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THERMALLY STIMULATED DEPOLARIZATION CURRENTS IN NEUTRON IRRADIATED Al_2O_3

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Two thermally stimulated depolarization current peaks due to dipolar relaxation processes have been observed at 242 and 273 K in neutron irradiated Al_2O_3. The results on fluence dependence and thermal stability indicate that the second one may be related to F-type centres or their corresponding interstitials induced by irradiation.

Introduction

Radiofrequency heating systems in fusion reactors require transmission windows to isolate the source from the plasma. Low loss materials are then needed for these windows, Al_2O_3 being one of the candidates. Besides other physical properties, the effect of neutron irradiation on dielectric loss in a wide frequency range has to be studied. Work performed up to now shows increases in permittivity and loss tangent induced by particle irradiation (1-5). The results obtained at low frequencies (0.02 to 2 MHz) by Fowler (2) seem to indicate that there is a loss increase at lower frequencies induced by neutron irradiation, which might arise from dipolar defects. The thermally stimulated depolarization currents (TSDC) technique (6) is a powerful tool for the study of dipolar defects in dielectric materials. Its main advantages are its high sensitivity and ability to resolve competing relaxation processes (this last can not usually be made by frequency dependent loss measurements). It can also be used to detect any type of relaxation processes, such as Maxwell-Wagner effects or space charge polarization.

The purpose of this paper is to present results on TSDC in the single crystal and ceramic Al_2O_3 samples used by Fowler (2). Optical absorption spectra of single crystals have also been obtained. Two dipolar relaxation peaks have been detected, the behaviour of one of them being similar to that of F-type centres.

Experimental

Samples of single crystal Al_2O_3 (0 degree cut Linde) and polycrystalline Al_2O_3 (Coors AD995) were irradiated at around 400 K up to $5x10^{18}$, 10^{19}, $2x10^{21}$ and 10^{22}n m^{-2}, in the spallation neutron source at the Los Alamos Meson Physics Facility (LAMPF). This neutron source is characterized as having a fast fission reactor-like spectrum, but with a higher energy tail (7).

TSDC spectra between 10 and 310 K have been measured in vacuum in a Displex CSW-202 closed-cycle refrigeration system at a heating rate of 7 K min^{-1}. At this rate the thermal gradients across the sample are negligible. The depolarization currents were measured with a Keithley 617 electrometer. A Fluke 415 B high voltage power supply has been used for electric field (0-20.000 V cm^{-1}) application to the sample electrodes, made either from indium or evaporated platinum. Optical absorption spectra were obtained with a Cary 17 spectrophotometer.

Results and Discussion

Unirradiated control samples show only a weak TSDC peak above room temperature. In neutron irradiated Al_2O_3 single crystals peaks at 242, 273 and 305 K appear (figure 1). Only the 273 and 305 K peaks are observed in polycrystalline samples (figure 2). No detectable TSD currents have been obtained for a neutron fluence of $5x10^{18}$n m^{-2} in single crystals and for $5x10^{18}$ and 10^{19}n m^{-2} in polycrystalline Al_2O_3. These peaks have been isolated by choosing adequate polarization temperatures and by using the thermal cleaning method, as can also be seen in figures 1 and 2.

The 242 and 273 K peaks have been fitted to first order kinetics processes by a recently proposed fitting method (8). Their activation energies (E_o) and relaxation times (τ_g) are, respectively, 0.66 $\pm$ 0.02 eV and 1 x 10^{-12} s for the first peak, and 0.72 $\pm$ 0.01 eV and 3 x 10^{-12} s for the second one. Their temperature at the maximum (T_m) does not depend on the polarization temperature (T_p). A linear relationship between the peak area and the polarizing field E_p, at least from 0 to 20.000 V cm^{-1}, has been found for both. These facts allow us to ascribe these peaks to dipolar processes induced by neutron irradiation.

On the other hand, for the 305 K TSDC peak, T_m increases with increasing T_p. It might be due to a polarization process other than a dipolar one, perhaps a space charge one. This process has not been investigated any further.

The dependence on the neutron fluence (D) of the TSDC is shown in figure 3. In this figure the values of $N\mu^2$, which are proportional to the

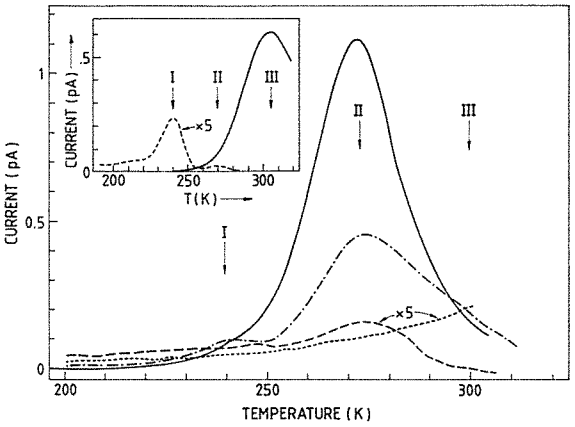

Fig. 1. TSDC spectra (T_p=270 K, E_p=20000 V cm^{-1}) in unirradiated (.....) and in neutron irradiated up to 10^{19} (———), 2 x 10^{21} (-.-.-.-) and 10^{22} (———) n m^{-2} single crystal Al_2O_3 samples. The inset shows the effect of thermal cleaning for 1 minute at 270 K after polarization at 295 K (———) and a TSDC spectrum by polarizing at 233 K (------)

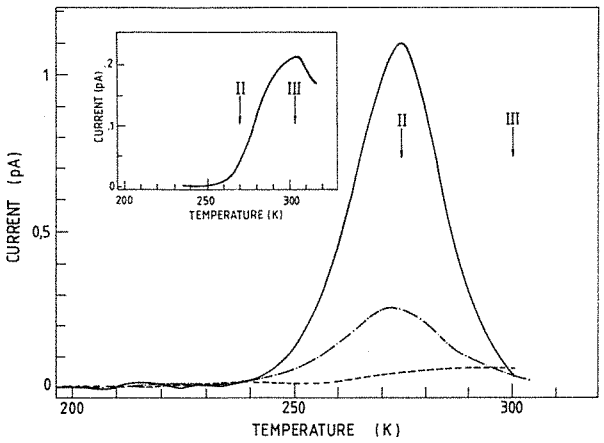

Fig. 2. TSDC spectra (in the same conditions as single crystals) in unirradiated (-----) and in neutron irradiated up to 2 x 10^{21} (-·-·-) and 10^{22} (———) n m^{-2} polycrystalline Al_2O_3 samples. The inset shows the effect of thermal cleaning for 1 minute at 270 K after polarization at 309 K.

peak areas (6), are plotted against fluence (N being the dipole concentration and μ the dipolar moment). From the available experimental data, there seems to be a linear relationship in a log-log plot for the 273 K peak in single crystals, this would indicate that $N\mu^2$ is proportional to D^{α}, α being 0.6 approximately. A similar behaviour might occur for this peak in polycrystalline samples. The 242 K peak behaves with fluence in a different way, as can also be seen in this figure.

The well known F band at 203 nm and F+ bands at 227 and 255 nm (9, 10) are clearly observed in the optical absorption spectrum of

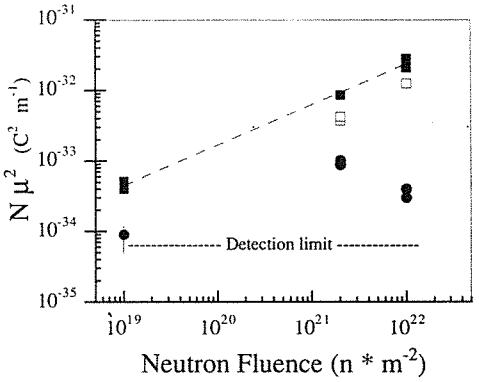

Fig. 3. Dependence on fluence of the 273 K peak area in single crystal (■) and polycrystalline (□) Al_2O_3 samples and of the 242 K peak area in Al_2O_3 single crystals (●).

neutron-irradiated single crystals. Their fluence dependence is linear in a log-log plot, in good agreement with other authors (see ref. 11). The slope α is approximately the same as that for the fluence dependence of the 273 K TSDC peak. Therefore the ratio of the $N\mu^2$ value for this peak to the F-type centre concentration is roughly constant for all neutron fluence values.

In order to study the thermal stability of defects related to these neutron-induced TSDC processes, a pulsed thermal annealing method has been used. Two single crystal samples (both showing exactly the same 273 K peak area in the as neutron irradiated condition) and a polycrystalline one, all of them irradiated up to 10^{22}n m^{-2}, have been employed. A TSDC spectrum is obtained after each heating pulse up to a temperature value in all samples, one of the single crystals being each time X-ray irradiated at room temperature at a fixed dose before polarization. These X-ray irradiations are intended to distinguish between electronic annealing processes (by thermal emptying of the centres or annihilation by recombination with a carrier released from other traps) and ionic annealing processes (by thermal destruction of the centres), since ionizing irradiation repopulates those centres which remain in case that an electronic process takes place (12). The peak areas are plotted against the corresponding annealing temperature. The results for the two peaks are shown in figure 4. The annealing curve for the 273 K peak in both types of samples shows one step between 420 and 800 K. The 242 K peak in single crystals vanishes between 370 and 470 K. X-ray irradiations only induce a slight decrease of both peak intensities corresponding to each annealing temperature, as seen in this figure. It is worth noting that this effect is more pronounced in the temperature range where the first peak exists. It is then reasonable to conclude that ionic annealing processes are taking place for both TSDC processes, since these are not regenerated by X-ray irradiation. Nevertheless charge rearrangement phenomena involving the dipolar defects are induced by

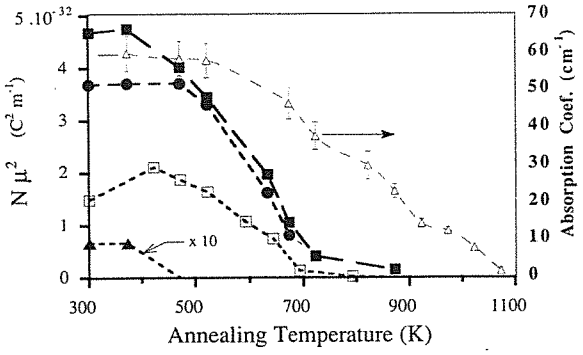

Fig. 4. Annealing curves of the 273 K TSDC process in single crystals (■) and polycrystals (□) irradiated up to 10^{22} n m^{-2}. Effect of X-ray irradiation after each heating pulse (●) for the single crystals. Also shown the 242 K TSDC peak (▲). Annealing curve for the 255 nm F+ band (△) (the F centres behave as the F+ ones).

these irradiations, as indicated by those slight decreases in the peak areas. The fact that these phenomena are more effective below 470 K can be explained by the existence of trapping centres (for these charges released by X-ray irradiation) whose concentration is higher below this temperature than above it. Dipolar defects related to the 242 K peak, which anneal out at 470 K, are then good candidates for being these trapping centres.

The thermal annealing curves for the F and F+ centres in neutron irradiated samples show three steps, one between 450 and 800 K and the others from 800 to 1100 K (figure 4), in good agreement with previous results (12-16), although some authors have only observed one step coinciding with our first one. This occurs in the same temperature range as that for the 273 K TSDC process. About 40% of the initial F and F+ centres anneal out in the first stage. This means about 8 x 10^{23} and 2 x 10^{23} F and F+ centres per m^3, respectively, for the 10^{22}n m^{-2} irradiated sample. It is known that the F-type centres anneal by recombination with interstitial oxygen ions (13).

From our results on neutron fluence dependence and thermal stability it is sensible to ascribe the 273 K TSDC process to those anionic vacancies and/or their corresponding interstitials induced by neutron irradiation which are thermally unstable below 800 K. Taking into account the $N\mu^2$ value for a neutron fluence of 10^{22}n m^{-2} dipolar moment values of 3.8 x 10^{-28} C m (that is, 24 electronic charge x Å) and 1.6 x 10^{-28}C m (10 electronic charge x Å) are obtained for dipole concentrations of 2 x 10^{23} and 10^{24} m^{-3} respectively. The first value seems to be too high, hence dipoles based on defects related only to F+ centres (as F+-negatively charged interstitial pairs) can not explain the 273 K process. On the other hand the second dipolar moment value may be a proper one, thus indicating that this TSDC process may arise from dipolar defects related to both F and F+ centres. Since F centres are neutral they can be disregarded as being part of them. So dipoles would be formed by charged interstitial pairs. Some defects due to charged interstitials of oxygen and aluminium ions have been proposed to explain ESR signals observed in neutron irradiated Al_2O_3 (17, 18), but at the present moment we cannot elucidate which type of lattice defects may be involved in these dipolar defects.

By applying the Debye model (19) calculations of permittivity and loss tangent from the activation parameters of the TSDC peaks show that there must be a loss peak at very low frequencies (around 10 Hz at room temperature) which may be related to the results found by Fowler (2).

In summary, two neutron induced dipolar relaxation processes have been observed for the first time. At least one of them must be related to charged interstitials. More work is in progress to understand these processes observed by TSDC.

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