Journal of Nuclear Materials 155–157 (1988) 357–360
North-Holland, Amsterdam

DOSE RATE DEPENDENCE OF THE RADIATION-INDUCED ELECTRICAL CONDUCTIVITY IN MgO

E.R. HODGSON and S. CLEMENT

CIEMAT/Euratom Fusion Association, 28040 Madrid, Spain

The radiation-induced electrical conductivity (RIC), has been measured for single crystal MgO at dose rates between 10^2 and 10^7 Gy h^{-1} and temperatures from 14 to 450 °C. A correlation is observed between the RIC dose rate dependence, and the thermal stability of well defined electron and hole traps, in agreement with the model of Klaffky et al. The dose rate dependence factor is observed not to be constant, but to depend on dose rate. The possible effects of displacement damage and radiation induced impurity diffusion are noted.

1. Introduction

Recent work has shown that the radiation induced electrical conductivity (RIC) in refractory ceramic oxides depends in a complex way on dose rate, temperature, and sample impurity content [1–3]. The RIC is in general well described at any given temperature by $\sigma = KR^{\delta} + \sigma_0$, where σ_0 is the conductivity in the absence of radiation, R the dose rate, and K and δ constants. The exponent of the dose rate dependence δ, is determined at any given temperature by the number and type of trapped charges (electrons and holes), and as a consequences shows a dependence on increasing temperature due to the different trapping centres becoming thermally unstable [1,4].

The initial work of Klaffky et al. [1] on single crystal Al_2O_3 indicated some correlation between the temperature variation of δ and the observed thermally stimulated current (TSC) peaks, their results gave $\delta \leqslant 1$. In contrast, Pells et al. [2] observed for polycrystalline alumina and spinel, temperature regions in which $\delta > 1$.

The results presented here for single crystal MgO show a clear correlation between the observed thermal stability of identified electron and hole trapping centres, and the temperature variation of δ. Values for δ, greater than unity, are observed at certain temperatures. Furthermore the results indicate that δ, hitherto considered constant at a given temperature, is in fact dose rate dependent, its value decreasing as the dose rate increases.

2. Experimental procedure

The experiments have been performed in a sample chamber mounted in the beam line of a HVEC Van de Graaff accelerator, in which samples were irradiated in high vacuum ($\sim 3 \times 10^{-6}$ mbar) directly with 1.8-MeV electrons, and with bremsstrahlung γ-rays produced by stopping the electron beam in a gold target. The sample chamber contains a small oven and an insulated spring loaded electrical contact, which permits samples with sputtered platinum electrodes to be held in good thermal contact with the oven. An adjustable double collimator system is used to measure the electron beam current and align the beam onto the sample. In this way the RIC may be measured at any temperature between 14 and 750 °C during irradiation. Following irradiation the sample may be linearly heated at 10 °C min^{-1} to measure the TSC and the associated thermoluminescence (TL).

Measurements have been made for three nominally pure MgO single crystal samples (99.99 and 99.9% purity from W.C. Spicer, U.K.) containing approximately 160, 180, and 650 wppm iron. The three samples classified in terms of radiation induced optical absorption bands and thermal annealing following irradiation, together with TL and TSC in previous experiments [5,6], have been irradiated with bremsstrahlung γ-rays at between 44 and 440 Gy h^{-1} and directly with electrons at beam currents of between 10^{-7} and 10^{-2} A m^{-2}. The lower end of the beam current overlaps the γ-irradiations in equivalent dose rate, permitting a reliable calibration of the direct electron dose rates. Values of RIC have been obtained for the three samples at temperatures between 14 and ~ 400 °C. At each value of temperature and dose rate the RIC has been measured for polarization voltages of 65, 113, and 160 V. This checks for the ohmic behaviour of the electrical conductivity, and permits one to obtain a value of the beam current on the sample which may be checked with that determined from the collimators.

The samples have also been irradiated for 3 h at 14 °C with bremsstrahlung γ-rays at 350 Gy h^{-1} and directly with electrons at 3.0×10^{-7} A m^{-2} (equivalent to 350 Gy h^{-1}), and then heated following irradiation to obtain the TSC and TL spectra.

In this way values of δ for the three samples have been obtained over a range of temperatures, together with the characteristic TSC and TL spectra which permit direct comparison with the previously measured optical absorption spectra and avoid any possible error due to temperature measurement.

3. Results

Fig. 1 shows a typical result for the RIC as a function of dose rate for the 180 ppm Fe sample at

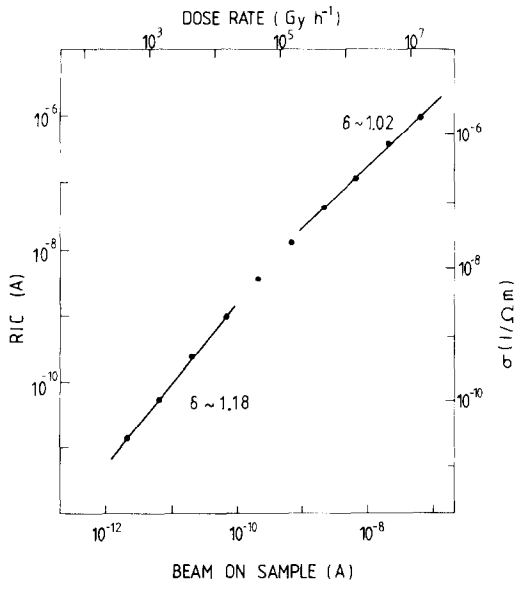

Fig. 1. Radiation induced electrical conductivity in the MgO single crystal as a function of dose rate for the 180 ppm Fe sample at 14 °C.

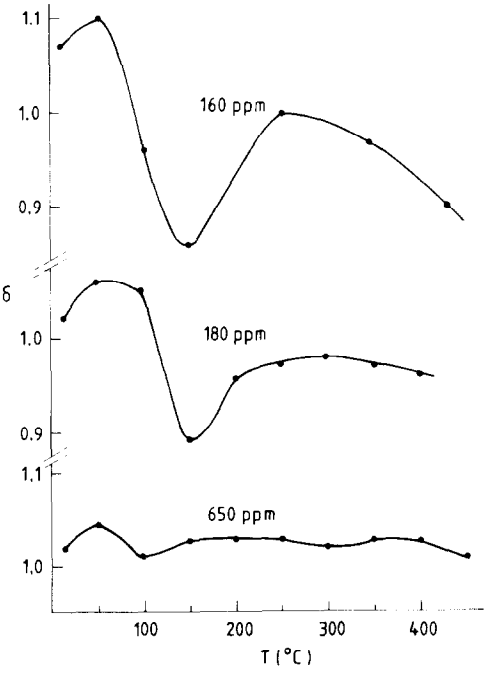

Fig. 2. High dose rate δ values in $\sigma = KR^{\delta} + \sigma_0$ for the samples with different Fe impurity contents as a function of temperature.

14 °C, in which one observes a definite curve in the log–log plot, the slope (equivalent to δ) being less at higher dose rates. This is general in all the results and only in a few isolated cases is δ approximately constant over the whole dose rate range. Fig. 2 shows the high dose rate δ values ($\gtrsim 10^5$ Gy h^{-1}) as a function of temperature for the three samples. The 160 and 180 ppm Fe samples show essentially the same structure: a narrow maximum around 50 °C, a sharp decrease near 100 °C with a minimum at about 150 °C, and a further broad maximum near 250–300 °C. In contrast, the 650 ppm Fe sample shows little structure, with only the 50 °C maximum being observed.

In fig. 3 the TSC spectra are given, each following a 3-h electron irradiation at 3.0×10^{-7} A m^{-2}. Identical results are obtained under bremsstrahlung γ-irradiation at the equivalent dose rate. Again the two low iron content samples give very similar results, a prominent structured peak being observed at about 100 °C. In the 650 ppm Fe sample, the 100 °C peak is about a factor 50 weaker. In all the samples the higher temperature peaks are similar in magnitude.

Previous experiments on these three samples have been carried out, in which the radiation induced growth and subsequent thermal annealing of the Fe^{3+} band at 290 nm and the V-type centre bands at ~ 540 nm, together with corresponding TL and TSC spectra were reported [5,6]. Fig. 4 shows the radiation induced Fe^{3+} thermal annealing for the samples following irradiation at 350 Gy h^{-1}. The large annealing steps observed at about 100 °C for the 160 and 180 ppm Fe samples in which the Fe^{3+} content decreases, correspond to approximately 25 and 18 appm Fe^{3+}, respectively. Follow-

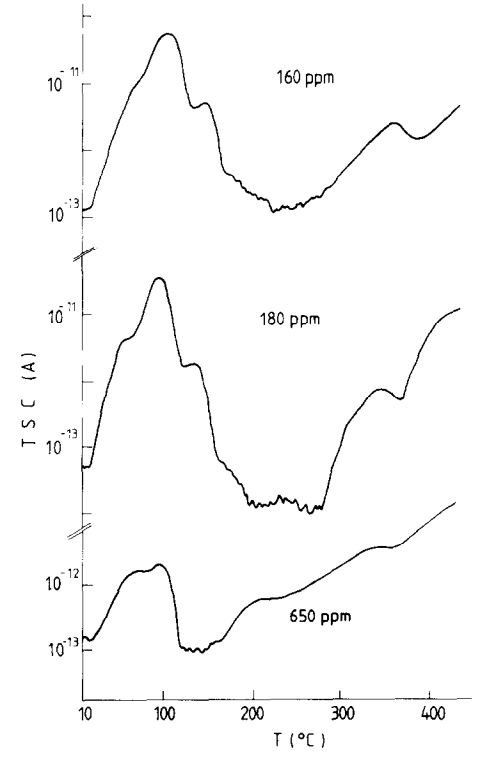

Fig. 3. TSC spectra for the samples with different Fe impurity contents, following irradiation at 3.0×10^{-7} A m^{-2} ($\triangleq$ 350 Gy h^{-1}) at 14 °C for 3 h.

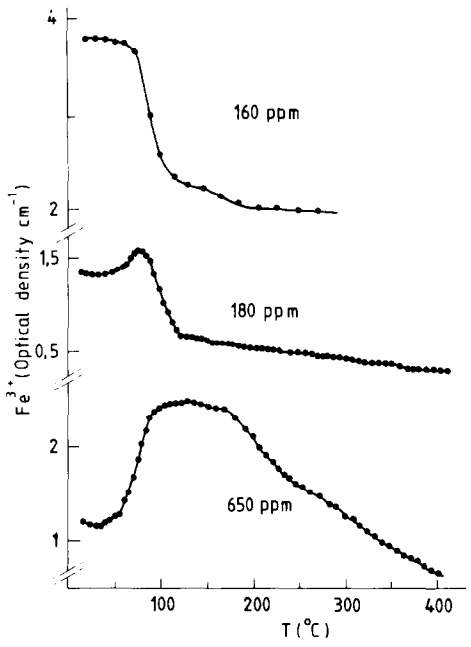

Fig. 4. Radiation induced Fe^{3+} annealing for the samples, γ irradiated at 350 Gy h^{-1}, 14 °C for 3 h.

ing this initial step, the radiation induced Fe^{3+} decreases slowly in a series of much smaller steps. The behaviour of the 650 ppm Fe sample is completely different, an important increase in the Fe^{3+} being observed between about 50 and 90 °C. In the same previous experiments, V-type centres (trapped hole centres) were observed to form during irradiation, and anneal out in all three samples between about 50 and 90 °C.

4. Discussion

Klaffky et al. [1] and Fisher [4] have been successful in fitting the general features of the temperature variation of δ, using a model based on electron and hole trapping [1]. The general features may be predicted with a reduced number of trapping centres (one hole and two electron traps). Some relation was found between observed TSC peaks in Al_2O_3 and the δ variation [1], however due to the numerous TSC peaks in Al_2O_3 a definite correlation was not possible. Other work on polycrystalline alumina and spinel has been limited to fitting the observed δ variation [4].

In the work presented here it has been possible to correlate the general features in the δ–temperature dependence with well defined electron and hole trapping centres. The previously observed and reported correlation between the annealing of radiation induced Fe^{3+} and V-type centres and the TL and TSC in MgO, served to identify the TSC current peaks. The low temperature peak at about 70 °C correlates with V-centre annealing and the associated hole release, the other peaks at ~100, ~150 °C and higher being due to electron release [5,6]. In fig. 2 one observes that all the samples show a maximum in δ between about 50 and 70 °C, which correlates with a peak in the TSC (fig. 3) at ~70 °C previously shown to be due to hole release from V-type centres. The Fe^{3+} increase observed at this temperature (fig. 4) for the 180 and 650 ppm Fe samples is also due to this hole release. The number of holes released in all three cases, as determined from the V-centre annealing, is $\sim 2 \times 10^{23}$ m^{-3}, and the observed effect on δ is very similar in all three samples. The sharp decrease in δ at about 100 °C for the 160 and 180 ppm Fe samples, correlates with the Fe^{3+} annealing stage (fig. 4). This Fe^{3+} annealing stage has been identified as being due to electron release from other impurities (in the main Cr) leading to Fe^{2+}, and gives rise to the large TSC peak observed at 100 °C. The effect on δ is greater in the 160 ppm sample and agrees with the observed larger Fe^{3+} annealing stage and TSC peak in this sample. The number of electrons released in this stage is $\sim 2 \times 10^{24}$ m^{-3}. Little if any δ variation is observed at this temperature in the 650 ppm sample. This is reflected in the absence of any Fe^{3+} annealing stage at 100 °C in this sample (fig. 4) and the considerably weaker TSC peak (fig. 3). Following the second electron TSC peak at between 120 and 150 °C in the 160 and 180 ppm samples, the δ value reaches a minimum and begins to increase towards a second broad maximum. In this region several overlapping electron TSC peaks are observed.

All these results help to substantiate the model predictions, in particular the lower temperature features in the δ variation. It is clear that for these samples the most important effect is due to the low temperature electron trapping centre becoming thermally unstable and giving rise to the sharp decrease in δ. The temperature region corresponding to supralinearity in the dose rate dependence of the RIC ($\delta > 1$) for the 160 and 180 ppm samples is the region in which the hole traps are thermally unstable, and agrees with the predictions of the model that thermal detrapping of holes must be included to obtain $\delta > 1$ [4]. The small variation in δ observed for the 650 ppm sample also confirms the model predictions for situations in which high temperature deep electron traps dominate.

The results obtained for δ indicate that this factor is dose rate dependent. A double dose rate dependence can be observed in some of the data of Klaffky et al. [1], although no comment is made on this point. Recent work using 20 MeV protons also shows a clear dose rate dependence for δ in polycrystalline spinel [7]. This non-constant aspect of δ is in effect included in the model of Klaffky et al., and is evident in some of their computer simulations for δ. It arises from the fact that the contribution to the free electron concentration due to thermal detrapping tends towards saturation as the dose rate increases. The actual critical dose rate at

which the number of trapped electrons tends towards saturation for any given trap, depends on the trap concentration, cross section, and thermal stability, and will in general be different for each trap [3]. Furthermore, it will be a strong function of temperature as the thermal stability is an exponential function of temperature. But in general one would expect the deviation in δ to become apparent on increasing dose rate, and to cause δ to decrease. This is precisely what is observed (fig. 1), the critical dose rate in this case being about 10^4–10^5 Gy h^{-1}. In the data of Klaffky et al. [1] one can observe a variation in δ for this dose rate range. The lower dose rate X-ray data of Pells et al. [2] ($\lesssim 10^4$ Gy h^{-1}), on the other hand shows no dose rate dependence in δ.

One may conclude that the results presented here, in which definite electron and hole traps have been observed to correlate with the salient features of the δ–temperature dependence, substantiate the Klaffky et al. model predictions. It is also clear that one must take into account the dose rate dependence of δ, which has to date been ignored.

It is to be expected that under prolonged irradiation in which displacement damage is accumulating, the V-type centre concentration will increase giving rise to an important $\delta > 1$ contribution, and furthermore radiation enhanced impurity diffusion will lead to electron trap modification [8].

Our thanks are due to Mr. E. Sánchez-Cabezudo for his help with these experiments.

References

[1] R.W. Klaffky, B.H. Rose, A.N. Goland and G.J. Dienes, Phys. Rev. B21 (1980) 3610.
[2] G.P. Pells, S.N. Buckley, G.J. Hill and M.J. Murphy, AERE Harwell Report R-11715 (1985).
[3] E.R. Hodgson and S. Clement, Radiat. Eff. 97 (1986) 251.
[4] A.J. Fisher, AERE Harwell Report M-3574 (1986).
[5] S. Clement and E.R. Hodgson, Phys. Rev. B30 (1984) 4684.
[6] S. Clement and E.R. Hodgson, Radiat. Eff. 97 (1986) 215.
[7] G.P. Pells, Harwell, private communication.
[8] S. Clement and E.R. Hodgson, Phys. Rev. B36 (1987) 3359.