Charge trapping induced by plasma in alumina electrode surface investigated by thermoluminescence and optically stimulated luminescence

P. F. Ambrico,1 M. Ambrico,1,2,a) L. Schiavulli,2,3,4 T. Ligono,2,4 and V. Augelli2,4

1CNR-IMIP Sezione di Bari, Via Orabona, 4 I-70126 Bari, Italy
2CNISM Unità di Ricerca Bari Università, Università degli Studi di Bari, Via Amendola, 73 I-70126-Bari, Italy
3INFN–Sezione di Bari, Via Amendola, 173 I-70126 Bari, Italy
4Dipartimento Interateneo di Fisica, Università degli Studi di Bari, Via Amendola, 173 I-70126 Bari, Italy

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The plasma of a dielectric barrier discharge can fill traps in the alumina that cover the electrode. Trap energies and lifetimes are estimated by thermoluminescence and optically stimulated luminescence. Comparison with similar results for traps created by other radiation sources clarifies the mechanisms regulating this effect. Alumina’s trap energies are approximately 1 eV, and the traps remain active for several days after plasma exposure. These results could be important to keep dielectric barrier discharge plasmas uniform since a trapped charge can be an electron reservoir. © 2009 American Institute of Physics. [DOI: 10.1063/1.3076122]

Various groups have recently investigated how the dielectric barrier discharge plasma interacts with its electrodes. Laser light at grazing incidence on alumina electrodes1–3 enhances plasma emission and affects the plasma current. Li et al.4 showed that the charge remains on quartz and Teflon electrodes after the discharge. However, their technique using a thermally stimulated current could not confirm charging in alumina nor give a reliable estimate for the trap energies filled by exposure to the plasma.

This letter reports experimental evidence of the plasma induced trap filling effect on alumina electrodes used in dielectric barrier discharge with purely optical characterizations techniques such as thermoluminescence and optically stimulated luminescence. Both techniques are commonly used in radiation dosimetry and, additionally, are performed on a wide range of insulating materials that, after radiation exposure, display luminescence effects. Basically, the thermoluminescence and optically stimulated luminescence effects originate from traps, previously filled by an incident radiation, that are depleted by heating5 or, in a selective way, by using an incident monochromatic light6 (termed bleaching). The thermoluminescence output is a glow curve representing the luminescence intensities versus the heating temperature,5 while the optically stimulated readout is a decay of the luminescence intensity versus time.6 The energy and lifetime of the trap can then be determined by the complementary use of these techniques. Despite intrinsic limitation of both characterizations due to sensitivity to luminescence phenomena, delayed from seconds to years with respect to the excitation, unlike the thermally stimulated current technique, thermoluminescence and optical stimulated luminescence are not limited by material low carrier mobility. Moreover, thermoluminescence is particularly suitable for testing alumina since this material is competitive with LiF (Ref. 7) in radiation dosimetry.8,9

The measurements were made on a series of alumina slabs (COORSTEK fine grain alumina, purity 96%, area 5×5 mm², 0.7 mm thick) each cut out from those used as electrodes in the region exposed to the volume parallel plate dielectric barrier discharge plasma. The discharge apparatus is the same type as the one described in Ref. 2. For the present measurements, a more specialized reactor with smaller electrodes was used to run an atmospheric pressure air discharge (N₂/O₃). The parallel plates discharge is composed of two 20×12 mm² copper electrodes, which are completely covered with two alumina plates with a surface of 30×40 mm², and four spacers that fix the discharge gap at 2 mm. The high voltage applied is a sine wave with 2 kHz frequency and a peak to peak value of 19 kV. The plasma exposure time was approximately 6 h. It is worth noting that only the central region (20×12 mm²) of the alumina plate exposed directly to the plasma has been examined, corresponding to about ten samples for each discharge process. Thermoluminescence spectra have been collected in the temperature range 320–720 K at a heating rate of 5 K/s with an automated RISØ TL/OSL-DA-15 reader. The beta particle radiation, produced in the same experimental apparatus, with a 90Sr/90Y source (Emax=2.28 MeV; beta current of 1.5 GBq/s, i.e., 1.5×10⁹ electrons/s, dose rate of ~0.119 Gy/s, and exposure time of t=90 s), and the ultraviolet (UV) light emission (exposure time 10 min) of a Lot Oriel LSP 035 mercury pen lamp10 have been used as alternative radiation sources. It was inferred that multitrap emission/recombination mechanisms occurred since the glow curves consisted of a weighted superposition of thermoluminescence peaks. The peak temperatures Tm and energies Em have been extracted by deconvolution of the experimental data using GLOW FIT, a free downloadable software.11,12

Optically stimulated luminescence has been performed on the same set of sample, and the bleaching was achieved by using a blue light emitting diode (LED) (λ=470 nm, Pmax=36 mW) at an incident power fixed at the 90% Pmax for t=600 s. In order to provide evidence for the bleaching effect in selectively depleting plasma filled traps, the thermoluminescence glow curves were collected before and after the optically stimulated luminescence. It is noteworthy that because after thermoluminescence experiments all plasma...
filled traps are fully depleted, the comparison of glow curves required a set of two samples, which, of course, must refer to the same discharge process.

In Fig. 1, the collected thermoluminescence glow curves for slabs used as electrodes in barrier discharge plasma (1) and after beta particle (2) or UV light irradiation (3) are displayed. Virgin alumina has never shown any glow peaks, thus indicating that no previously filled traps due to accidental environmental radiation exposure are present (4). The full set of the calculated fitting parameters of glow curves in Fig. 1 is summarized in Table I. Figure 2 shows an example of the data fitting results and specifically the experimental data on plasma exposed alumina together with the calculated glow curves and residuals versus temperature values. Evidence of the accuracy of the fit is shown by the residual values lower than 10% for plasma exposed and 5% for beta particle and UV light exposed alumina in the range of 400–700 K. Four trap energy levels have been determined with corresponding values lower than 1.0 eV in agreement with values hypothesized in Ref. 5.

The glow curve shows similar behavior for the plasma and beta irradiated sample, while the UV light irradiated samples are quite different. It can be inferred that plasma induced charge trapping is mainly due to the energy transfer from the plasma electrons to the alumina ones rather than to the phototransfer effect. The lower thermoluminescence intensity can be attributed to the electron energy whose mean value in plasma is typically of the order of a few eV, i.e., five orders of magnitude lower than the beta source, leading to an electron penetration depth of around tens of nanometer. Therefore the plasma electrons are effective mostly on the first monolayers of alumina surface (where they are readily stopped) rather than on the overall bulk, which is instead fully crossed by beta radiation. Under the hypothesis that the area under the thermoluminescence glow curve is linearly dependent on the total absorbed radiation dose (i.e., dose rate × exposure time), the plasma radiation was found to be approximately 2 Gy, i.e., about five times lower than total beta dose, resulting in a radiation dose rate of 9 × 10⁻⁵ Gy/s. Moreover, the ratio between the total area values under the thermoluminescence glow curve of beta and plasma radiation allows a rough estimate of the energy of plasma electrons at about 1 eV, in good agreement with those calculated by numerical modeling simulation.

The comparison with luminescence intensities due to UV photons suggests that the plasma radiation dose is also much lower than the mercury lamp dose and also that charge trapping due to photons is more effective. Furthermore, while in plasma and beta irradiated alumina, the relative intensities of the main peaks are almost the same; those of UV irradiated alumina display a strong reduction in the low temperature peak with respect to the high temperature peak. This could be ascribed to a bleaching superimposed to trap filling during mercury lamp irradiation; in fact, the collected glow curve is similar to that commonly observed after optically stimulated luminescence readout, since the lamp emission includes also lines in the wavelength region of the blue LED (see above and Ref. 10). Figure 3 shows the optically stimulated decay curve of the plasma exposed alumina together with the data fitting to a second order exponential behavior; the trap decay times were \( t_1 = 79 \pm 4 \) s and \( t_2 = 2010 \pm 132 \) s, higher than the corresponding decay time observed in alumina exposed to beta (\( t_1 = 42 \pm 2 \) s, \( t_2 = 179 \pm 10 \) s) or UV radiations (\( t_1 = 154 \pm 3 \) s). In the inset, the glow curves before and after optically stimulated luminescence readout exhibit the bleaching effect on traps up to \( T = 525 \) K, while for a trap at \( T = 622 \) K, the depletion is not effective. The longer trap lifetimes also explain the glow curve detection after 2 weeks from plasma exposure.

In conclusion, thermoluminescence and optically stimulated luminescences have been demonstrated as new and original tools to accurately explain the plasma induced trap filling effect on alumina. With respect to previous findings,

FIG. 1. Thermoluminescence glow curves of alumina (1) used as electrodes in \( N_2/O_2 \) DBD plasma, (2) after 90 s exposure time to beta radiation, (3) after 10 min exposure to the UV radiation of Hg lamp, and (4) virgin alumina not exposed to any radiation source. The thermoluminescence curve (1) for plasma exposure has been multiplied by a factor of 20 for clarity.

FIG. 2. Experimental thermoluminescence glow curve of \( N_2/O_2 \) plasma exposed alumina (curve 1 in Fig. 1) and corresponding fitting by using the GLOW FIT program. Here the glow peaks contributing to the glow curve have been also displayed. It can be observed that the peak intensities follow the first order kinetics (see Refs. 5 and 11). The inset shows the calculated residuals vs temperature; residual values below 10% have been found in the range between 400 and 700 K. Some of the glow peak intensities have been multiplied by a factor of 10 for clarity.

TABLE I. Thermoluminescence glow peak temperatures \( T_m \) and corresponding trap energy levels \( E_o \) extracted as fitting parameters by applying the GLOW FIT program to glow curves in Fig. 1. It could be observed that almost the same set of fitting parameters resulted for beta and plasma irradiated sample, while slightly different values have been observed in mercury lamp irradiated alumina.

<table>
<thead>
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<th>No.</th>
<th>( T_m ) (K)</th>
<th>( E_o ) (eV)</th>
<th>( T_m ) (β) (K)</th>
<th>( E_o ) (β) (eV)</th>
<th>( T_m ) (plasma) (K)</th>
<th>( E_o ) (plasma) (eV)</th>
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<tr>
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<td>0.575</td>
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<td>3</td>
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<td>0.628</td>
<td>626</td>
<td>0.543</td>
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</tr>
</tbody>
</table>
for fruitful scientific discussion on DBD plasma. This work was possible thanks to the Laboratorio di Ricerca per la Diagnostica dei Beni Culturali dell’Università degli Studi di Bari.

12The GF program works under the hypothesis that the general one trap model holds for all TL peaks in the slow retrapping conditions, i.e., following the Randall–Wilkins equation (see Refs. 5 and 11). The measurements performed after a long interval of time between plasma exposure and thermoluminescence measurements demonstrate that the effect is long lasting and that the presence of deep filled levels suggests that the electron release could probably not be easily achieved on the overall trapped charge levels. These results could be also helpful for a better understanding of the effect of trapped charge on influencing the pressure range where uniform dielectric barrier discharge occurs and explaining the origin of the laser surface plasma emission phenomena.

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