Study of Neutron Induced Defects in Ceramics using the GiPS Facility

S May-Tal Beck,[1] M Butterling[2], W Anwand[2], A Beck[1], A Wagner[2], G Brauer[2] , A Ocherashvili[1], I Israelashvili[1], O Hen[3]

[1] *Physics department, NRCN,P.O.Box 9001, Beer-Sheva, Israel*
[2] *Institut für Strahlenphysik, Helmholtz-Zentrum Dresden-Rossendorf, 01314 Dresden, Germany*
[3]*School of Physics and Astronomy, Tel-Aviv University, Tel-Aviv 69978, Israel*

smtbeck@gmail.com

Abstract. Preliminary results are presented from a study of neutron irradiation damage in Sapphire and B_4C, produced with a fluence of 6×10^{18} n/cm^2 and ~10^{15} n/cm^2, respectively. Measurements were performed at the GiPS facility and the SPONSOR beam at HZDR, and in the PAL spectrometer at NRCN. Bulk and vacancies lifetimes were identified in the Sapphire, ~150ps and ~188ps, respectively, with complete trapping in the irradiation induced vacancies. Irradiation damage in B_4C found to be limited to the surface. A single lifetime of ~166ps was measured in both irradiated and non-irradiated samples, and was associated with the bulk.

1. Introduction

There has been an increased interest in defects within structural materials motivated by future fission and fusion reactor needs. While reactor steels are extensively studied, much more research effort is needed in order to understand radiation damage in ceramic materials and its effect on their macroscopic characteristics [1].

Sapphire and B_4C are interesting to study. Sapphire is a candidate material to be used in diagnostic systems for burning plasma experiments [2], due to its transparency to a wide range of wavelengths (200-5000 nm), high melting temperature (~2300 K) and hardness close to that of a diamond. Its optical and electronic properties are expected to be affected by the harsh radiation environment. The B_4C is interesting for the nuclear industry due to the high cross-section for thermal neutron capture in Boron, which produces helium inside the material. The much higher neutron flux expected in future reactors can cause swelling of the material and macroscopic crack formation.

PAS methods are suited [3-5] and applied to study neutron-induced defects in Sapphire and B_4C samples. In the present paper first but preliminary results are presented.

2. Samples and Measuring Techniques

Two pairs of Sapphire samples were available as disks of 14 mm in diameter and a total thickness of 6 mm. One pair was neutron irradiated in the Israel Research Reactor 1 (IRR1) [6] some few years ago, to a fluence of 6×10^{18} n/cm^2. The other was not irradiated and was annealed at 1773 K for 30 minutes before measurements. Two pairs of cubic samples of B_4C were available, each having a size of $10\times10\times8$ mm^3. One pair had been previously irradiated to a neutron fluence of ~10^{15} n/cm^2 in the IRR1 [6]. Despite of the high penetrability of neutrons, predictive calculations for B_4C [7] show that damage can be produced only on the surface due to the high cross section for the $^{10}B(n,\alpha)^7Li$ process, of ~3800 barn for thermal neutrons. Based on the calculations of Ref.7, and taking into account the neutron fluence of ~10^{15} n/cm^2 on the B_4C samples, a damage of ~10^{-4} -10^{-3} dpa is expected to exist on their surface. A single crystalline Si sample was used as a reference.

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All Sapphire and B_4C samples were measured at the GiPS facility [3]. B_4C samples were measured also at the SPONSOR beam [4] since neutrons were expected to create damage only on their surface [7]. Lifetime measurements on the same Sapphire samples were performed also in the PAL spectrometer at NRCN [5]. The GiPS facility is suitable for thick samples (>= 1cm³) of solids and liquids. At the SPONSOR beam mono-energetic positrons from 0.027 keV to 37 keV are available. The PAL spectrometer at NRCN had an average time resolution ~190ps during measurements.

2.1. GiPS

The data from GiPS measurements are collected in list mode, namely, for each event the data from each of the detectors is recorded, making possible an offline correlative data analysis. Selection of good annihilation events was done by requiring that only one pair of (BaF_2, HPGe) detectors fired, and by applying energy cuts on the spectra from these detectors. The first criterion selected ~0.25 of the collected events, and energy cuts further reduced the number of good events by a factor of ~6, leaving ~1.5×10^5 events from each measurement for final analysis. The energy scales were not calibrated, but due to the fact that the 511 keV photons were the only ones detected, the selection criteria were defined in channel units. These criteria included events within one and five FWHM from the center of a fitted Gaussian to the 511 keV photo-peaks in the BaF_2 and HPGe energy spectra, respectively. As an example, figure1 shows one HPGe energy spectrum of events that passed all selection criteria from the non-irradiated B_4C measurement. Lifetime data, gained from the BaF_2 time stamps and selected by the same set of criteria, are shown in figure 2 for the four BaF_2 detectors.

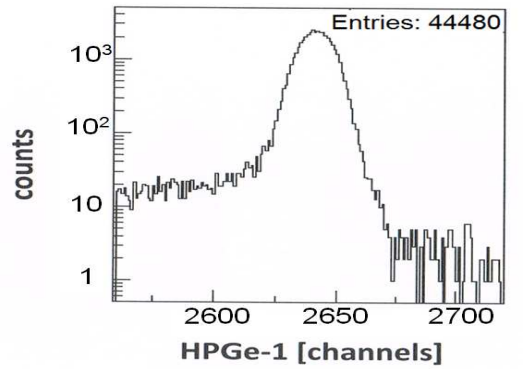

Figure 1. Energy spectrum from one HPGe detector, for the selected events of the non-irradiated B_4C measurement.

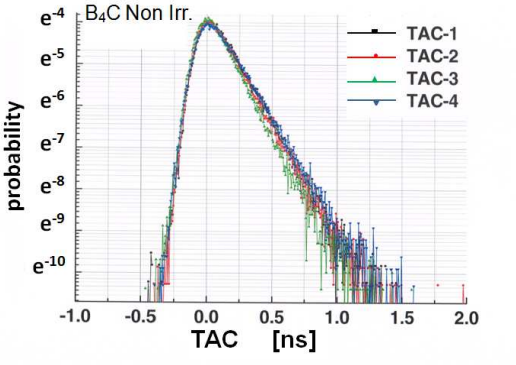

Figure 2. Positron lifetime spectra from the four BaF_2 detectors, for the selected events of the non-irradiated B_4C measurement.

Mean positron lifetimes were fitted using the RESOLUTION program [8]. The time resolution for each BaF_2 detector was defined from the Si measurement, ~180ps. For all spectra it was possible to fit only one decaying exponential. Figure 3 presents the averaged mean values of positron lifetimes extracted from the four different detectors per each measurement. The error bars include the statistical error, as well as the spread of results from the different detectors. DB analysis is still pre-mature and is not presented here.

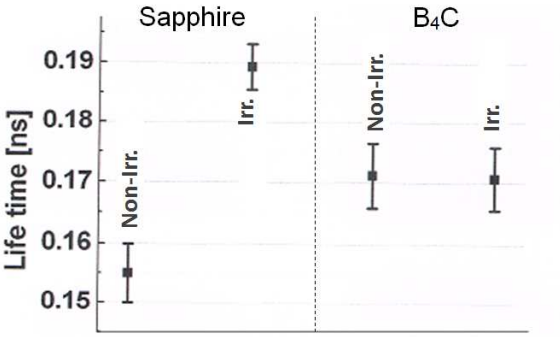

Figure 3. Averaged mean positron lifetimes for the four measured samples (see text for further details).

2.2. SPONSOR

DB spectra for B_4C samples, irradiated and non-irradiated, were collected with one high-purity Germanium (HPGe) detector in varied steps of the positron energy. The energy resolution of the HPGe

detector was (1.09±0.01) keV at 511 keV. The number of counts accumulated in the photo-peak was 9.5×10^5 per positron penetration energy. Energy regions defined for S and W calculations were |Eγ-511keV|<0.92keV and 2.3keV<|Eγ-511keV|<3.0keV, respectively. Figure 4a shows the S values as a function of positron penetration energies, for the two measurements. Slight differences between the irradiated and non-irradiated samples appear on the surface. Figure 4b shows the S-W plot, where this difference is more pronounced. Positron diffusion lengths, calculated using the VEPFIT program [9], were found to be identical: L_+ = (96 ± 4) nm and L_+ = (97 ± 6) nm for the irradiated and non-irradiated samples, respectively. This is expected since the irradiation damage is concentrated on the surface of the sample and therefore has little effect on the average diffusion length.

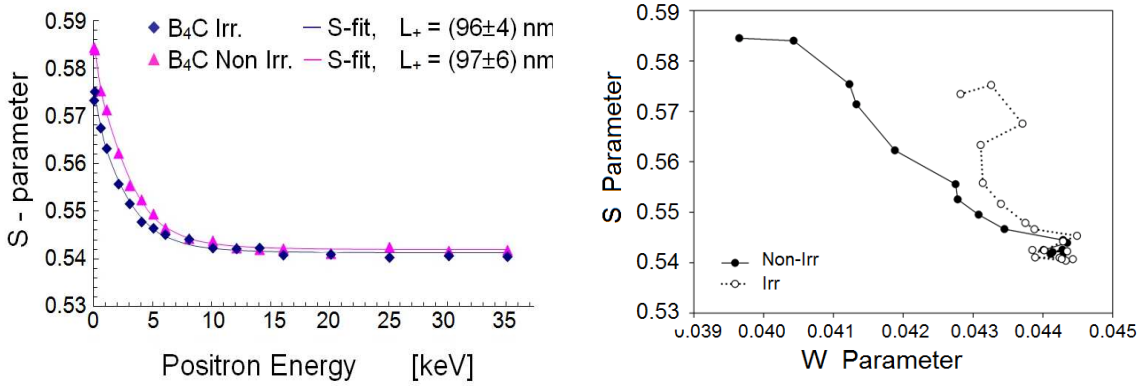

Figure 4: (a) S values extracted from two B_4C samples, non-irradiated and irradiated to a fluence of ~10^{15} n/cm^2, as a function of positron beam energy. The marker size includes the uncertainties, dS=0.0006. **(b)** S-W plot for the values extracted for these two samples.

2.3. PAL spectrometer

The source and samples assembly used included two 1μm Ni foils, as beddings for the ^{22}Na source, and an air gap between the foils and the samples for safety reasons. The Ni foils and the samples were held by Perspex housings.

Positron lifetimes in Perspex and their relative intensities were found from a Perspex measurement performed with a similar assembly (7.6 μm Kapton foils instead of Ni), and based on the simulated relative annihilation intensities in the assembly materials (done in a similar way to that described in detail in [10]). The lifetimes: 125ps (p-Ps), ~360ps and ~1770ps (o-Ps) contribute relative intensities of ~0.3 and ~1.96 of that of the long component. These values are similar to other published values for nylon materials [11]. A measurement of well annealed Fe samples was used to extract the τ_{e+}(Ni)~235ps. The measurement of the Sapphire non-irradiated and annealed samples, were used to extract the relative intensity in Ni, ~8.5%, as well as the relative intensities in the Perspex components. Time resolution was defined from ^{60}Co measurements [5], taken in between Sapphire measurements, ~190ps. Finally, for each Sapphire measurement, all source correction components were fixed, except for the intensity of the o-Ps component that might include annihilations in the air gap. The POSITRONFIT package [8] was used to analyse the lifetime spectra. The resulted mean lifetimes are 144±4ps, 159±1ps and 188±2ps for the non-irradiated and well-annealed, non-irradiated and irradiated to 6×10^{18} n/cm^2 samples, respectively. The errors are statistical only.

3. Results and Discussion

The measurements presented here, of ceramic samples that were exposed to low dose of neutron irradiation, can lead to few conclusions:

The lack of background events in lifetime measurements in the GiPS facility is evident by the single decay obtained for each of the samples. The changes between the results obtained from the

different detectors, and taken into account in the error estimation (figure 3), reflect the systematic uncertainty of the measurement.

For the Sapphire, two distinguished lifetime were measured for the non-irradiated and annealed, 155±5ps, and for the irradiated samples, 189±4ps, in similarity to previously reported values, and thus can be attributed to the bulk region and to trapping in vacancies [12]. The criteria applied to the data selected a clean sample of events, with relatively low statistics. More events can be selected in further analysis of the same data, containing more than one pair of detectors, but adequate background reduction criteria should be studied and applied. Positron lifetimes resulted from measurements of the same samples at the NRCN spectrometer (listed above), agree within 1σ with the results of GiPS. While in [13] two lifetimes are reported for Sapphire irradiated to ~10^{20}-10^{21} n/cm^2, it is evident from the present measurement that for the fluence of 6×10^{18} n/cm^2, only one type of defect is visible, which fully traps the positrons.

Lifetime results from the B_4C measurements do not show any difference between the irradiated and non-irradiated samples, and agree with published results for the non-irradiated material [14]. However, since the GiPS facility probes the entire volume of the sample, if damage has been created in few micron depth of the material, a maximum intensity of 2% is expected. This amount cannot be observed with the current low statistics. The DB measurement in the SPONSOR beam is much more sensitive to the surface of materials, and shows slight change on the surface between the two B_4C samples, which is not apparent by the VEPFIT results. Since the 10^{-3} dpa in a depth of few microns should be visible in slow positron beam measurements, further investigation is needed to understand the measurement results in relation with the damage estimation. The two distinguished regions seen on the S-W plot, resulted mainly from different W values on the surface, might indicate on the existence of He bubbles there. Further measurements with correlated DB data should enhance the sensitivity to the W parameter and produce a better picture of the neutron irradiation induced defects.

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