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Ion/Electron Induced Luminescence for Radiation Damage Process Interpretation and *In Situ* Material Verification

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Abstract

The Research Centre for Energy, Environment and Technology (CIEMAT), Madrid, Spain, features two installations (2 MeV Van de Graaff electron accelerator and a 60 kV ion implanter) specifically developed for *in situ* material characterization during irradiation, and focused on the study of volume and surface electrical degradation in insulating materials for fusion applications. These installations have been equipped with optical systems which permit electron and ion beam induced luminescence to be measured at the same time as recording the electrical conductivity. Recent results for combined ion-induced luminescence and surface electrical degradation experiments in alumina confirmed a correlation between conductivity changes and evolution of emission bands with irradiation dose. Similar experiments under electron irradiation in silicon carbide have also shown the usefulness of luminescence for material characterization and evaluation of radiation effects.

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Keywords: electron accelerator, ion implanter, luminescence, radiation damage

Introduction

One of the main research activities within the Fusion Materials group at CIEMAT (Research Centre for Energy, Environment and Technology), Spanish public research body located in Madrid, is currently the investigation of radiation effects on electrical properties of insulating materials. Numerous systems in fusion devices, including

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plasma heating, diagnostics and remote handling, require the use of insulating elements. Depending on the location, these materials will be subjected to different levels of neutron, gamma, and charged particle fluxes, and will be required to maintain for as long as possible not only their mechanical properties but also the very sensitive physical properties such as electrical conductivity. Expected radiation damage such as Radiation Induced Conductivity (RIC), Radiation Induced Electrical Degradation (RIED), and surface electrical conductivity degradation must be examined for the candidate materials [1-3].

Although simulation of radiation damage through theoretical calculations and models provides valuable information in the prediction of materials behaviour during irradiation at experimentally unattainable dose scales, testing of materials under real radiation conditions is essential in order to establish their operation capabilities. The use of charged particle accelerators for radiation damage investigation is widespread, as a versatile means of reproducing radiation effects under properly controlled conditions of temperature, pressure, and dose rate.

The CIEMAT features two installations specifically developed for *in situ* material characterization during irradiation, where experimental setups are mainly focused on the study of volume and surface electrical degradation: The 2 MeV Van de Graaff electron accelerator, which permits the study of ionizing effects and displacement in the volume, and a 60 kV ion implanter to examine surface effects. The major goal of these facilities is the possibility of in situ and real-time measurements as a function or irradiation dose under different conditions.

Among other effects, the transfer of energy from the incident particle to atomic or molecular electrons in the insulator material may give rise to the emission of light, defined in this text according to excitation source as radioluminescence (RL) for high energy electrons, and ion beam induced luminescence (IBIL) for ions. Consequence of electronic transitions between characteristic energy levels, it provides information about the state of solid lattice structure and defects. Radiation induced material modification including point and extended defects creation, impurity valence changes, or segregation which affect physical properties may cause identifiable changes in luminescence bands, enabling direct and continuous material performance verification during irradiation.

The accelerator and implanter installations have been equipped with optical systems which permit electron and ion beam induced luminescence to be measured at the same time as recording the electrical conductivity. This allows one to observe a correlation between the two processes, considered to be the first step to permit *in situ* monitoring of radiation induced degradation in materials by means of luminescence.

Examples for experiments made in silicon carbide and aluminium oxide which illustrate luminescence potential for this purpose as well as for the characterization and study of ceramic materials for fusion applications, are presented.

1. Installations

1.1. The 2 MV Van de Graaff accelerator (VDG)

The Van de Graaff high voltage generator was acquired in 1953 and was first dedicated to fundamental nuclear physics research activities involving proton, deuterium and He induced reactions. Two decades later, when more recent and higher voltage Van de Graaff accelerators had entered the scene and the 2MV machine could no longer contribute effectively in this field, the installation operation was redefined. Important modifications in the VDG system transformed the installation into the present 2 MV electron accelerator dedicated to the study of radiation effects in materials, becoming the European reference laboratory for fusion research in ceramics insulators with unique experimental systems for in-situ optical absorption and emission measurement, and the study of electrical and dielectric properties (DC to GHz), as well as He and H isotope diffusion.

A simplified experimental scheme is given in Figure 1, where radioluminescence measuring system, including

photomultiplier detection unit and a monochromator, is shown at the end of the accelerator line. Due to the high radiation levels during operation (breemstrahlung), shielding of sensitive elements is required and synchronous detection used to reduce the radiation background. Remote control is needed for every action, greatly complicating experimental designs.

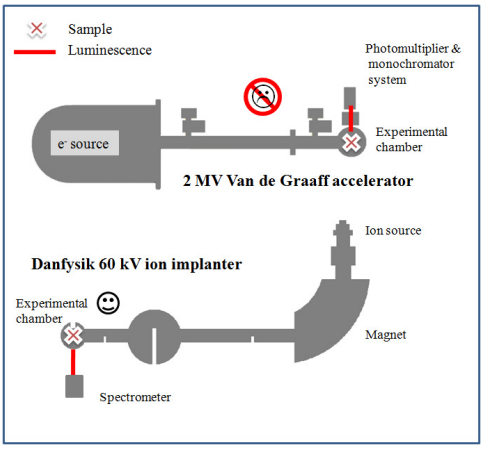

Fig. 1. Schematic view of Van de Graaff and Ion Implanter installations

1.2. The Danfysik 60 KV ion implanter

Ion implantation accelerators, of multiple energies and currents, are widely used in industry e.g. for semiconductor doping processes. Ion implantation implies material damage: incident ions transfer their energy and momentum to target atoms causing near-surface alteration of important properties such as optical transmission or electrical conductivity. This aspect must be considered particularly in insulating elements for fusion applications, highly susceptive to the expected constant ion bombardment due to local electric field induced acceleration of residual ionized ionized gas in future fusion devices [3-4].

The CIEMAT 60 kV ion implanter is mainly used for the generation of surface radiation effects comprising sputtering, ionization, and displacement and the study of their influence on physical properties, in particular electrical conductivity. Unlike the VDG, the absence of radiation background permits direct handling of different mechanisms and the use of modern solid-state devices for light collection. In this installation the ion induced luminescence from samples is focused by means of lenses into an optical fibre connected to a spectrometer with a CCD image sensor, (Hamamatsu, TM series C10083CAH). This way, full IBIL spectra from 200 to 1100 nm may be recorded in few milliseconds.

For both the electron and ion accelerators similar experimental chambers and sample-holders are used, including an oven and a cooling system that permits experiments to be made at different temperatures. Volume and surface electrical conductivity are respectively measured by applying a voltage between different electrode configurations sputtered onto the samples [5-6].

The long term usefulness and adaptability of these two laboratories, where experimental setups are under continuous development according to new needs, is now well established. The lastest modifications in the ion implanter, including replacement of complete ion source unit, responded to the need to improve ion beam stability, of great importance for luminescence experiments [7]. Furthermore, the implementation of optical fibre plus spectrometer system has been planned in the VDG line in order to simplify RL measurements: photomultiplier tubes are extremely fast, sensitive devices and are less susceptible to radiation than multichannel digital image sensors, making them particularly appropriated for use in a radiation environment. However, photomultipliers do not

distinguish photon energies, hence the use of a monochromator is necessary in order to select desired wavelength. With the combined system, taking complete spectra from 200 to 900 nm requires as much as 20 min, far too long to consider the same state being maintained in high dose rate irradiations. The planned spectrometer change is being carried out in the first place by providing adequate protection for the most sensitive elements through physical shielding and light transmission by means of mirrors in order to avoid areas with the highest radiation exposure.

2. Luminescence as a characterization tool

Luminescence techniques are commonly used in different scientific and industrial areas as simple non-destructive characterization tools. However, implementation of radiation induced luminescence based on *in situ* characterization systems clearly involves a number of difficulties. Well controlled conditions, characteristic of typical cathodoluminescence or thermoluminescence tests, cannot be assured in radiation facilities where different types of radiation will be present, and temperatures will rise depending on the location up to several hundred degrees.

Characteristic emissions corresponding to well defined level transitions of luminescence spectra at low temperature broaden into wide bands as temperature increases, reducing resolution. Moreover, signal intensity may significantly decrease due to a enhanced probability of non-radiative de-excitation, also reducing sensitivity.

Experiments carried out in SiC and Al_2O_3 show that the luminescence maintains both signal intensity and sufficiently spectral structure well above room temperature [7-8] for this purpose.

A clear example of this for SiC is given in Figures 2 and 3. Figure 2 shows electrical conductivity as a function of 1.8 MeV electron irradiation dose for two samples (HP SiC B1 and HP SiC B2) which correspond to different batches of “hot pressed” SiC obtained from the same company and nominally identical. As may be seen, the two lots of material showed more than one order of magnitude difference in initial conductivity values.

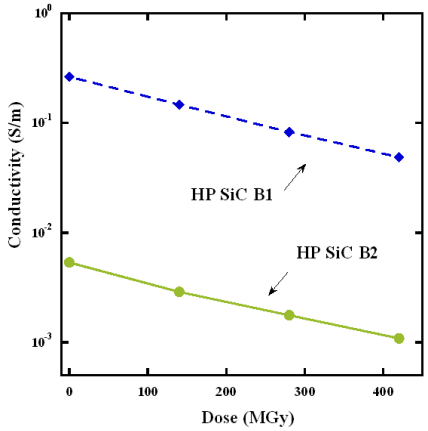

Fig. 2. Electrical conductivity for HP SiCas a function of 1.8 MeV electron irradiation at 450 ºC, 7kGy/s. Data from [12]

In Figure 3, RL spectra for both batches show the similar difference, the intensity for B2 being almost four times higher than B1. The behaviour with dose has been found to be the same for the two samples in both cases, indicating similar mechanisms are responsible for changes in conductivity and luminescence. This illustrates radioluminescence capability as a characterization tool for material quality control above room temperature, and a clear correlation between luminescence and conductivity, as discussed bellow.

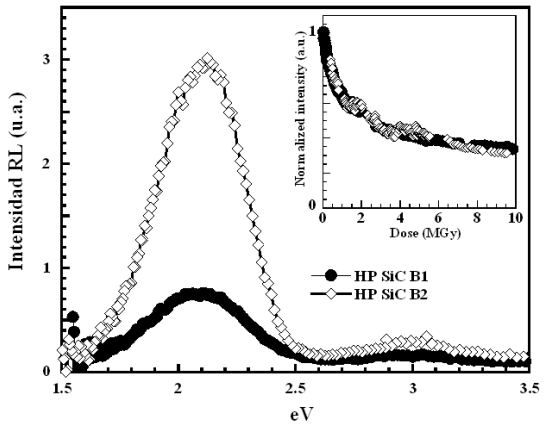

Fig. 3. Radioluminescence spectra for HP SiCas a function of 1.8 MeV electron irradiation at 450 ºC, 7kGy/s.

For the same type of material, experiments performed at IMR, Sendai (Japan) during collaborative experiments for 1 MeV proton irradiation show that comparable results are obtained using very different systems, energies, temperatures and dose rates if compared as a function of ionizing dose (fig.4), suggesting a general use of this technique for different irradiation conditions is feasible [9].

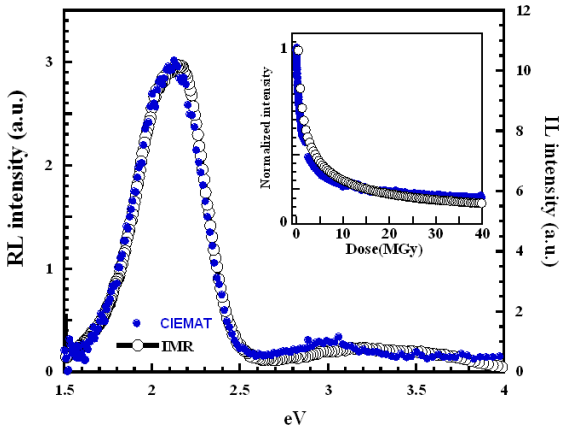

Fig. 4. Radioluminescence (CIEMAT) and IBIL (IMR) spectra for HP SiC. Inset: evolution as a function of ionizing dose. Data from [9]

3. Luminescence for in situ material verification

Alternating measurements of luminescence and surface electrical conductivity induced by 45 keV He^+ ion bombardment carried out for different grades of polycrystalline α-Al_2O_3 and high purity sapphire indicate severe damage in surface conductivity can be predicted by identifiable changes in oxygen vacancy related luminescence bands [7].

In Figure 5 an example for sapphire, where luminescence intensity at 340 nm corresponding to F^+ centre emission (oxygen vacancy with one electron trapped [10]) and surface electrical conductivity evolution as a function of irradiation time are compared, is given. A reduction of about 60 % of the maximum intensity reached by F+ band has been observed to be an evident precursor to the high conductivity increase. This has been shown to occur

irrespective of temperature, dose rate, and material type, indicating a clear correlation between luminescence and surface electrical degradation.

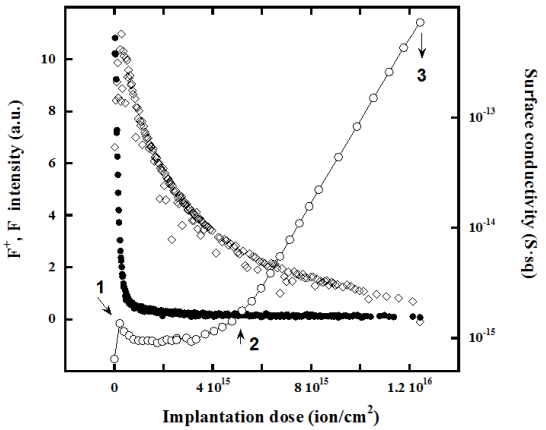

Fig. 5. F and F^+ peak intensities for sapphire plotted as a function of dose, together with the surface conductivity. Figure from [7].

Luminescence has also been successfully employed to qualify the degree of recovery of ion bombardment induced damage in alumina accomplished by posterior thermal treatments in vacuum and in air, as described elsewhere [7]. This correlation between IBIL and surface electrical degradation as a function of ion dose show the capability of the technique to remotely monitor the degradation of insulating materials during operation, giving the possibility to anticipate material degradation and hence contribute to device safety.

4. Luminescence for radiation damage process interpretation

Understanding of basic phenomena leading to degradation is essential for the development of new advanced functional materials and composites. These are usually complex materials for which radiation effects have not been successfully interpreted and available computational models fail to describe. Luminescence in this case might be of special interest in order to help clarify underlying mechanisms which take place under irradiation.

Silicon carbide, a covalent compound with more than 200 possible structures or ‘polytypes’, represents a very illustrative example. Monolithic SiC and SiC based composites are considered for different applications in fusion [11]. In spite of its nominally attractive low activation and radiation resistant characteristics, it has been found to be an extremely unpredictable material, showing a variety of results for different polytypes and fabrication methods.

Although certainly insufficient, significant results for hot pressed SiC have been obtained that suggest the same mechanisms dominate luminescence and conductivity evolution with irradiation dose [12]. Figure 6 shows volume electrical conductivity evolution as a function of ionizing dose normalized to initial values.

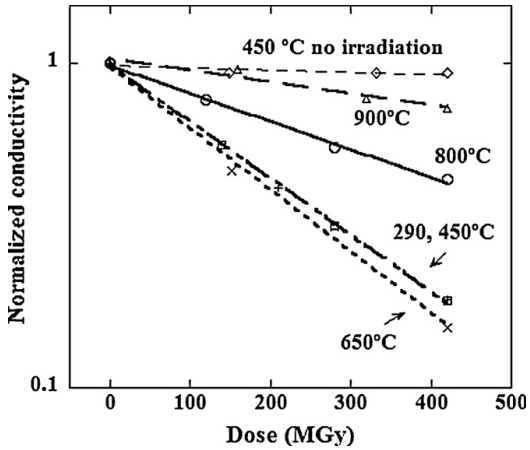

Fig. 6. Normalized volume electrical conductivity as a function of dose for HP SiC irradiated at 290, 450, 650, and 900 ºC. Figure taken from [12].

In Figure 7 the initial RL spectrum is compared with RL spectra taken after 1.8 MeV electron irradiation at different temperatures up to 420 MGy. For irradiations between 290 and 650 ºC the radiation damage in HP SiC did not show any significant temperature dependence. However, for higher temperatures the rate of change decreases, according with partial damage recovery during irradiation. The luminescence intensity decrease for the high dose samples irradiated between 290 and 650 ºC is very similar, while for the sample irradiated at 800 ºC the reduction in RL is minimum (fig. 7), in agreement with conductivity results (fig. 6). Besides, whereas no matches were found for different examined materials (CVD SiC, RB SiC, and HP SiC) in similar experiments regarding electrical conductivity, it was possible to identify common RL bands for all three materials, and could be the supporting point for the development of a common theory for radiation damage in SiC.

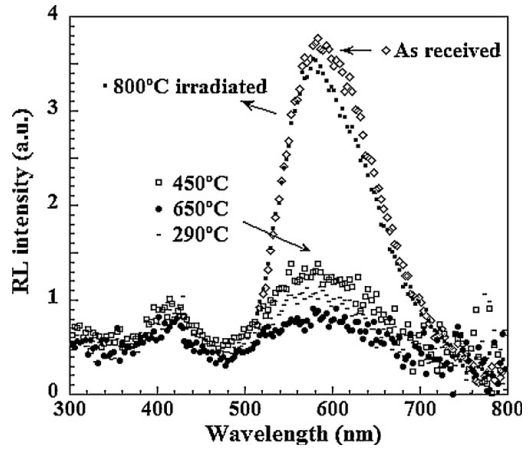

Fig. 7. RL spectra for HP SiC at 35 °C, for as-received samples and following prior irradiation to 420 MGy at 290, 450 and 650, and 800°C. Figure taken from [12].

Although considerably more information concerning basic radiation effects is available for Al_2O_3, luminescence has provided important data which has helped to develop a model for surface damage under ion bombardment. Different stages for surface electrical conductivity degradation observed in several types of alumina and sapphire, 1, 2, and 3 in Figure 5, have been interpreted according to oxygen vacancy (F and F^+) luminescence centre intensity evolution as a function of irradiation time [7,13]. Luminescence spectra also reveal the presence of oxygen vacancy complexes which could be precursors to higher order aggregates leading to severe damage.

Conclusions

Main characteristics of two radiation facilities at the CIEMAT, where parallel electrical conductivity and ion/electron beam induced luminescence measurements are carried out, have been briefly described.

Important results which illustrate the utility of luminescence for fusion applications have been selected for this overview. Although the task of implementing a luminescence based method for monitoring material performance during irradiation is difficult, the potential advantages justify further work being developed in this regard.

Moreover, the study of luminescence constitutes an end in itself, having been shown to be an efficient means to interpret radiation damage processes.

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