8.5/20(Standard TRIGA) and 30/20 Low Enriched Uranium core. Samples are being irradiated by max fast flux of approximately 4.10^{12} $n.cm^{-2}.s^{-1}$. The desired radiation damage is below 1 dpa, to prevent saturation trapping.

After receiving samples from the reactor, we will apply PALS to observe changes in irradiated materials with the aim to study their response to radiation. We predict higher positron trapping rate due to higher concentration of defects caused by displacements in the lattice. Also positron lifetime of the defect component should rise. The anticipated changes would lead to hardening of steels and degradation of ductility.

3. Discussions

The trapping rate of positrons into vacancy clusters and concentration of these defects in KAERI ODS steels has been calculated. Positron lifetime measurements prior to planned irradiation by neutrons showed clusters of 3-4 vacancies to be the main defect throughout the samples. High intensity of this component together with the nature of yttria particles to attract positrons shows a high amount of positrons annihilating near yttria particles.

After irradiation we predict changes in microstructure. Aim of our future work is to determine the resistance of ODS steels towards neutron irradiation and prove that radiation induced hardening of these steels is shifted towards higher neutron doses in comparison to traditional ferritic/martensitic steels which are not strengthened by yttria particles.

Acknowledgement

Financial support from the National Science Foundation (DMR 1359523 grant) as well as from Slovak scientific agencies VEGA 1/0204/2013 and APVV AK48 are highly acknowledged.

4. References

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