T. A. E. C.
ÇEKMECE NUCLEAR RESEARCH AND TRAINING CENTER
İSTANBUL - TURKEY

ÇNAEM - R - 112

HIGH - FIELD TRANSPORT PROPERTIES OF ALUMINUM - EMBEDDED ALUMINUM OXIDE FILMS

By

H. Birey

P. K. 1, Hava Alanı, İstanbul, Turkey

1973

High-field transport properties of aluminum-embedded aluminum oxide films

Hülyâ Birey

Physics Department, Çekmece Nuclear Research and Training Center, P.K. 1 Hava Alani, Istanbul, Turkey
(Received 7 March 1973; in final form 2 July 1973)

Current-voltage characteristics of aluminum-embedded aluminum oxide thin films with Al or Au electrodes, between 150–1000 Å in thickness, prepared by thermic evaporation of pure aluminum in partial air pressure are studied. I-V characteristics of these films showed metallic conductivity, switching, and memory effects different than those observed in amorphous materials, and metal-oxide-metal diode characteristics as the amount of metallic aluminum within the oxide is decreased in respective samples. The switching and memory effects are found to be independent of oxide thickness and electrode material.

In recent years, charge storage phenomena[1,2] in electron-beam-evaporated aluminum oxide films, negative resistance characteristics,[3,4] and swiching phenomena[5,6] in anodic oxide films, tunneling in thermally grown metal-Al_2O_3-metal sandwiches,[7] and dielectric properties[8–10] of aluminum oxide films deposited by evaporation in oxygen have been studied.

In this letter, static current-voltage (I-V) characteristics of aluminum-embedded aluminum oxide thin films, which showed switching and memory characteristics, are studied. These films are prepared by thermic evaporation of pure aluminum in different air (or oxygen) deposition pressures and are sandwiched between aluminum or gold electrodes that are also prepared by thermic evaporation.

The samples are prepared by first evaporating 2-mm strips of Al or Au films, 2000–4000 Å thick, onto carefully cleaned microscope slides. 4-mm-wide oxide films are next evaporated onto these lower electrodes. The thickness of the oxide (150–1000 Å) depends on the amount of aluminum evaporated. The concentration of the metallic aluminum within the oxide is controlled by the deposition pressure, the evaporation rate, and the distance of the substrate from the source. Thus, it is possible to change the amount of aluminum within the oxide. Al or Au upper electrodes are then evaporated across the oxide through a suitable mask. The effective areas of the sandwiches prepared in this manner are 4 mm^2.

The experimental setup used to study I-V characteristics of the samples is shown in Fig. 1. A variable-slope ramp generator supplies the necessary driving voltage. The total series load resistance R_L is the sum of the external load resistance R_s, which is variable from 47 Ω to 470 kΩ, and the current sampling resistance R_i, which is fixed at 100 Ω. Voltage and current outputs are connected to the X and Y inputs of a 7005 B Hewlett-Packard X-Y recorder. Since the current passing through the sample is very small, shunting of the sample is prevented by a source follower with 100-MΩ input impedance. An oscilloscope, Tektronix 555, is used to check these I-V plots for possible adverse effects during switching due to the mechanical time response of the recorder.

Optical measurements are made on separate samples simultaneously prepared during oxide formation. Transmittance and reflectance of the films are measured using 6328-Å-wavelength radiation of a He-Ne gas laser. Total light scattering is obtained through integration of the angular scattering of the same laser light. Wavelength dependence of scattering is not investigated. The sample thickness measurements are made by an optical interferometer which produced Fizeau fringes of equal thickness.

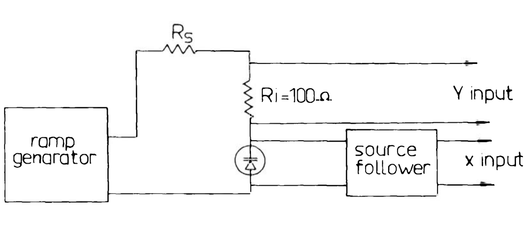

FIG. 1. Schematic diagram of the electrical circuit used to study I-V characteristics of sample S. Series load resistance is $R_L = R_s + R_i$. X and Y inputs are connected either to an X-Y recorder or to an oscilloscope.

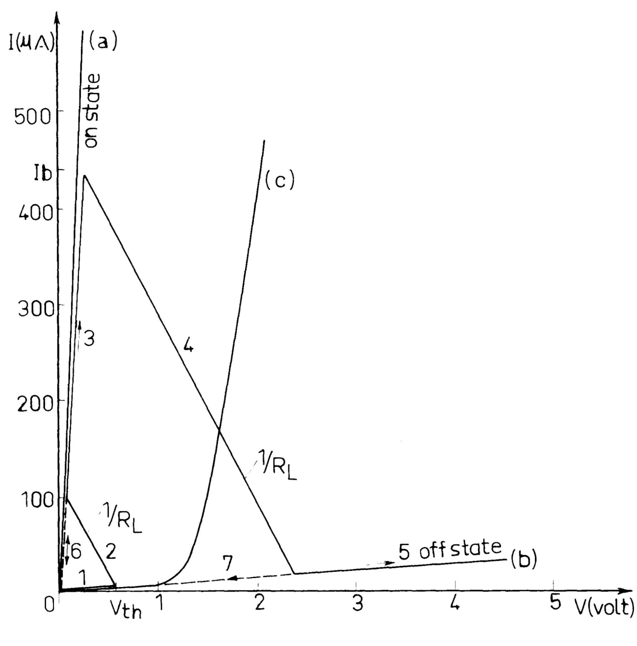

FIG. 2. I-V characteristics of aluminum-embedded aluminum oxide sandwiches: (a) metallic conductivity, (b) switching and memory effects, (c) metal-oxide-metal characteristics. Sample showing switching and memory effects follows curves 1–5. V_{th}, threshold voltage; I_b, blocking current. Conductance of the on state is directly proportional to I_b.

TABLE I. List of typical optical and electrical properties of Au-oxide-Au sandwiches with increasing deposition pressures.

Sample No.	P (torr)	d_{in} (Å)	R (%)	T (%)	A_m (%)	$k = \frac{d(\text{oxide})}{d(\text{metal})}$	R_{ps} (Ω)	Sample properties
1	10^{-5} (air)	500	92	0	8	1	~1	Pure metal; metallic conductivity
2	Increasing Pressure ↓	505	35	37	28	1.01	500	Large optical absorption; high electrical conductivity
3		510	14.4	59	26.6	1.02	$(0.2-0.05) \times 10^6$	Switching but erratic
4		520	14.2	73.9	11.9	1.04	1.5×10^6	Excellent switching and memory effects
5		550	11.9	78	10.1	1.10	2.8×10^6	Both switching effects and oxide I-V properties
6	4×10^{-3} (air)	1070	7.78	92.2	0.02	2.14	8×10^6	No optical absorption; oxide I-V properties
7	4×10^{-3} (oxygen)	1110	7.5	92.5	0	2.21	10×10^6	No optical absorption; oxide I-V properties

The switching properties of these devices did not depend on film thickness regardless of the electrode material, but sandwiches with Al electrodes showed erratic electrical behavior. Therefore, only Au electrodes are used, and equal amounts of metallic aluminum are evaporated during film deposition for the rest of the experiments.

The deposition pressure p, interferometric thickness d_{in}, reflectance R, transmittance T, total light absorption $A_t = A_m + A_s$ (where A_m is the metallic absorption and A_s is the absorption due to scattering), augmentation of the thickness k [11] and the initial preswitching low-field resistance R_{ps} of some typical samples are listed in Table I Total light scattering A_s, measured at 6328 Å, is found to be negligible compared to the total absorption. Therefore, total light absorption is assumed to be only due to metallic absorption A_m of the films, and A_m is calculated from $R + T + A_m = 1$.

From Table I, it can be seen that films which are prepared at low pressures show high optical absorption due to large amounts of randomly distributed excess metallic particles embedded in the oxide. The thickness augmentation k of these low-pressure films is nearly unity. As the pressure is increased, k increases, which indicates a rough oxide formation probably due to the change in grain size with the oxygen pressure during evaporation. As can also be seen from Table I, the preswitching resistance values of the mixtures fall between the pure metal and the pure insulator. The structure of the films is not investigated in this work. Static electrical I-V characteristics of some typical samples are illustrated in Fig. 2, and these correspond to samples Nos. 2, 4, and 7 in Table I. As the deposition pressure is increased, first metallic conductivity (curve a), then switching and memory effects (curve b, 1–5), and finally oxide characteristics (curve c) are observed in respective samples.

Curve a in Fig. 2 shows typical electrical characteristics of a sample containing considerable aluminum within the oxide. The process of switching and memory effects can be explained by using curve b. Under the initial application of an increasing voltage, high resistance or off-state condition (line 1) is observed until a threshold voltage V_{th} is reached. Then the sample makes a transition to the conduction or on state following the load line (line 2). In this on state (line 3), the device is also found to be in its memory state, i.e., if the applied voltage is reduced to zero, it returns to the origin following line 6 and it remains in this conduction state for subsequent traces unless the current exceeds a blocking value I_b. After reaching I_b, the sample switches back following the load line (line 4) to its high-resistance state (line 5). As the voltage is then reduced, the

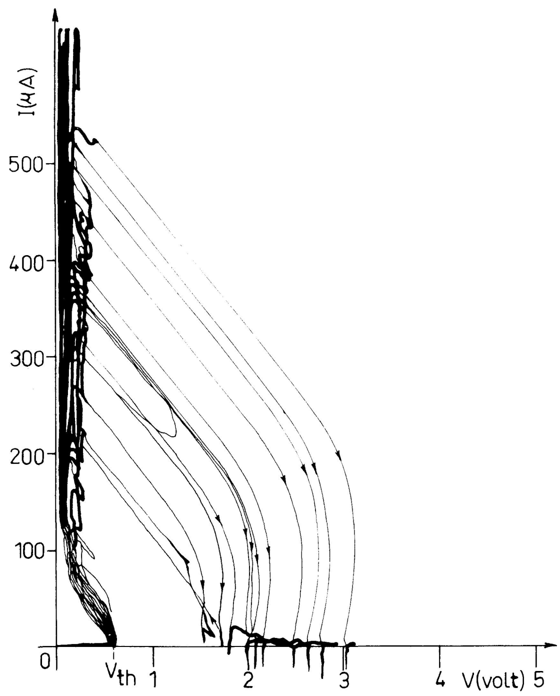

FIG. 3. I-V characteristic of a typical switching element (Au electrodes, $k = 1.04$, $d_{int} = 520$ Å). Traced by the X-Y recorder.

sample returns to the origin following lines 5, 7, and 1, and the next trace repeats the original characteristics (lines 1–5). Segments 4 and 5 of curve b do not conform to the switching behavior of amorphous materials currently found in the literature.[12] The I-V characteristics of the films investigated in this work do not show hysteresis effects, and a single sample exhibits both threshold switching and memory.

I-V characteristics of nearly oxide films which are similar to those found in metal-oxide-metal sandwiches[13] are shown in curve c of Fig. 2. The current increases linearly (Ohmic region) at low voltages (up to ~ 0.5 V), and then there is an exponential dependence on voltage up to ~ 1.2 V. From then on the current is proportional to V^n, where n is nearly 5 for the given sample.

Figure 3 gives I-V characteristics of a typical switching element traced many times. The time slope of the sawtooth generator used is 0.25 V/sec, and the series load resistance R_L is 4.8 kΩ. The transition curves from the off to on and from the on to off states are due to the mechanical time response of the X-Y recorder. Oscilloscope traces confirmed that these transitions should be straight lines and follow the external load line.

The values of V_{th} and I_b depend on the series load resistance R_L and the time slope of the ramp voltage but not on the polarity of the applied voltage. V_{th} depends also on the Al concentration of the films and the electrode material but shows considerable dispersion for Al electrodes.

The off- and on-state resistance values of the samples tested are around 1 MΩ and a few hundred Ω, respectively. The on-state resistances are directly proportional to the blocking current I_b, as can be seen from Fig. 3. The higher is the value of I_b, the steeper is the slope of the corresponding curve.

Some samples showed only switching effects, whereas others had both switching and memory characteristics. Some virgin films that initially had metallic conductivity showed switching properties after sufficient current is passed through them. The electrical properties of the samples did not change for a long time provided the sandwiches are kept in dry air.

All these experimental results indicate that the switching properties of the samples depend on the amount of metallic aluminum embedded in the oxide. The transitions from the off to on and from the on to the off states may be explained by filament formation and its rupturing process, respectively. A thin oxide film which is placed between two electrodes and subjected to an increasing electric field may initiate the growing of a metallic bridge by the migration of aluminum within the oxide. The heat dissipation may help in forming the filament. Depending on the filament size and number, the blocking current is different for each trace for the same sample, which indicates a lower on-state resistance for large filaments and a corresponding increase in I_b to break these filaments. The load resistance and the voltage sweep rate may effect this formation. However, the threshold V_{th} is constant irrespective of the size and number of the filaments for the same sweep rate and load resistance.

Electronic processes initiated by the high electric field may create energetic conditions which provide the limited atomic motion allowing structural reorganization to take place. Electric field, current density, and thermal gradients may effect the filamentary ordering since the samples are assumed to be amorphous and anisotropic in nature because of the presence of metallic aluminum within the oxide, due to the evaporation process.

I would like to thank Dr. Altan M. Ferendeci for many helpful discussions and critical reading of the manuscript during the course of this work.

[1]D. Mayerhofer and S.A. Ochs, J. Appl. Phys. **34**, 2535 (1963).
[2]K.M. Eisele, in *Basic problems in thin film physics*, edited by R. Neidermayer and H. Mayer (Vanderhoeck and Ruprecht, Göttingen, 1966), p. 672.
[3]T.W. Hickmott, J. Appl. Phys. **33**, 2669 (1962).
[4]G.A. Mead, J. Appl. Phys. **32**, 646 (1961).
[5]P.O. Sliva, G. Dir, and C. Griffiths, J. Non-Cryst. Solids 2, 320 (1970).
[6]K. Sawamura and M. Saito, Jap. J. Appl. Phys. **5**, 182 (1966).
[7]J.C. Fisher and I. Giaever, J. Appl. Phys. **32**, 172 (1961).
[8]E.M. Da Siliva and P. White, J. Electrochem. Soc. **109**, 12 (1962).
[9]J.A. Bennett, Electron. Components 5, 737 (1964).
[10]B. Lewis, Microelectron. Reliab. **3**, 109 (1964).
[11]Equal amounts of pure aluminum are used as the source material during evaporation, and the ratio of metallic aluminum thickness deposited at the lowest chamber pressure to the thickness of the evaporated material at the given pressure p in Table I is called the thickness augmentation k.
[12]For a review and list of references on amorphous semiconductors, see IEEE Trans. Electron Devices **ED-20**, No. 2 (1973).
[13]C.A. Mead, in *Basic problems in thin film physics*, edited by R. Neidermayer and H. Mayer (Vanderhoeck and Ruprecht, Göttingen, 1966), p. 674.