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A study on a numerical solution of the transport equations for thin insulating films with two blocking electrodes

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Abstract. The transport equations in the case of thin insulating films, sandwiched between two blocking electrodes, have been solved numerically. The mobility of the charge is considered to be field and temperature dependent and it is related to the time dependence of the electric field. A trial function for this dependence is introduced. The time evolution of the charge and voltage distribution within the thin film are obtained and the resulting transient current is compared with experimental results.

1. Introduction

The behaviour of dielectrics under the application of steady voltage depends mainly on the kind of contacts between the metal electrodes and the dielectric material. For blocking contacts no charge crosses the electrodes from the dielectric material to the metal electrode and vice versa. Therefore, the observed transient current will be due to the polarisation of the material, which may be due either to the orientation of electric dipoles or to hopping positive and/or negative charges or both.

Experimental results of the transient electrical conductivity of thin alkali halide films (Georga and Pisanias 1983, 1984), sandwiched between aluminium electrodes, show that the current decreases with time according to the general relation,

$$I(t) = \alpha t^{-m} \tag{1}$$

where α and m are constants, which depend mainly on the material. This relation is the universal dielectric response in the time domain (Jonscher 1983, Ngai and White 1979, Scher and Montroll 1975). Other factors, which affect slightly the values of the constants α and m are the applied voltage, the temperature and the film thickness. It should be pointed out, that thin alkali halide films formed by thermal evaporation are highly disordered (Smart 1971) and conductivity is achieved through the hopping mechanism. Recent results of AC conductivity on KI thin films (Pisanias *et al* 1990) have shown, that the observed transient conductivity is due to the superposition of two mechanisms which are identified as due to anions and to the ionisation of trapped electrons.

The problem of evolution of the internal electric field for blocking electrodes has first been treated by Macdonald (1959), who has solved the transport equations under steady state conditions for cases in which only the anion is considered to be the mobile charge.

The aim of the present work is to incorporate in the solution of the one-dimensional transport equations the time dependence of the mobility through the time dependence of the electric field. The resulting voltage and charge distributions and their time dependence allow the determination of the transient current and its comparison with experimental data.

2. Theoretical considerations

In order to write down the proper transport equations it is necessary to describe first the system under consideration. It is assumed that a dielectric material of thickness L in the x direction is placed between two parallel metallic plates, which are normal to the x axis. The work function b_m of the electrodes is larger than the work function b_i of the dielectric. Therefore they are blocking electrodes and no charges can cross the electrodes in either direction.

2.1. Governing equations and numerical solution

The mobile charges contained in the insulating thin film due to the presence of structural defects are considered to be negative only and any recombination with positive charges is practically zero. With these assumptions the time-dependent one-dimensional governing equations are

$$-\varepsilon_s \frac{\partial^2 \psi(x, t)}{\partial x^2} = \rho(x, t) \tag{2}$$

$$J(t) = J_n(x, t) - \varepsilon_s \frac{\partial}{\partial t}\left(\frac{\partial \psi(x, t)}{\partial x}\right) \tag{3}$$

$$J_n(x, t) = -q\mu_n n(x, t) \frac{\partial \psi(x, t)}{\partial x} + qD_n \frac{\partial n(x, t)}{\partial x} \tag{4}$$

$$\frac{\partial n(x, t)}{\partial t} = \frac{1}{q} \frac{\partial J_n(x, t)}{\partial x} \tag{5}$$

where the symbols used in the above equations and their physical meaning are listed in table 1.

The above set of equations, rewritten in a normalised form with normalisation coefficients for each variable given in table 1 (De Mari 1968), are reduced to the following two equations:

$$\frac{\partial^2 \psi(x, t)}{\partial x^2} = n(x, t) - p \tag{6}$$

$$\frac{\partial n(x, t)}{\partial t} = -\mu_n n^2(x, t) + \mu_n n(x, t)p - \mu_n \frac{\partial \psi(x, t)}{\partial x} \frac{\partial n(x, t)}{\partial x} + \mu_n \frac{\partial^2 n(x, t)}{\partial x^2}. \tag{7}$$

Uniform charge carrier distribution and linear potential drop throughout the sample are the initial conditions. These, together with equations (6) and (7) as well as the boundary conditions of zero current at the electrodes and constant applied potential, completely specify the problem.

Table 1. List of symbols.

Symbol	Physical quantity	Normalisation factor
ψ	potential	$V_t = kT/q$
n	anion concentration	N
p	cation concentration	N
ρ	total charge density	N
J_n	anion current density	$-qD_0N/L_D$
$J(t)$	total current density	$-qD_0N/L_D$
$I(t)$	total current	$-qD_0NL_D$
μ_n	anion mobility	D_0/V_t
ε_s	permittivity	ε_s
L	film thickness	L_D
V	applied voltage	V_t
x	distance from the anode	$L_D = \sqrt{(\varepsilon_s V_t/qN)}$
t	time	L_D^2/D_0
D_n	diffusion coefficient	D_0
D_0	unit of carrier diffusion coefficient	$1\ \mathrm{m^2\,s^{-1}}$

The above set of equations (6) and (7) is solved numerically by applying a finite difference method, following well established techniques (Smith 1971, Press *et al* 1987). With a first-order accuracy in time and second-order accuracy in space this method leads to an optimum space mesh of 180 points throughout the sample under considera- tion. Timestep Δt is calculated according to the convergency condition

$$\Delta t \leqslant \frac{\Delta x^2}{\mu_n} \frac{1}{3\Delta x^2 - |\Delta\psi_i|\Delta x + 2} \tag{8}$$

where i is the index of the uniform space-mesh point (Smith 1971).

3. Results and discussion

The method, which was previously analysed, has been applied to MIM (metal–insulator– metal) structures of thin insulating films. In particular the concentration N is equal to $10^{23}\ \mathrm{m^{-3}}$ and the dielectric constant ε is equal to 2.2. These values are comparable to the corresponding values of alkali halides. Thus direct comparison with the experi- mental results by Pisanias *et al* (1990), as mentioned in the introduction, can be made. The thickness of the film is greater than twice the depth of the depletion region λ_0 (Simmons 1971) and specifically it is equal to 2000 Å.

The results, which are presented in this section, are strongly dependent on the anion mobility, which enters the equations describing the system under consideration. Although it is well known that mobility is field and temperature dependent, constant values have often been used in similar works and have proved to be a good approxima- tion (Kitani *et al* 1984, Mizutani *et al* 1981).

In our case however, constant mobility has proved to be a rather poor assumption. This is clearly shown in figure 1 where the large deviation (*a*) from the experimental results (*c*) is obvious. Empirical formulae for field- and temperature-dependent mobil- ity are reported in the literature (Gill 1972, Yoshino 1978) as well as formulae derived theoretically with certain assumptions for the transport mechanisms (Bryskin and Gol'tsev 1988, Saglam and Friedman 1975).
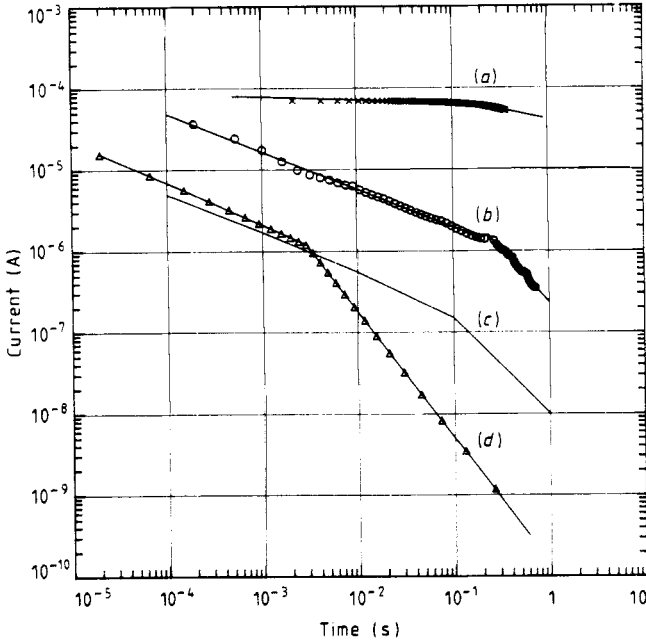
Figure 1. Transient current for different mobilities. (a) μ = constant; (b) $\mu = \mu_0 \exp(k|E - E_0|^l)$; ($c$) experimental results (Pisanias *et al* 1990); (d) $\mu = \alpha t^{-m}$. Applied voltage 0.9 V. Film thickness 2000 Å.

None of the existing formulae in the literature to the best of our knowledge could be used satisfactorily in this work. Therefore, as a starting point, the time dependence of the mobility in dispersive transport was considered. In this case it has been shown that the mobility may be expressed through the relation (Orenstein and Kastner 1979, Hamill 1982)

$$\mu_n = \begin{cases} bt^{-n} & t < t_\tau \\ ct^{-m} & t > t_\tau \end{cases} \qquad\qquad (9a) \\ (9b)$$

where t_τ corresponds to the transit time of the fastest carriers and b, c, n and m are parameters. The sum of the exponents n and m should be equal to 2 (Scher and Montroll 1975). For known values of b, t_τ, n and m, the value of the parameter c is obtained through the relation

$$c = bt_\tau^{2(1-n)}. \qquad\qquad (10)$$

The use of relations 9(a) and 9(b) in the numerical solution requires the knowledge of the transit time t_τ. This transit time could be deduced from the slight change of the slope of the current against time in a log–log presentation when only relation 9(a) was used. By forcing the mobility to take values according to the relation 9(b) for $t > t_\tau$, the solution of the transport equations leads to a time dependence of the current, which is improved (figure 1, plot (d)) and tends to the experimentally obtained behaviour of the current (c).

However, our efforts were focused on using a formula for the mobility as a function of the electric field, in which the time dependence would be implicit through the time dependence of the electric field. The best results (figure 1, plot (b)) were obtained

with the trial function

$$\mu(E) = \mu_0 \exp(k|E(x, t) - E(x, 0)|^l) \tag{11}$$

where μ_0, k and l are parameters, which are evaluated through a fitting program to the mobility results obtained from relation 9(a). The value of b in the relation 9(a) in the present study has been chosen in such a way so that the resulting mobility at extremely short times ($\simeq 10^{-6}$ s) is equal to the mobility, which results from the Nernst–Einstein relation for diffusion coefficients, taken from the work by Laurent and Benard (1957). It is interesting to point out that the results of the fitting lead to a value for l equal to 0.4, which is independent of the applied voltage and the temperature of the thin film.

By using the mobility expression given by relation (11) the time dependence of the current was determined for two cases: (i) as a function of the temperature for a given applied voltage and (ii) as a function of the applied voltage for a given temperature. The results are shown in figures 2 and 3 respectively. The plots (b) and (c) in figure 2 are shifted by one decade for clarity reasons. The effect of increasing temperature is not only the increase of the current but also a shift to lower values of the transit time t_r. Such a shift has been observed by Georga and Pisanias (1984), Hamill (1982) and Pfister and Scher (1977). On the other hand, by increasing the applied voltage for a given temperature the current increases and a shift of the transit time t_r to lower values is observed. Such a shift has been reported by Pfister and Scher (1977).

Furthermore, the charge and voltage distribution within the thin film are presented in figures 4 and 5, from which it is concluded that the charge distribution develops in such a way that the potential drop near the cathode becomes steeper as time proceeds; that is, the internal field becomes higher. This is clearly shown in figures 6 and 7, where the space-time dependences of the charge and the electric field distributions are
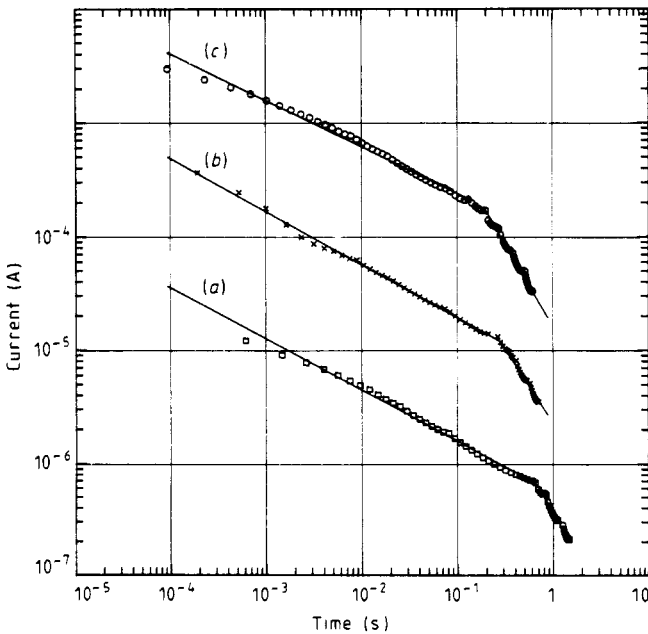
Figure 2. Transient current at different temperatures for mobility given by relation (11). (a) $T = 180$ K; (b) $T = 290$ K; (c) $T = 360$ K.
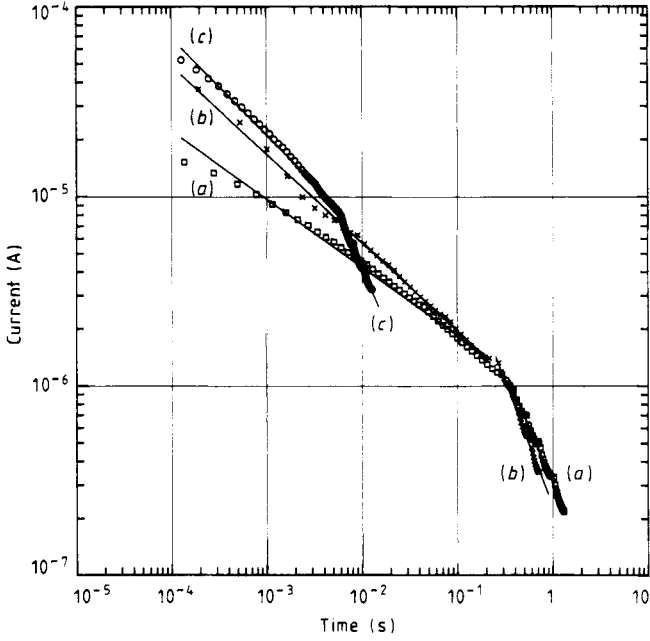
Figure 3. Transient current for different applied voltages and for mobility given by relation (11). (*a*) $V = 0.5$ V; (*b*) $V = 0.9$ V; (*c*) $V = 2.0$ V.
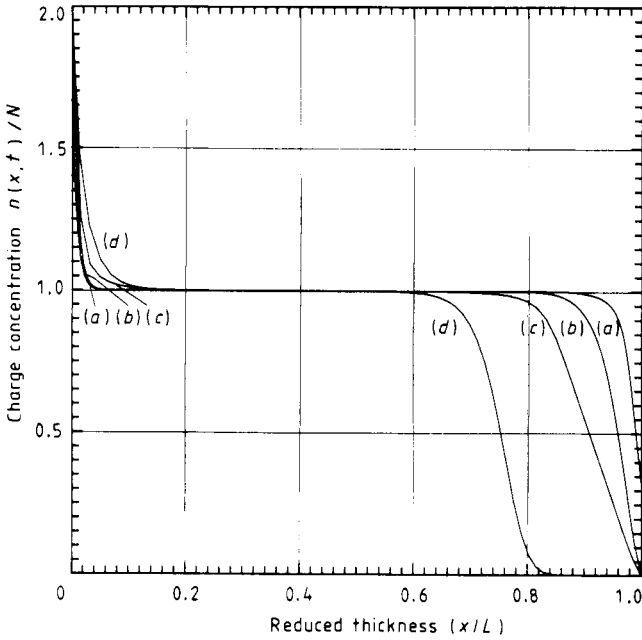

Figure 4. Charge distribution at different instants. (*a*) $t = 3.8 \times 10^{-1}$ s; (*b*) $t = 8.8 \times 10^{-1}$ s; (*c*) $t = 10$ s; (*d*) steady state.
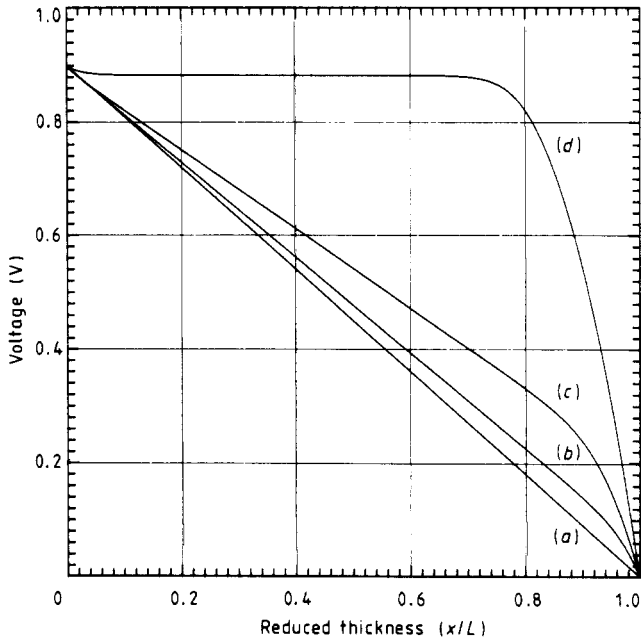
Figure 5. Voltage distribution at different instants. (*a*) $t = 0$ s; (*b*) $t = 8.8 \times 10^{-1}$ s; (*c*) $t = 10$ s; (*d*) steady state.
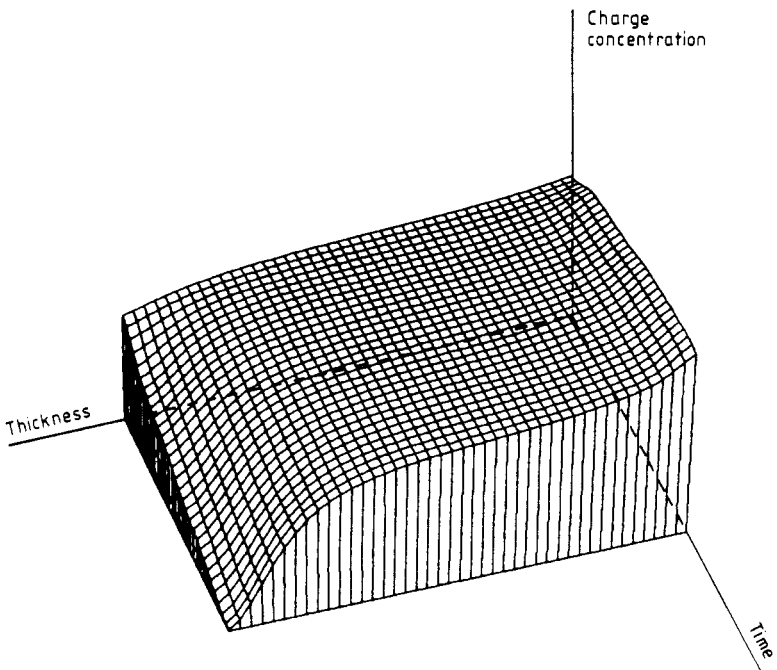

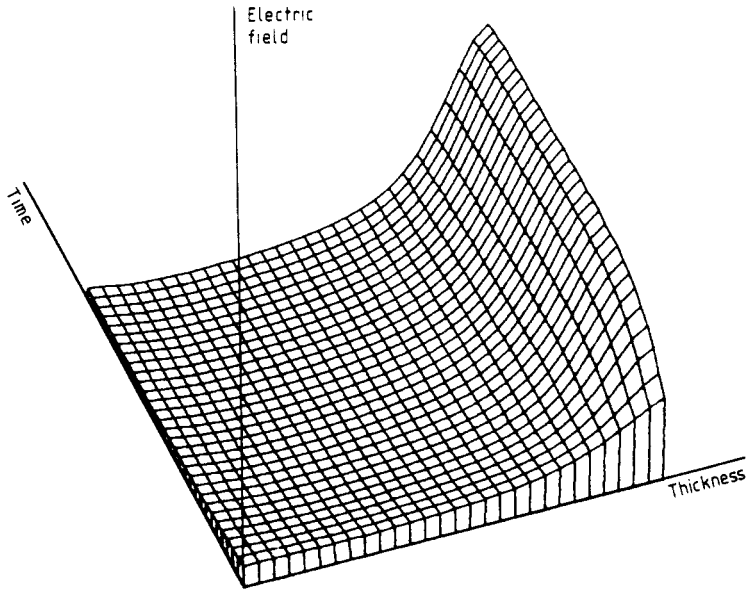**Figure 6.** Space-time dependence of the charge concentration.

Figure 7. Space-time dependence of the electric field.

presented respectively. This high internal field may lead to the onset of other effects, like the Poole–Frenkel effect (Frenkel 1938, Hill 1971), which will contribute to the observed current. This effect has not been taken into consideration in this study, but it is under investigation and will be reported in the future. The steady state charge and voltage distributions are presented as plots (*d*) in figures 4 and 5 respectively.

4. Conclusions

The transport equations have been solved numerically for the determination of the voltage and charge distribution within the sample and the time dependence of the resulting current under a steady bias voltage. By comparing the numerically obtained transient current with experimental results, it is concluded that the field dependence of the mobility for the anions, which is proposed in this work, is satisfactory. The application of this method for the case of two mobile charges is now under investigation.

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