

The distribution of the relaxation times and the thermally stimulated depolarization currents

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It is analysed the possibility to determine the relaxation parameters and their distributions by use of the Fractional Polarization Thermally Stimulated Discharge Current (FPTSDC) technique. For every elementary peak i there is a relationship between the activation energy W_i , the pre-exponential factor τ_{0i} , the temperature of the maximum current of the peak T_{mi} and the heating rate b , that can be used to obtain useful information concerning the values of the relaxation parameters or about their distribution. Realistic data for elementary peaks, in the range of the local β relaxation, are used for model simulations to analyze the lower limits below which we cannot discriminate between the existence of a distribution of the relaxation parameters and the normal uncertainty intervals. We will assume that at any temperature T the incertitude interval in activation energies ΔW is at least equal with $\pm kT$ and will determine the incertitude interval for τ_{0i} and for T_{mi} . The minimum or natural incertitude interval for the relaxation parameters will be analysed. As long as the experimental current can be fitted with an elementary peak and the uncertainties affecting the relaxation parameters are low, i.e. $\Delta W \cong kT_m$ and $\Delta\tau_0 \cong \tau_0$, the distribution functions cannot be determined accurately.

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1. Introduction

The main methods used for characterizing the dielectric properties of materials are: (i) alternating current (AC) measurements; (ii) isothermal charging and discharging current measurement (time domain spectroscopy) and (iii) thermally stimulated discharge current (TSDC) measurement. The TSDC method is widely used for determining the most important parameters to characterise the dielectric relaxation process, i.e. the activation energy W , the natural relaxation frequency α_0 (or the pre-exponential factor $\tau_0 = 1/\alpha_0$), the equilibrium polarization P_0 and the distribution functions for the pre-exponential factors and/or activation energies. The measured current is mainly determined by dipoles disorientation [1-4], but we must keep in mind that there is a polarization determined by the charge trapped at interfaces [1,5-7]. A global thermogram can be decomposed, at least theoretically, into a number of elementary peaks and consequently a distribution of relaxation times, determined by a distribution of pre-exponential factors and/or of activation energies can be assumed. For the time being, it is assumed that the only experimental technique allowing the decomposition of a distributed relaxation into its elementary processes or in subsets of the distributions, is the thermal sampling or fractional polarization (FP) or windowing polarization of TSDC. The assumption behind such an interpretation of the experimental results is that the system contains states with fixed relaxation times. Two different behaviour patterns are assumed: (i) When T_{mi} of the maximum of each elementary peak i is a function of the conditions of

polarization, a continuous distribution of relaxation times should be observed. This may be particularly true for secondary relaxations. (ii) If T_{mi} is independent of the polarization conditions, a discrete distribution of relaxation times should be postulated. On the other hand it has been claimed for more than 20 years that the universality of the non-exponential decay of polarization which is observed in many dielectrics results from the dynamics of correlated or cooperative systems rather than from a distribution of relaxation times [8, 9]. In this case it is assumed that the isothermal relaxation time is time dependent [8].

A good method to determine the relaxation parameters is by fitting the experimental data to the analytical expression of the current, using a small number of parameters and a small number of constraints on the parameters. The problem of distribution, whichever it is in activation energies or pre-exponential factors, is an inverse and ill-posed problem that generally cannot be uniquely solved. In this respect it is opportune to try and find some ways of using clear assumptions and simulations for very realistic values of parameters, to obtain information about the distributions. It is necessary to try to understand better what information can be obtained from elementary peaks analysis.

In a previous paper we have analysed the possibility to determine the relaxation parameters and their distributions by use of the FPTSDC technique in the temperature range of the primary (non-local) α relaxation [10].

The aim of this paper is to analyse the possibility to determine the relaxation parameters and their distributions by use of the FPTSDC technique in the temperature range

of the secondary (local) β relaxation. It will be employed the fact that for every elementary peak i there is a relationship between W_i , τ_{0i} , the temperature of the maximum current of the peak T_{mi} and the heating rate b , that can be used to obtain useful information concerning the values of the relaxation parameters or about their distributions. Realistic data for elementary peaks, obtained by FPTSDC technique in the range of the local β relaxation for polyethylene terephthalate (PET), are used for model simulations to analyze the lower limits below which we cannot discriminate between the existence of a distribution in the relaxation parameters and the normal uncertainty intervals in which the values of the relaxation parameters are expected.

2. Background considerations

The current density measured during the TSDC experiment can be written as a function of temperature [1]

$$j(T) = \frac{P_0(T_p)}{\tau(T)} \exp\left(-\frac{1}{b} \int_{T_0}^T \frac{dT'}{\tau(T')}\right) \quad (1)$$

where $P_0(T_p)$ is the equilibrium polarization produced during the polarization of the sample at T_p , $\tau(T)$ the relaxation time, $b=dT/dt$ the heating rate and T_0 the initial temperature. The maximum current occurs when

$$\frac{d\tau(T)}{dT} = -\frac{1}{b} \quad (2)$$

The relaxation time usually depends on a certain number of variable parameter like T , time (when there are coupling interactions between the relaxation species like in the case of ionically conducting solids [8,9] pressure and concentration. In simpler cases only one parameter, such as temperature T , is of primary interest. At low temperature in the range of the local β relaxation, the temperature variation of τ is given by an Arrhenius type relation

$$\tau(T) = \tau_0 \exp\left(\frac{W}{kT}\right) \quad (3)$$

where τ_0 represents the relaxation time at infinite temperature, W the activation energy of dipole (re)orientation. In a first approximation τ_0 is considered constant. In general τ_0 is a function of W , i. e. for a system characterized by a compensation behavior [11].

From Eqs. (2) and (3) results

$$\tau_0 \frac{W}{kT_m^2} \exp\left(\frac{W}{kT_m}\right) = \frac{1}{b} \quad (4)$$

where T_m is the temperature of maximum current of the peak. The Eq. (4) shows that W and τ_0 are connected. The

results obtained by taking into consideration only the variation or the distribution for one parameter, considering the other parameter constant, will conduct to an incorrect result. The relaxation time at T_m is

$$\tau(W, T_m) = \frac{kT_m^2}{bW} \quad (5)$$

It is useful to eliminate the pre-exponential factor τ_0 from Eq. (1) using Eq. (4). It results for the TSDC current density

$$j(T) = P_0(T_p) \frac{bW}{kT_m^2} \exp\left(\frac{W}{kT_m}\right) \exp\left[-\frac{W}{kT} - \frac{W}{kT_m^2} \exp\left(\frac{W}{kT_m}\right) \int_{T_0}^T \exp\left(-\frac{W}{kT'}\right) dT'\right] \quad (6)$$

The above relation can be used to determine the two adjustable parameters (W and $P_0(T_p)$) by fitting it to the experimental profile. $P_0(T_p)$ can be determined by integration of the experimental current in order to reduce the number of adjustable parameters.

Equation (1) can be generalized for the case when there is a Distribution in Activation Energies (DAE) that in turn will produce a Distribution in Pre-exponential Factors (DPF). In general it is assumed that there exists a distribution for the relaxation times $G(\tau)$. Assuming that the resulting TSDC arises from contributions of independent and parallel relaxation process, $j(T)$ [see Eq. (1)] can be written:

$$j(T) = \int_{-\infty}^{\infty} \frac{P_0(T_p, \tau) G(\tau)}{\tau} \exp\left[-\frac{1}{b} \int_{T_0}^T \frac{dT'}{\tau(T')}\right] d\tau \quad (7)$$

The determination of $G(\tau)$ is not straightforward. A relaxation mechanism with distribution in both W and τ_0 requires an over-parameterized fitting procedure. On the other hand Eq. (4) shows that for a particular peak (with b and T_m known) any change in W will result in a change in τ_0 . Consequently, any analysis that assumes one parameter to be fixed (could be either W or τ_0) and one parameter variable, will lead to inappropriate results.

3. Experimental

We will use the experimental results obtained for the parameters W , τ_0 and T_m on biaxially drawn sheets of polyethylene terephthalate (PET) of 8 μm thickness to generate realistic data for the analysis and simulations. Disk specimens of 40 mm diameter were provided with two measuring electrodes of 25 mm diameter by evaporating silver at a pressure of 2×10^{-5} Torr. Measurements of FPTSDC, at a chamber pressure of 5×10^{-3} Torr, were made for different polarization temperatures. The electrical current in the circuit was measured with an electrometer (Keithley 617) coupled to a PC for data acquisition and analysis. The fractional polarization peaks were recorded with the following polarization conditions. Isothermal polarization time $t_p = 5$ min at selected temperatures T_p , followed by cooling to $(T_p - 5)$ K in 2 min keeping the electric field on. At $(T_p - 5)$ K the electric field was removed and the sample was kept for

$t_s = 2$ min in short circuit. Further the sample was cooled down (0.5 K/s) at approximately $(T_p - 50)$ K in short circuit. The temperature of measurements was varied over the range 60 to 320 K. A Cu-Constantan thermocouple, mounted in the sample holder and adjacent to the film, allowed the temperature measurement with a precision of 0.1 K. The sample temperature was maintained constant to within ± 0.1 K. The current measurements were taken at a heating rate $b = 5$ K/min. The linear heating rate was controlled by the computer and the difference between the prescribed and the measured temperature was always lower than 0.3 K. Prior to every TSDC measurements the sample was heated to 463 K to anneal out any thermal and electrical pre-history. It was kept there for a sufficient long time to insure that the discharging current becomes insignificant. It was then slowly cooled down to room temperature. This way, the sample is conditioned so that prior history does not influence the coming measurements, therefore avoiding impairing the repeatability and interpretation of results [4,8]. The degree of crystallinity determined by X-ray diffraction was 68 % and was found to be practically unchanged by the conditioning procedure.

4. Results and discussion

Fig. 1 displays the FP spectra for samples polarized at $T_p = 120$ K under a field $E_p = 30$ MV m⁻¹. The full curve represents best fit lines to Eq. (1). The values of the fitting parameters are presented in the inset. In Table 1 are presented T_m and W for samples polarized at different temperatures from 80 to 180 K. Using Eqs. (4) and (5) the values for τ_0 and for $\tau(T_m)$ were determined.

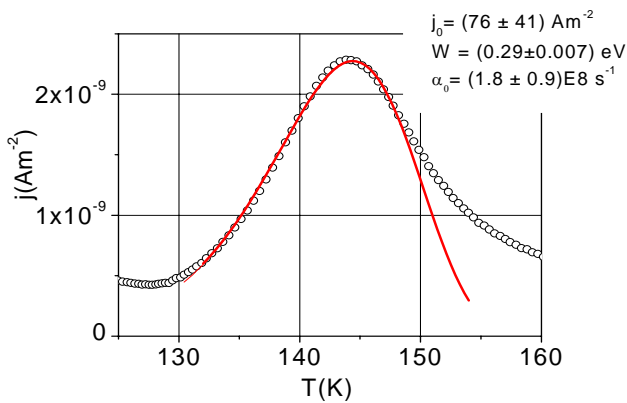


Fig. 1. Thermally stimulated discharge currents (fractional polarization) for PET samples polarized under a field of 30 MV m⁻¹ at 120 K. The full curve represents the best fit line obtained using Eq. (1).

Table 1. The results obtained for W by a fast estimation using the initial rise method. The values for τ_0 are obtained from Eq. (4) and the values for $\tau(T_m)$ are obtained from Eq. (5).

T_p K	T_m K	W eV	τ_0 s	$\tau(T_m)$ s
80	96.4	0.07	0.05	145
100	117.1	0.12	0.006	120
125	144.0	0.12	0.007	163
140	157.9	0.15	0.002	169
160	176.5	0.15	0.009	210
180	197.7	0.20	0.002	205

Equation (4) can be solved numerically to obtain a three-dimensional plot of $\tau_0(W, T_m)$. For the parameters, we will use values very close to those determined from the experimental data and presented in Table 1. The results obtained for 0.1 eV $< W < 0.6$ eV, 150 K $< T < 250$ K and $b = 1$ K/min or 5 K/min are presented in Figs. 2 and 3, respectively. Figs. 2 and 3 imply that, for narrow intervals of the input parameters, there are approximately linear relationships between W , T_m and $\log \tau_0$. It can be observed that W decreases as τ_0 increases. Assuming that τ_0 is constant, Eq. (4) implies a relationship between W and T_m . Conversely, assuming that τ_0 is a function of W , Eq. (2) implies either (i) a relationship between W and T_m or (ii) a relationship between τ_0 and T_m .

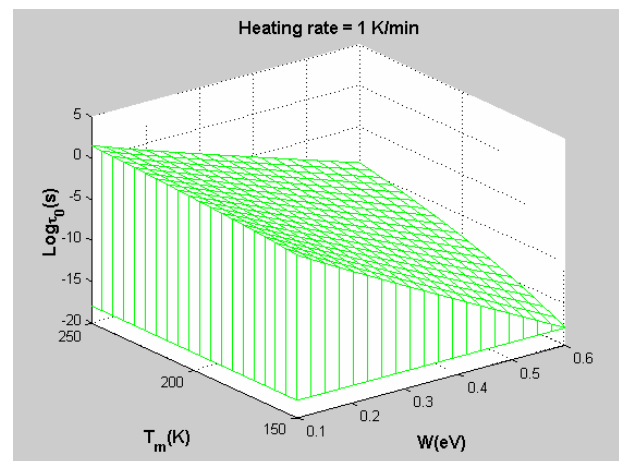


Fig. 2. Three-dimensional plot of the expected values for $\tau_0(W, T_m)$ obtained by numerical solving of Eq. (4) for a heating rate of 1 K/min.

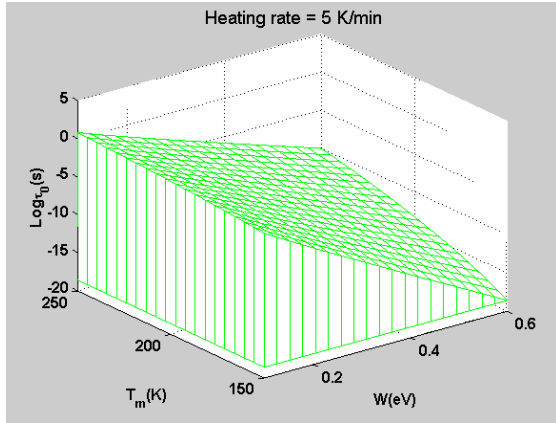


Fig. 3. Three-dimensional plot of the expected values for $\tau_0(W, T_m)$ obtained by numerical solving of Eq. (4) for a heating rate of 5 K/min.

The variation of $\tau(W, T_m)$ for the same parameter intervals as above is presented in Figs. 4 and 5. The relaxation time varies in a narrow interval suggesting a narrow distribution. The variation is smaller for a high heating rate. Consequently, a low heating rate is recommended in order to study a possible distribution.

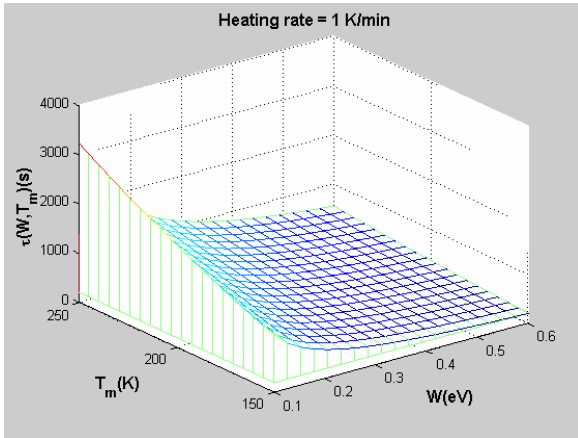


Fig. 4. Three-dimensional plot of the expected values for $\tau(W, T_m)$ obtained by numerical solving of Eq. (5) for a heating rate of 1 K/min.

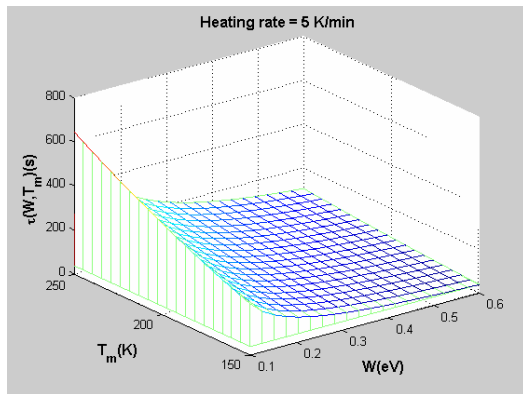


Fig. 5. Three-dimensional plot of the expected values for $\tau(W, T_m)$ obtained by numerical solving of Eq. (5) for a heating rate of 5 K/min.

General relationships involving any two of the relaxation parameters and the ranges in which these parameters vary, may be obtained using Eq. (4) for a row i of elementary peaks. In this case T_m becomes a variable parameter T_{mi} , meaning that

a) all elementary currents (characterised by different values of W_i , τ_{0i} and T_{mi} for b constant) that can contribute to a measured current are considered. This is especially the case when a peak is decomposed into a sum of elementary peaks;

b) we can consider all possible sets of parameters W_i , τ_{0i} and T_{mi} for a constant b that define an elementary peak, and assume that any of these parameters can be affected by some uncertainty. This is especially the case when we try to determine whether there is a distribution in the values of the relaxation parameters obtained from a peak, keeping in mind the uncertainties affecting the relaxation parameters. In fact, the uncertainties affecting the parameters determine a broadening of the theoretical peak.

c) we can consider all parameters W_i , τ_{0i} and T_{mi} obtained for a peak when b is a variable parameter. As can be observed from the simulations in Figs. 2 to 5 the values obtained for the relaxation parameters are dependent on the heating rate.

From Eq. (4) results that for a given heating rate, for example 1 K/min, and for every elementary peak, e.g. the one in Fig. 1, there are several possible solutions for the pair W_i , τ_{0i} . We will consider the peak at $T_{mi} = 144$ K and let us try to solve Eq. (4) to find some possible solutions for W_i and τ_{0i} . We will assume that τ_{0i} varies in a large range 10^{-6} to 10^{-16} s. The results for W_i are presented in Table 2, column 2. From the data in Table 2 it can be observed that if we impose a value for one of the parameter the value obtained for the other parameter can be very different from its actual value. The value obtained for $\tau_{0i} = 10^{-6}$ s is close to the one determined from experimental data.

An estimate of the possible uncertainties for relaxation parameters can be obtained using Eq. (4). By taking the logarithm of Eq. (4), differentiating and replacing the infinitesimal variations d with the finite variations Δ , it results

$$\frac{\Delta \tau_{0i}}{\tau_{0i}} + \left(1 + \frac{W_i}{kT_{mi}}\right) \frac{\Delta W_i}{W_i} - \left(2 + \frac{W_i}{kT_{mi}}\right) \frac{\Delta T_{mi}}{T_{mi}} = 0 \quad (8)$$

Equation (8) can be used to obtain useful relationships between the relaxation parameters τ_{0i} , T_i and W_i and the finite variations of these parameters $\Delta \tau_{0i}$, ΔT_{mi} and ΔW_i , which in turn can be used as approximations for the uncertainty intervals of τ_{0i} , T_{mi} and W_i .

The lack of periodicity in random solids, such as semicrystalline PET, implies that the constituent atoms, molecules, or molecular subunits are located in statistically different environments. We will assume that the thermal fluctuation gives rise to a Gauss-type distribution function for W . It is known that at any temperature there is a thermal energy kT . Consequently, we will assume that at any temperature T the uncertainty interval ΔW is at least

equal with $\pm kT$. Because this interval represents the minimum uncertainty that influences any determination of W , we will call it the *natural* uncertainty interval in activation energy [20]. Equation (8) will be further discussed in three particular cases to follow.

In the first case we will assume that τ_{0i} is constant ($\Delta\tau_{0i} = 0$). From Eq. (8) it results

$$\Delta T_{mi} = T_{mi} \left(1 + \frac{kT_{mi}}{W_i}\right) \left(2 + \frac{W_i}{kT_{mi}}\right)^{-1} \quad (9)$$

Equation (9) gives the minimum range for ΔT_{mi} (because ΔW_i has the minimum value) as a function of T_{mi} , W_i and implicitly of τ_{0i} . Using the values in column 2 (Table 2) for W_i (determined for a particular case $T_{mi} = 144$ K) and Eq. (9), the values for ΔT_{mi} may be calculated. The calculated values are reported in Table 2, column 3. In Table 1 are reported the values calculated using the results determined from the experimental thermograms. The quantity ΔT_{mi} represents the temperature range in which an elementary peak can be located, i.e. the peak can be located in a certain temperature interval for different choices of the relaxation parameters. We can read this result in a reverse way: all the peaks having the temperature of the maximum current in an interval $\cong \Delta T_{mi}$ will provide similar relaxation parameters. The immediate useful implication of this estimation is from an experimental point of view: if the window polarization technique is to be used to separate the elementary contribution, it is useless to chose the experimental conditions so that for two successive elementary peaks $T_{mj} - T_{mi} < \Delta T_{mi}$. Similar conclusions are also valid for the case when a global peak is decomposed (by fitting) into a number of elementary components. At the same time from the data in Tables 1 and 2 it can be concluded that the elementary peaks obtained by windowing polarization need not be uniformly spaced in temperature. For the presented conditions, if the activation energy increases from 0.2 to 0.5 eV or the pre-exponential factor decreases from 10^{-6} to 10^{-16} s, ΔT_i decreases from 6.6 to 3.47 K. For the situation when $\Delta W_i > kT_{mi}$, meaning that we have a variance (standard deviation) in activation energies, ΔT_{mi} increases which, in turn, results in the elementary peaks being less accurately resolved.

In the second case W_i is assumed constant but ΔW_i is equal to the thermal energy kT_{mi} . In a worst-case estimate of error, that is, assuming that $\Delta T_{mi} = 1$ K is the lower limit for ΔT_{mi} (we can not expect to have a better separation of the elemental peaks) we have

$$\frac{\Delta\tau_{0i}}{\tau_{0i}} = \left(1 + \frac{kT_{mi}}{W_i}\right) + \left(2 + \frac{W_i}{kT_{mi}}\right) \frac{1}{T_{mi}} \quad (10)$$

Using the data in Tables 1 and 2, the values of $\Delta\tau_{0i}/\tau_{0i}$ can be computed. The results are listed in Table 2, column 4. It can be observed that $\Delta\tau_{0i}/\tau_{0i}$ is very close to unity. Because in Eq. (10) we have used a good resolution for ΔT_{mi} (1 K) the interval $\Delta\tau_{0i} \cong \tau_{0i}$ represents the higher limit for the resolution of determining the pre-exponential factor. This

resolution is higher than the one reported in literature $\Delta\alpha_0/\alpha_0 = 5.8$ [2], as expected. It can be concluded that by assuming $W_i = \text{constant}$, and under $\Delta W_i = kT_{mi}$, the width of the peak increases as compared to the width determined in the $\tau_{0i} = \text{constant}$ case; a broad experimental peak indicates that there is, at minimum, a distribution in the activation energies.

The third case is $T_{mi} = \text{constant}$ and applies to situations when there are relaxation entities having different values for W_i and τ_{0i} but the current peak appears at the same temperature T_{mi} . In this case Eq. (8) becomes

$$\frac{\Delta\tau_{0i}}{\tau_{0i}} = -\left(1 + \frac{W_i}{kT_{mi}}\right) \frac{\Delta W_i}{W_i} \quad (11)$$

To obtain the natural uncertainty interval for τ_{0i} we will assume again that $\Delta W_i = kT_{mi}$. Using the values for W_i in Table 1, we can determine the values of $\Delta\tau_{0i}$ from Eq. (11). The results are reported in Table 2, column 5. It can be observed that the ratio $\Delta\tau_{0i}/\tau_{0i}$ is very close to unity. In fact, the values for $\Delta\tau_{0i}/\tau_{0i}$ in Table 2 do not differ significantly.

Table 2. The values obtained for the activation energy W_i by numerical solving of Eq. (4) for $b = 5/60$ K/s, $T_m = 144$ K and different values for the pre-exponential factor τ_{0i} in the range 10^{-16} to 10^{-6} s. There are reported also the natural uncertainty intervals ΔT_{mi} corresponding to the natural uncertainty interval for the activation energy and the ratio between the natural uncertainty interval $\Delta\tau_{0i}$ and τ_{0i} in the case when there is no distribution in τ_{0i} .

τ_{0i} s	W_i Eq. 4 eV	ΔT_{mi} Eq. 9 K	$\Delta\tau_{0i}/\tau_{0i}$ Eq. 10	$\Delta\tau_{0i}/\tau_{0i}$ Eq. 11
10^{-16}	0.53	3.47	1.32	1.02
10^{-14}	0.44	3.9	1.29	1.02
10^{-12}	0.39	4.4	1.26	1.03
10^{-10}	0.34	5.1	1.24	1.04
10^{-8}	0.28	5.8	1.23	1.04
10^{-6}	0.21	6.6	1.21	1.05

Further we will apply the above results for the experimental thermogram on PET displayed in Fig. 1. The uncertainty in W for the best fit line in Fig. 1 is 0.007 eV meaning that the fitting process is very good; but there is no physical reason to consider that $\Delta W < kT_m$ ($kT_m = 0.014$ eV). The fact that the experimental data in Fig. 1 are very well fitted for a unique value of W does not necessarily mean that there exists no distribution in W . There could be a distribution in the activation energies and the important question is how wide this distribution is. In this respect it is useful to simulate thermograms for small parameter variations. Fig. 6 presents two simulated thermograms for $W = 0.27$ eV and for $W = 0.31$ eV (meaning that the variation of W is slightly higher than the thermal energy $kT_m = 0.014$ eV), the other two parameters having the same values as above. Fig. 6 suggests that by choosing ΔW in

narrow limits, the peak shifts by about 10 K. This result is in good agreement with the estimations presented in Table 2.

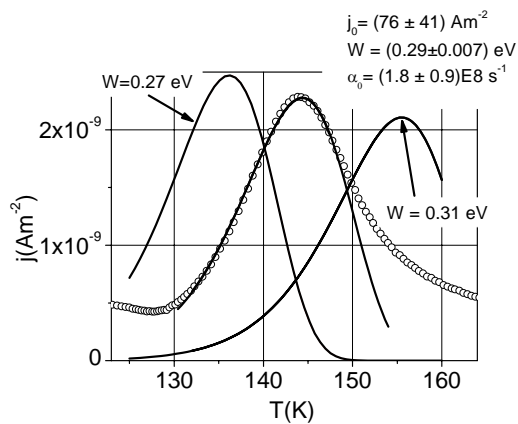


Fig. 6. TSDC (circles), the best fit of the data and two simulated curves assuming W as variable parameter ($\alpha_0 = \text{constant}$).

In Fig. 7 we present two simulated thermograms (assuming α_0 a variable parameter) and the experimental data from Fig. 1. It can be observed that the peak changes significantly for a small variation in α_0 . It is to be underlined that the variation in α_0 is lower than the natural uncertainty interval $\Delta\tau_0$ reported in Table 2.

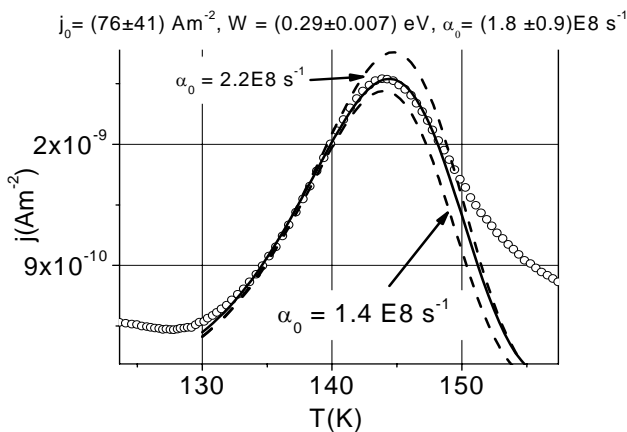


Fig. 7. TSDC (circles), the best fit of the data and two simulated curves assuming α_0 as variable parameter ($W = \text{constant}$).

Fig. 8 captures the experimental data from Fig. 1, the best fit line using Eq. (6), and two simulated thermograms (assuming P_0 constant). It can be observed that, since W increases from 0.28 to 0.32 eV, the maximum current increases but does not shift due to the fact that T_m is kept a constant parameter. The results in Fig. 8 are in very good agreement with a resolving limit of $W_2/W_1 = 1.09$ reported in the literature [2]. There is a very good agreement between the values obtained for W using Eqs. (1) and (5)

and qualitative agreement between the values obtained for τ_0 . As long as T_m is known it is advisable to use Eq. (6) for fittings, simulations or to decompose a global current into a sum of elementary peaks.

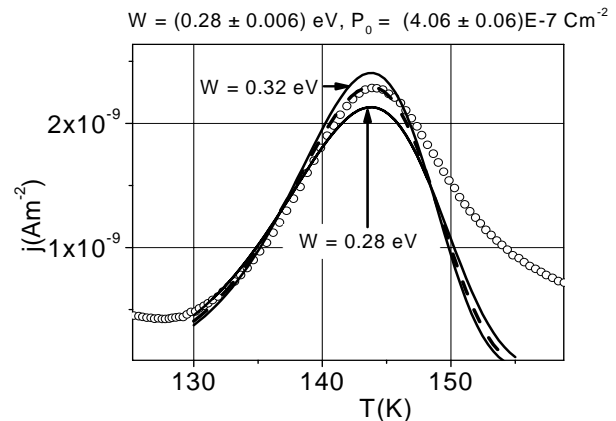


Fig. 8. TSDC (circles), the best fit curve to Eq. (5) (dashed line) and two simulated curves. W increases from 0.28 to 0.32 eV and the value calculated for τ_0 , using Eq. (4), decreases from 1.2×10^{-8} to 4.1×10^{-10} s.

From the data in Figs. 6 and 7, it can be concluded that very small variations in W or τ_0 can be discriminated when only one parameter varies. In reality both parameters change and the data in Fig. 8 show that variations lower than the *natural* uncertainty intervals can not be properly identified.

5. Conclusions

The values obtained for W or τ_0 by fitting the experimental data to the analytical equation of the current vary significantly for the case when one parameter is assumed constant. An equation is proposed to compute the relaxation parameters taking advantage of the fact that W and τ_0 change simultaneously. The relationship between W , τ_0 , b and T_m can be used to obtain information about the distribution of relaxation parameters and the temperature range corresponding to elementary peaks. As long as the experimental current can be fitted with an elementary peak and the uncertainties affecting the relaxation parameters are low, i.e. $\Delta W \cong kT_m$ and $\Delta\tau_0 \cong \tau_0$, the distribution functions cannot be determined accurately. The adjustable parameters are affected by uncertainties that progressively increase as the number of elementary currents, considered in a fitting step, increases.

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