Cryst. Res. Technol. **39**, No. 5, 404 – 407 (2004) / **DOI** 10.1002/crat.200310202

Mott-hopping processes in neodymium oxide thin films prepared on Si(100) substrates

A. A. Dakhel*

Dept. Of Physics, College of Science, University of Bahrain, P.O. Box 32038, Bahrain

Received 15 September 2003, revised 10 December 2003, accepted 14 January 2004
Published online 15 April 2004

Key words insulating films, neodymium oxide, dielectric phenomena, Mott VRH.
PACS 77.55.+f, 72.20.-I

Thin Nd oxide films were prepared on Si (P) substrates to form MOS structure. The oxide films were annealed at different conditions and their crystal structures were determined by X-ray diffraction (XRD). The dc electrical transport properties of the devices with amorphous and crystalline Nd oxide were investigated. The current-temperature J(T) characteristics suggest that the carrier transport through the insulator follows Mott's variable-range hopping (VRH) mechanism and its results were compared with the results obtained from X-ray diffraction.

© 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

1 Introduction

Rare earth oxides (REOs) have many applications in solid-state electronics, optics and transparent opto-ionic devices. The basic promise of REOs in electronics is that of enabling high-temperature and high-power devices. REOs have high resistivity ($\rho = 10^{12} – 10^{15}$ Ω.cm), high dielectric constant (ε = 7-20), large band gap (E_g = 4–6 eV.)[1], high recrystallization temperature and thermodynamically stable in contact with Si substrate at high temperatures (~800°C) under high vacuum or in N_2 atmosphere [2]. But annealing in oxygen atmosphere can produce a silicate layer due to Si atoms diffusion from substrate into the REO [2,3]. There is also another problem when dealing with REOs that is the hygroscopic nature, which leads to formation of hydroxide after exposure of the film to atmospheric conditions. The hygroscopic nature may noticeably affect the electrical properties.

In this work, we examine the hopping mechanism, which controls the carrier transport, of Nd_2O_3 films annealed at different conditions as a function of oxide structure. This requires the study of temperature dependence of the dc conductivity of samples annealed at different conditions.

REO as an insulator in MOS structures were investigated in many works [1-13]. But the study of Nd_2O_3 is seldom.

2 Experimental

Samples of Al/Nd_2O_3/Si MOS structure were prepared and examined in this investigation. The p-type Si wafers with (100) orientation at doping level of 1.2 x 10^{16} cm^{-3} were utilised as a substrates. These wafers were degreased in acetone, rinsed in deionised water, etched in 48% HF to remove the native oxide layer and then rinsed with deionised water, before introduction into the high vacuum chamber. Prior to deposition the wafers

* Corresponding author: e-mail: adakhil@sci.uob.bh

© 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

were degassed by heating at about 150 °C for 3h in vacuum of about 10^{-4} Pa, and Nd oxide was obtained by evaporation of pure Nd in residual pure oxygen atmosphere of pressure about 10^{-4} Pa. Thickness was monitored by thickness monitor and then measured after annealing by Gaertner L117 ellipsometer. The simultaneously prepared samples were classified into three groups relative to their annealing conditions and hence according to their crystal structures. Films annealed in air at 300 °C for 15 min are named AM300, annealed in air at 800 °C for 15 min. are named CR800 and annealed at 800°C in vacuum of 10^{-4}Pa for 15 min. are named CR800v samples. After sample annealing, aluminium film of around 150 nm was deposited to form a gate.

The crystal structure was investigated with Philips PW 1710 X-ray diffractometer. Keithley 3330 LCZ meter with a signal of 50 mV performed the capacitance measurements. The dc measurements were done using the standard technique with Keithley 614 electrometer. Measurements were done in room temperature and in temperature controlled furnace.

3 Structural characterisation

The X-ray diffraction of light blue neodymium oxide powder is shown in fig.1. It shows a mixed crystal structures A- and C-phases. The lattice constant of cubic phase is a = 11.08 Å and of hexagonal phase are: a= 3.83 Å and c= 6.00 Å; in agreement with published data [14]. Films AM300 were amorphous while films CR800 and CR800v show a polycrystalline structure of mixed A – and C-structure with a slightly shifted peaks relative to the powder peaks positions. This shift appears as results of residual structural microstresses in thin films. The average grain size (g.s) estimated from (222) line is 8.5 nm for CR800 and 68 nm for CR800v.

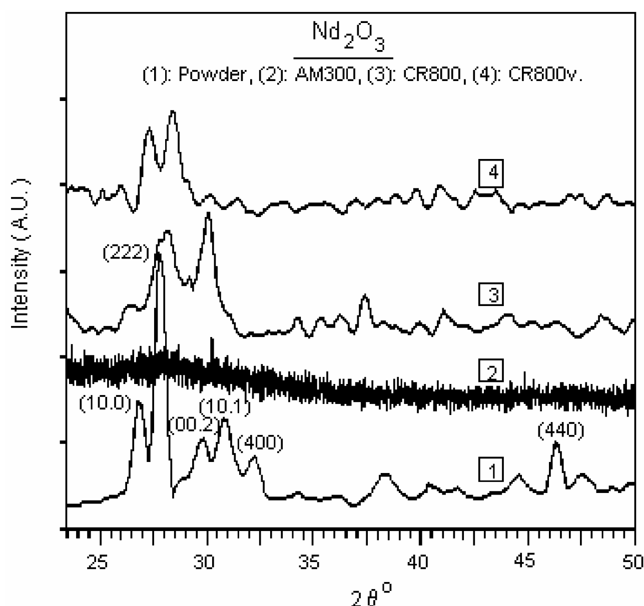

Fig. 1 X-ray diffraction pattern from Nd_2O_3 powder, amorphous AM300 film and polycrystalline CR800 and CR800v films. The scan speed was 0.01 °/s.

Results and discussion

The temperature dependence of the electrical conductivity σ (T) of Nd_2O_3 samples were studied at accumulation polarity in the temperature range (300 – 500 K).

The low average activation energy of the conductivity of AM300, CR800 and CR800v samples and the fulfilment of the conditions (as we will see latter) permit us to assume that the hopping process of the carriers

© 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

between localized states would be governed by a variable range-hopping (VRH) mechanism as characterized by Mott's expression of the form [15-17]:

$$\sigma(T) = (\sigma_0 / T^{0.5})\exp\{-(T_0 / T)^{0.25}\} \tag{1}$$

where σ_o is a pre-exponential factor, T_0 is the degree of disorder $T_0= \lambda\alpha^3 / k_B N(E_F)$, $N(E_F)$ is the density of the localized states at the Fermi level E_F , λ is a dimensionless constant (about 16 as given by ref [17], for other values for λ see the survey given by [18]) and α^{-1} ($\alpha=10^7$ cm^{-1}) represents the degree of localization.

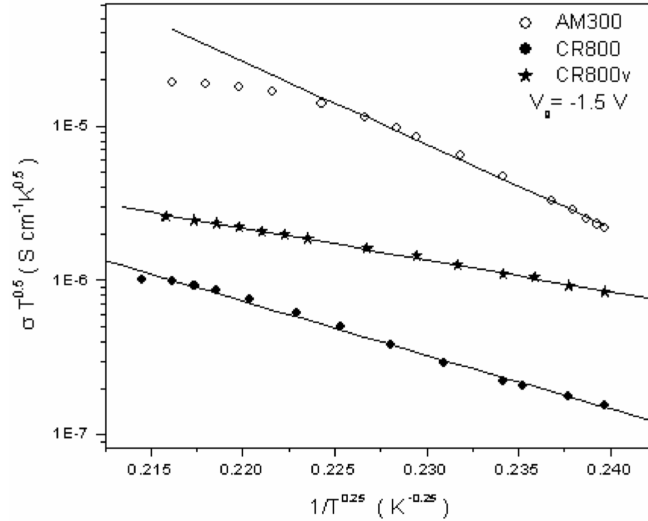

Fig. 2 $\ln(\sigma T^{0.5})$ vs. $T^{-0.25}$ plot (the Mott's VRH plot) for AM300, CR800 and CR800v structures. The conductivity was measured at gate voltage V_g= -1.5 V.

Table 1 Parameters of Mott's VRH mechanism: T_0 is the degree of disorder, $N(E_F)$ is the density density of the localized states at Fermi level, R is the hopping distance at 293 K and W is the average hopping energy. g.s is the average grain size. The dielectric constants of the samples ε were measured at 10 kHz.

Sample	T_0(K)	N (E_F) ($eV^{-1}cm^{-3}$)	R (nm)	W (meV)	g.s (nm)	ε
AM300	2.37×10^8	7.8×10^{17}	11.6	190	-	11.70
CR800	4.17×10^7	4.4×10^{18}	7.5	130	8.5	8.47
CR800v	4.86×10^6	3.8×10^{19}	4.3	70	68.0	10.14

Fig. 2 shows the Mott's VRH plot : $\ln(\sigma T^{0.5})$ vs. $T^{-0.25}$ for AM300, CR800 and CR800v structures. At about 373 K with only amorphous sample (annealed at 300 °C), a deviation from the linearity of Fig. 2 was observed. This related to the hygroscopic nature of the amorphous oxide. Sample CR800 has less conductivity and dielectric constant than CR800v; this is due to the formation of SiO_2 layer of thickness that can be estimated from capacitance measurements. From the relation of two "series-connected capacitors model" [19] and by using the dielectric constants (ε) of CR800 and CR800v measured from the accumulation capacitance at 100 kHz it is possible to estimate SiO_2 layer thickness in CR800 to be about 12.8 nm. The formation of a very thin SiO_2 layer, say 1- 2.5 nm [4,20], even for AM300 sample is also possible although the value of ε of AM300 is well-agree with the value (12.6) given at 1 kHz in metal-insulator-metal MIM structure [21]. But if the interfacial layer has more complicated structure like amorphous SiO_x layer [22], mixed REO with SiO_2 in non-stoichiometric composition [23] or Nd silicate with SiO_x [24] then its analysis needs direct-probe methods like Rutherford back scattering (RBS), electron microscopy, Auger or photo electron spectroscopy [23, 24].

The hopping distance R and the average hopping energy W are given by [17]: $R=\{9/8\pi\alpha k_B T N(E_F)\}^{0.25}$, and $W=3/4\pi R^3 N(E_F)$. The calculated values of T_0, $N(E_F)$, R and W for our samples are given in table 1. As T_0

© 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim

represents the degree of disorder or degree of amorphisity, it is clear from table 1 that sample CR800v is ordered more than CR800 or the average grain size of CR800v is larger than that of CR800, this conclusion is supported by the X-ray experimental results. It is also observed from our experimental data that as disorder degree increases, the density of localized states at Fermi level decreases; such observation was also given by the data published in [25,26] for amorphous glasses and their degree of amorphisity. It is clear from table1 that the conditions [15] of possibility of applying VHR model i.e.: $W > k_B T$ and $\alpha R > 1$ are satisfied.

References

[1] V. A. Rozhkov, A. Yu. Trusova, and I. G. Berezhnoy, Thin Solid Films **325**, 151 (1998).
[2] J. Kwo, M. Hong, B. Busch, D. A. Muller, Y. J. Chabal, A. R. Kortan, J. P. Mannaerts, B. Yang, P. Ye, H. Gossmann, A. M. Sergent, K. K. Ng, J. Bude, W. H. Schulte, E. Garfunkel, and T. Gustafsson, J. Cryst. Growth **251**, 645 (2003).
[3] Z. M. Wang, J. X. Wu, M. S. Ma, W. Chen, Q. Fang, and J.-Y. Zhang, Microelectronic Engineering (Article in press).
[4] V. Mikhelashvili and G. Eisenstein, J. Appl. Phys. **90**, 5447 (2001).
[5] H. J. Osten, J. P. Liu, E. Bugiel, H. J. Mussig, and P. Zaumseil, J. Cryst. Growth **235**, 229 (2002).
[6] H. Vidal, S. Bernal, R. T. Baker, D. Finol, J. A. Perez Omil, J. M. Pintado, and J. M. Rodriguez-Izquierdo, J. Catalysis **183**, 53 (1999).
[7] A. C. Rastogi and R. N. Sharma, J. Appl. Phys. **71**, 5041 (1992).
[8] Ya. G. Fedorenko, L. A. Otavina, and S. V. Korenyuk, Pis'ma v Zhurnal Tekhnicheskoi Fizika, (2000) pp. 916-18, ISSN of translation 0320-0116.
[9] R. J. Gaboriaud, F. Pailloux, P. Guerin, and F. Paumier, Thin Solid Films **400**, 106 (2001).
[10] D. Landheer, J. A. Gupta, G. I. Sproule, J. P. McCaffrey, M. J. Graham, K.-C. Yang, Z.-H. Lu, and W. N. Lennard, Journal of Electrochemical Society **148**, G29 (2001).
[11] J. A. Gupta, D. Landheer, G. I. Sproule, J. P. McCaffrey, M. J. Graham, K.-C. Yang, Z.-H. Lu, and W. N. Lennard, Appliedsurface Science **173**, 318 (2001).
[12] S. Wang, W. Wang, and Y. Qian, Materials Research Bulletin **35**, 2057 (2000).
[13] W. A. Henle, M. G. Ramsey, F. P. Netzer, and K. Horn, SurfaceScience **254,** 182 (1991).
[14] Roger M. Nix, Robert W. Jugg, and Richard M. Lambert, Surface Science **205**, 59 (1988).
[15] N. F. Mott and E. A. Davis, Philos. Mag. **22**, 903 (1970).
[16] N. F. Mott, Philos. Mag. **32**, 961 (1975).
[17] N. F. Mott and E. A. Davis, Electronic process in nanocrystalline materials, Clarendon, Oxford, 1979.
[18] A. Ganguly, S. K. Mandal, S. Chaudhuri, and A. K. Pal, J. Appl. Phys. **90**, 5652 (2001).
[19] Li Hong-Chengi, Si Weidong, D. W. Alexander, and X. X. Xi, Appl. Phys. Letters **73**, 464 (1998).
[20] Sanghun Jeon, Matsuda Takanori, Akira Unno, Kiyotaka Wasa, Yoko Ichikawa, and Hyunsang Hwang, Vacuum **65**, 19 (2002).
[21] Vineet S. Dharmadhikari and A. Goswami, Thin Solid Films **87**, 119 (1982).
[22] H. J. Osten, E. Bugiel, and A. Fissel, Solid-State electronics **47**, 2161 (2003).
[23] D. Schmeisser and H.-J. Muessig, Abstract of EMRS 2002- spring meeting, June 18-21, Strasbourg, France, Symposium title: The 300 mm Silicon era: Material, equipment, technology.
[24] H. Nohira, T. Shiraishi, T. Nakamura, K. Takahashi, M. Takeda, S. Ohmi, H. Iwai, and T. Hattori, Appl. Surf. Sci. **216**, 234 (2003).
[25] M. A. Majeed Khan, M. Zulfequar, and M. Husain, J. Phys. Chem. Solids **62**, 1093 (2001).
[26] E. A. Mohmoud, M. M. El-Samanoudy, and A. S. Abd Rabo, J. Phys. Chem. Solids **63**, 2003 (2002).

© 2004 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim