

Electrical conductivity, mobility and carrier concentration in Nb₂O₅ films: effect of NH₄OH Molarity

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ABSTRACT

This work presents, the effect of the NH₄OH molarities on the electrical characteristics of Nb₂O₅ films prepared using the precipitation method. Hall Effect measurements evidence the formation of n-type Nb₂O₅ films. The carrier concentration was found to increase with ammonium hydroxide molarities up to 12mol/L, while the carrier mobility decreased. Resistivity- temperature measurements confirm the semiconductor behaviors of the thin films, it also shows a decrease in the resistivity and activation energy with molarity increasing reaching its minimum value in 12 mol /L then they increased again.

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1. INTRODUCTION

Newly, there has been rising attention in materials representing metal-insulator transitions (MITs) on account of their promising implementations in electronic apparatus [1-3]. NbO₂ is amongst these materials which displays one of the elevated MIT temperatures of 1081K joined through a conversion in the structure from deformed rutile (depressed temperature phase) into a rutile construction (rise temperature) [4-7].

In new year's, Niobium Pentoxide (Nb₂O₅) appears as a multifunctional material as a result of its remarkable outstanding characteristics like electrochromic attitude, rise refractive index, superior chemical and thermal stabilization in an aqueous ambiance, excellent electrochemical responses, redox, photocatalytic and photoelectric activity, impedance to corrosion in alkaline and acidic ambiance together, displays band levels nearby water redox possibilities and surface acidity [8-19]. Thus, it has been deliberated in various implementations as a favorable alternate material to TiO₂.

Nb₂O₅ presents an inorganic semiconductor its band gap ranging bandgap from 2.8 to 3.8 eV relying on the procedures of the preparation [20-22], little in contrast to additional oxides. It has a small electrical conductivity at 300 K of about 3.4×10^{-6} S/cm [23] which is two orders of the amount at minimum lesser in contrast to its electrochromic metal oxides coordinates like WO₃ and MoO₃ [24, 25].

It has been found that with the insertion of ionic type, crystalline niobium pentoxide demonstrates an alteration of color to blue from transparent, however amorphous Nb₂O₅ variations into a brownish-grey color [10].

Nevertheless, Nb₂O₅ system is complex particularly, as slight aberrations from the particular stoichiometry in Nb₂O₅ affect powerfully the material's physical properties. As an example, a

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minor oxygen lack arrives at the conversion to n-type semiconducting from insulating behavior [26-28].

Further, several implementations in photonics request materials of rising refractive index with pretty optical goodness and insignificant scattering. Therefore, numerous new applications have been advanced and Nb₂O₅ thin films have originated implementations in photonics for beneficent the optical achievement of various equipment like optical filters, transparent conductive electrodes, waveguide-based optical circuits, electro-chromic show, gas sensing, as well as in field-emission presentations, optical coatings, capacitors, EC devices, microelectronic devices, such as optoelectronics detectors [29-39].

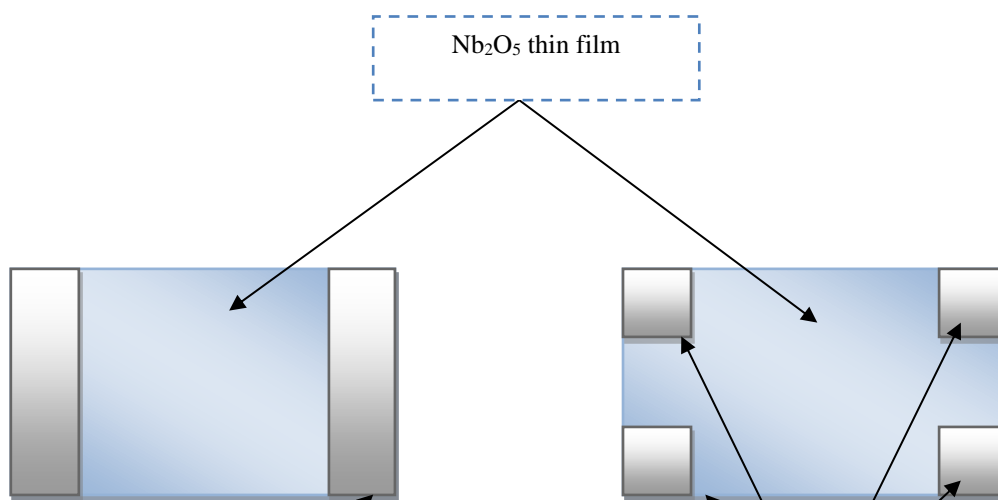
Novel implementations of Nb₂O₅ films depending on the capability to deposit rise fineness films employing comparatively unpretentious and low-cost procedures. Generally, Nb₂O₅ thin films can be intended with various optical and structural characteristics relying on the situations of deposition such as the beginning materials, procedures of synthesis, and the conditions of heat treatment [30, 40, and 41].

2. MATERIAL AND METHODS

Nb₂O₅ suspension was prepared by adding HF and ammonium hydroxide to Nb₂O₅ powder as shown in details [42, 43]. The suspension was deposited on a glass substrate using the spin coating method at 1500 rpm for 1 min. The film was subsequently dried in a drying oven at 100 °C for 15 min. This process was repeated three times for the formation of three film layers. The thickness of each layer was measured to be 150 nm using a scanning optical reflectometer (Filmetrics F20, USA). The thickness and the number of layers were selected based on the optimization process. The electrical resistant of the films on glass substrates was measured under different temperature ranged from 40 to 200 C employing (KEITHLEY 616) meter. Electrodes were made using aluminum metal through a standard mask as shown in figure (1A). The activation energy (Ea) was estimated using log σ vs. 1000/T curve, where it value could be found from the following equation (1) [44-46]:

$$E_a = 0.86 * \text{slope} \quad (1)$$

Using Hall Effect measurement (HMS3000 Ecopia) the mobility, type of conductivity, and carrier concentration has been investigated, by depositing aluminum metal electrodes through a standard mask as shown in figure (1B)



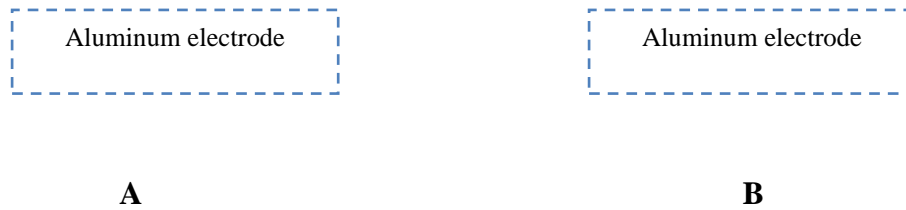


Figure 1. Aluminum electrode deposition for A: R-T B: Hall effect measurements.

3. RESULTS AND DISCUSSION

Figure (2) demonstrated the decrease of the hall coefficient values with an increase in the molarity. The negative sign of the coefficient indicates that the negative carrier is moderated for the films at various molarities; this indicates that n-type semiconducting films have been prepared. This may be related to the O²⁻ (donor) ions and the Nb⁺⁵ incorporation as shown in other work [47, 48]. Oxygen - vacancies of the oxide such as the electron donors induced the anodic oxide to act as an n-type material because of its smallest enthalpy in between the donors similar to a point defects, which results in the formation of deep state electrons, such defect could not be ionized and contribute to the conductivity as shown in other work [49, 50].

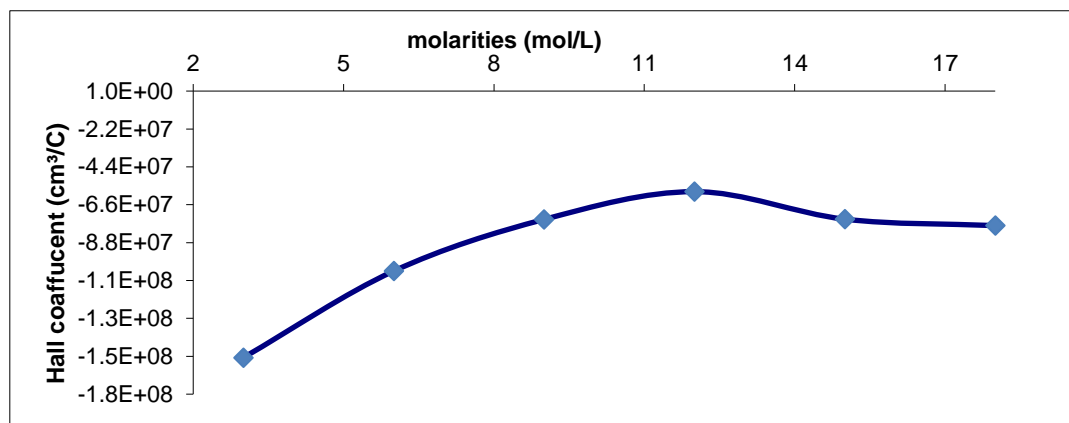


Figure 2. Hall coefficient for Nb₂O₅ films with the ammonium hydroxide molarities.

Carrier concentration was varied with molarities due to the variation in the Hall coefficient according to relation (2)[51]:

$$n = \frac{1}{eR_H} \quad (2)$$

Figure (3) shows the molarities as a function of charge carrier concentration. In this figure, the increase in the charge carrier concentration could be recognize as the ammonia molarities increase which may be related to the increase Nb₂O₅ molecules at the suspension which reached to its maximum amount at 12 mol/L. Further increase in hydroxide molarities will results in a reduction in the carrier concentration because of the separation in the formed Nb₂O₅ remaining a very small material amount within the suspension.

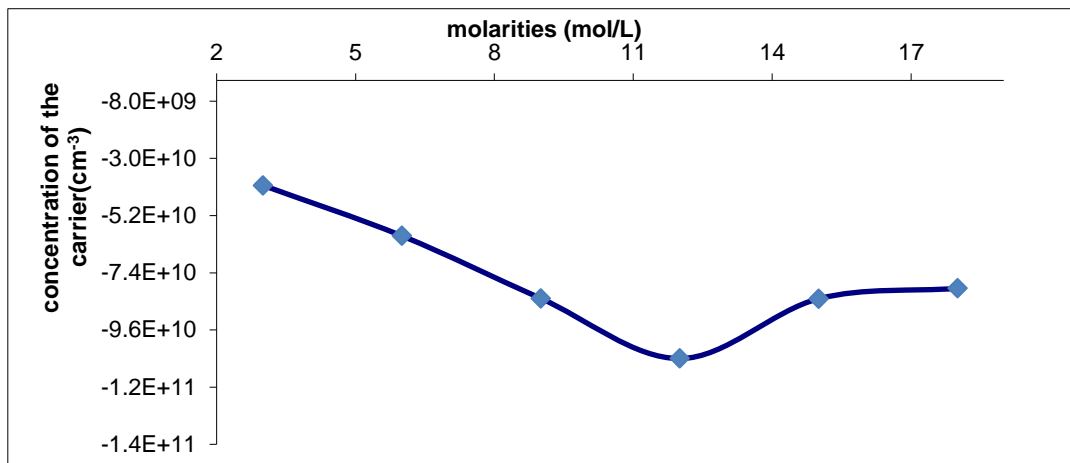


Figure 3. Nb₂O₅ films carrier cons. with the ammonium hydroxide molarities.

Electrical mobility of the carrier affected also by the molarity concentration of the ammonium hydroxide because of its direct effect on their concentration, since its value related inversely to the concentration due to the scattering effect as given in fig. (4), similar results could be shown in other work [52, 53].

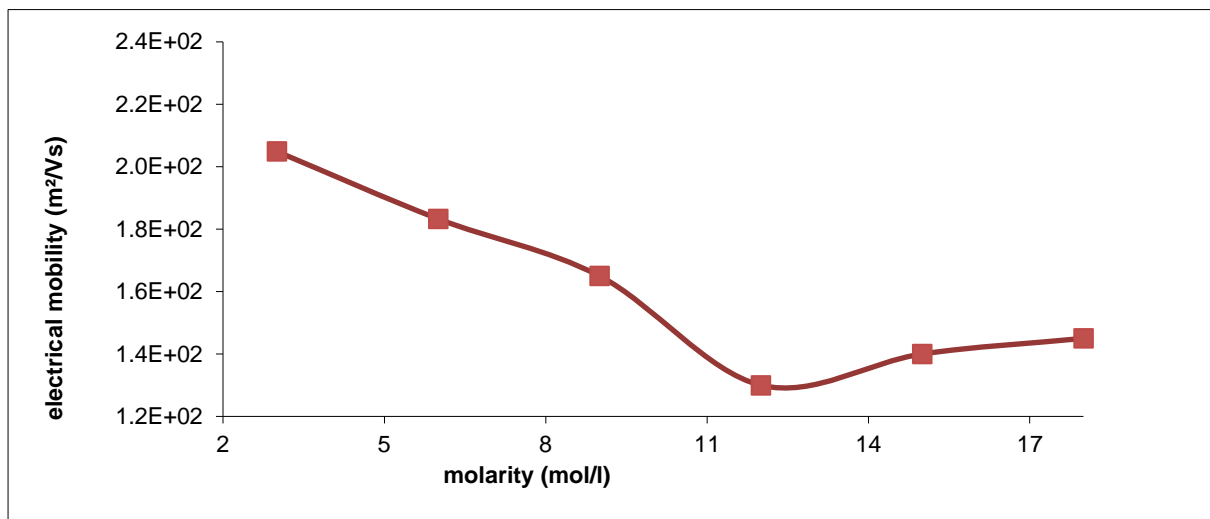


Figure 4. electrical mobility for Nb₂O₅ thin films with the ammonium molarities.

Figure (5) shows the resistivity and molarity relation at different molarities. It is found to related directly concentration of the carriers, the resistivity reaching to a minimum at 12 mol/L followed by a slight increase as the molar ratio increase.

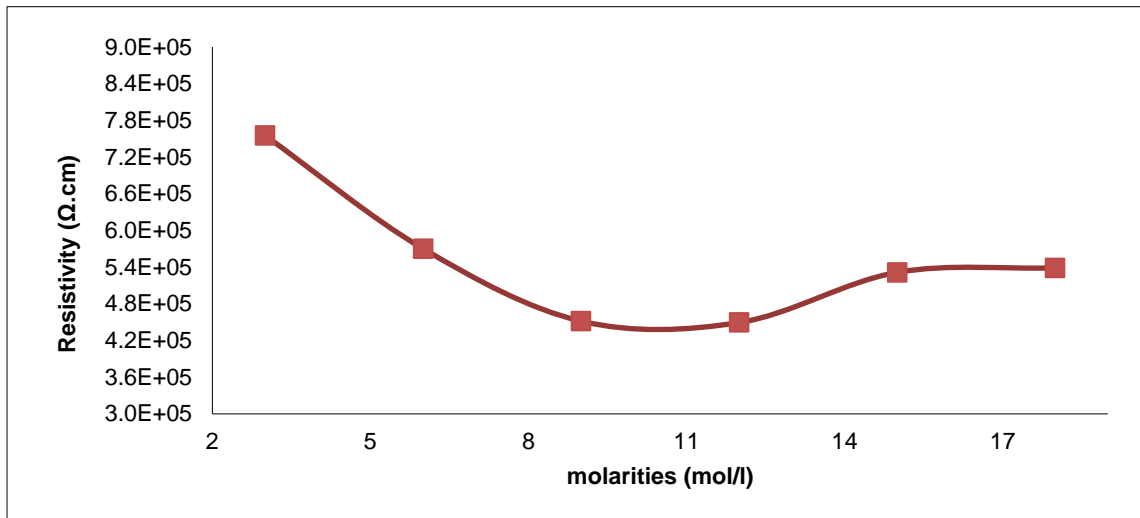


Figure 5. The electrical resistivity of Nb₂O₅ film with the ammonium molarities.

Figure (6) shows the investigation study of the resistivity of the Nb₂O₅ film at a wide temperature range of (40-200°C) in order to decide their semiconducting properties at different ammonium molarities of (3, 6, 9, 12, 15 and 18 mol/L). Films electrical resistivity found to reduce as the molarity of the ammonium hydroxide increase to reach minimum at 12mol/l, it return back to increase as ammonium molarity increase further beyond (12) mol/l.

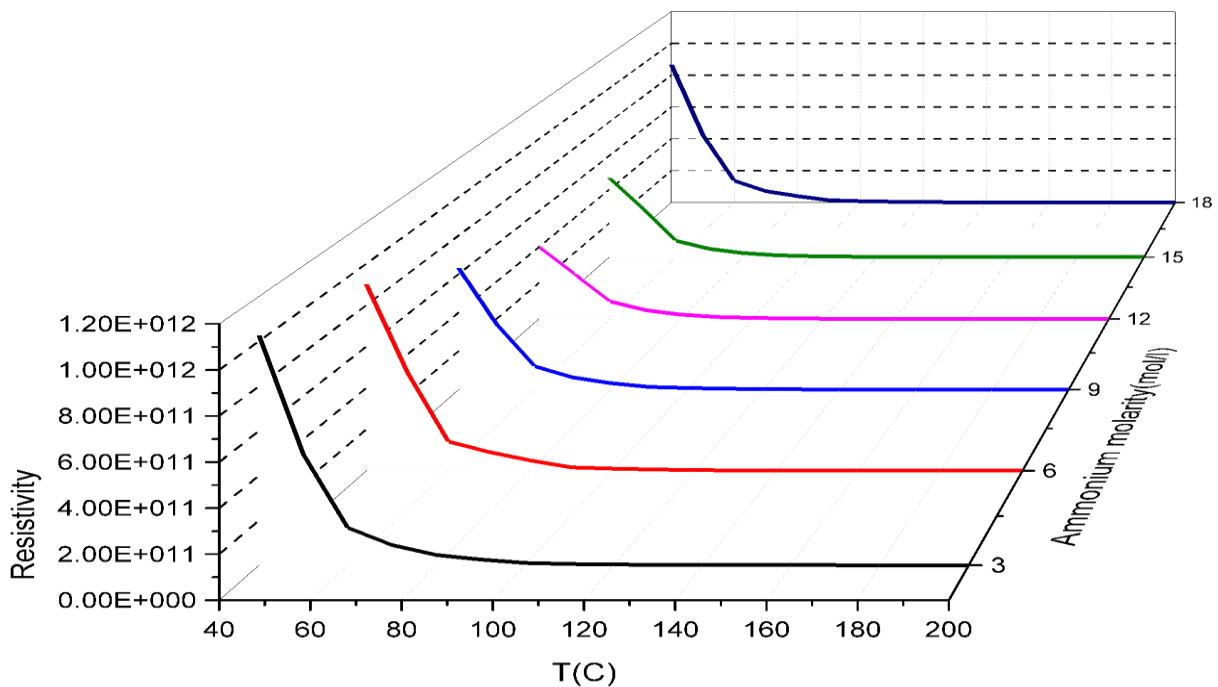


Figure 6. Resistivity of for Nb₂O₅ films with ammonium molarities at different temperatures.

This is because the increase in molar ratio resulting in increased the carrier concentration, so that resistance is decreasing with the increase in concentration as the number of ions is increasing in the same volume, supporting the flow of current. A simple explanation is as follows. By increasing the molarity of the precursor you increase the thickness (D) of the resulting film.

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As the molarity increase beyond 12M, the increase in material concentration began to act as trapping center and defects which play an inverse role leads to increase the resistivity again.

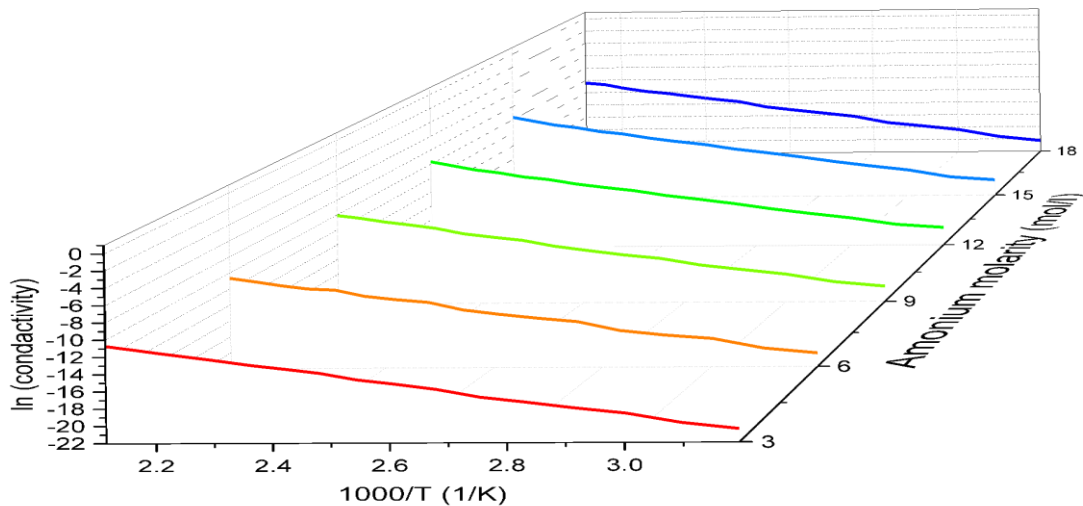


Figure7. The conductivity with $1/T$ for Nb_2O_5 thin films at different molarities.

This result obeyed to the estimation from Hall measurement. This also accomplished with semiconductor behavior as given equation (3) [54],

$$\sigma = \sigma_0 \exp\left(\frac{-E_a}{kT}\right) \quad (3)$$

σ = Electrical conductivity at specific temperature, σ_0 is the Electrical conductivity when $T \rightarrow \infty$, E_a is the activation energy and k is Boltzmann constant. The activation energy values could be extracted from the relation between the logarithm of the conductivity and $1000/T$, it could recognize the linear behavior with a negative slope as presents in Fig (7). Typical semiconducting behavior could be recognized. The calculated energy of activation is found in Figure (8) with the hydroxide molarities.

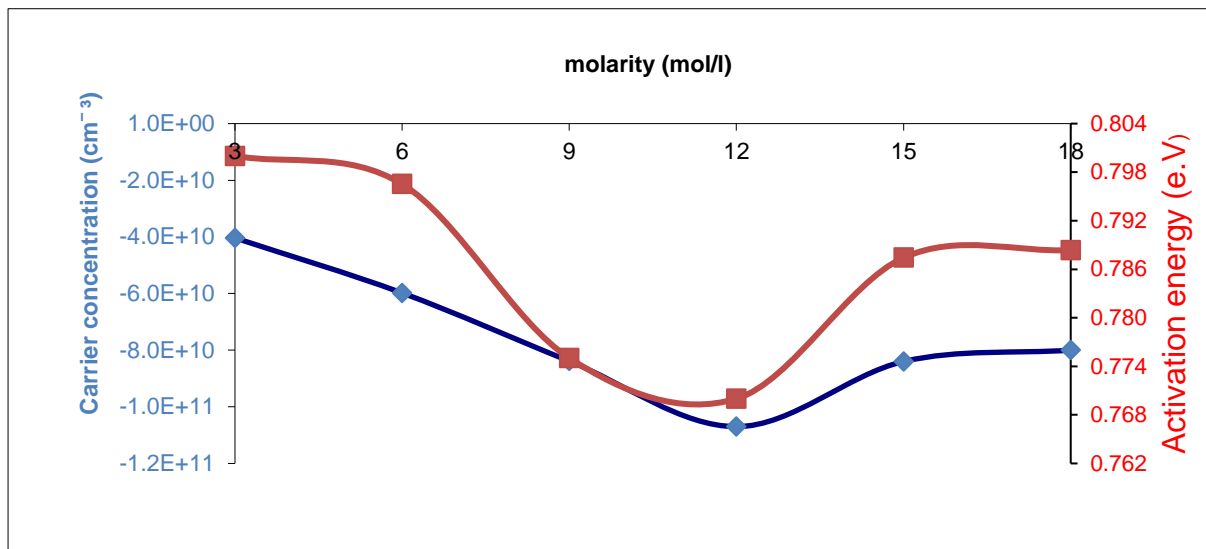


Figure 8. Activation energy, a carrier concentration for Nb₂O₅ thin at different molarities.

The inverse relation between the energy of activation is and hydroxide molarities up to 12mol/L could recognize after which it begins to increase again. This may be related to increasing the concentration of electronic carries since this energy represented the difference in the Fermi level energy, as a result, these values could be used to estimate the Fermi energy position. In the same manner, the reduction in the activation energy at low hydroxide concentration may be related to the increase in electronic concentration. This behavior could understand using the following equation (4) [55]:

$$n = N_c \exp [(E_F - E_c)/KT] \quad (4)$$

Where n is the free carrier concentration (electrons), N_c is the effective electrons concentration at the conduction band, E_c , E_F are the bottom of conduction band and Fermi level energy respectively.

4. CONCLUSION

By studying the effect of ammonium hydroxide concentration on electrical properties of Nb₂O₅ thin film, it could be concluded that 12M is the optimum preparation condition, where minimum film resistivity and maximum conductivity were obtained. In addition, the activation energy was also found to be minimum at this molarity.

REFERENCES

- [1] A.B.S.Posadas, A.O'Hara, S.Rangan, R.A.Bartynski, A.A.Demkov, Appl. Phys. Lett. 104 (2014) 092901.
- [2] F. J. Wong, N. Hong, S. Ramanathan, Phys. Rev. B 90 (2014) 115135.
- [3] E.T. Salim, R.A. Ismail, & H.T. Halbos, Appl. Phys. A 126 (2020) 891.
- [4] A. A. Bolzan, C. Fong, B. J. Kennedy, J. Solid St. Chem. 113 (1994) 9-14.

- [5] E.T. Salim, J.A. Saimon, M.K. Abood, M.A. Fakhri, *Optical and Quantum Electronics* 52(10) (2020) 463
- [6] F. Hashemzadeh, A. Gaffarimejad, R. Rahimi, *Journal of Hazardous Materials*, 286 (2015) 64-74.
- [7] T.Sreethawong, S.Ngamsinlapasathian, SH.Lim, S.Yoshikawa, *Chemical Engineering Journal*, 215-216 (2013) 322-330.
- [8] AD Faisal, RA Ismail, WK Khalef, ET Salim, *Optical and Quantum Electronics* 52 (2020) 1-12.
- [9] M.L.Marin, G.L.H.-Tapley, S.Impellizzeri, C.Fasciani, S.Simoncelli, J.C.N.-Ferreira, J.C.Scaiano, *Catalysis Science & Technology*, 4 (2014) 3044-3052.
- [10] A.V.Rosario, E.C.Pereira, *Sol. Energ. Mat. Sol. C.* 71 (2002) 41-50.
- [11] H Asady, ET Salim, RA Ismail, *AIP Conference Proceedings* 2213 (1) (2020) 020183
- [12] D. D.Yao, R. A.Rani, A. P.O'Mullane, K.K.-Zadeh, J. Z.Ou, *J. Phys. Chem. C*, 118 (2014) 476-481.
- [13] A.Verma, P. K.Singh, *Indian J. Chem. A*, 52 (2013) 593-598.
- [14] ET Salim, MA Fakhri, Z Tareq, U Hashim, *AIP Conference Proceedings* 2213 (1) (2020) 020230
- [15] Y. Zhao, X. Zhou, L. Ye, S.C.E. Tsang, *Nano Reviews*, 3 (2012) Article ID 17631(1-11).
- [16] Y. Huang, Y. Xu, S.-J. Ding, H.-L. Lu, Q.-Q. Sun, D. W. Zhang, Z. Chen, *Applied Surface Science* 257 (2011) 7305-7309.
- [17] MA Fakhri, ET Salim, MHA Wahid, ZT Salim, U Hashim, *AIP Conference Proceedings* 2213 (1) (2020) 020242
- [18] Y. Y.Zhou, Z. F.Qiu, M. K.Lü, A. Y.Zhang, Q.Ma, *J. Lumin.* 128 (2008) 1369-1372.
- [19] M.K. Abood, M.H.A Wahid, J.A. Saimon, ET Salim, *International Journal of Nanoelectronics and Materials*, 11 (Special Issue) (2018) 237-244.
- [20] H.Liu, N.Gao, M.Liao, X.Fang, *Scientific Reports*, 5 (2015) Article number: 7716 (1-9).
- [21] A.L.Viet, M.V.Reddy, R.Jose, B.V.R.Chowdri, S.Ramakrishan, *J. Phys. Chem. C* 114 (2010) 664-671.
- [22] MT Awayiz, ET Salim, *AIP Conference Proceedings* 2213 (1) (2020) 020247
- [23] G. R.Lee, J. A.Crayston, *J. Mater. Chem.* 1 (1991) 381.
- [24] J. Z.Ou, R. A.Rani, M.-H.Ham, M. R.Field, Y.Zhang, H.Zheng, P.Reece II, S.Zhuyikov, S.Sriram, M.Bhaskaran, R. B.Kaner, K.K.-Zadeh, *ACS Nano*, 6 (2012) 4045.
- [25] M. A Fakhri, M. J. AbdulRazzaq, A. A. Alwahib, W. H Muttlak, *Optical Materials* 109 (2020) 110363.
- [26] C.-C. Lee, C.-L. Tien, and J.-C. Hsu, *Applied Optics*, 41 (2002) 2043.
- [27] E. T. Salim, M. S. Al-Wazny, M. A. Fakhri, *Modern Physics Letters B*, 27(16) (2013) 1350122.
- [28] A. Dhar, T. L. Alford, *Journal of Applied Physics*, 112 (2012) 103113.
- [29] JM Taha, RA Nassif, NH Numan, MA Fakhri, *AIP Conference Proceedings* 2213 (1) (2020) 020235.
- [30] R. Georgiev, B. Georgieva, M. Vasileva, P. Ivanov, T. Babeva " *Advances in Condensed Matter Physics*, 2015 (2015) 3.

- [31] M. A. Muhsien, E. T. Salim, and I. R. Agool, International Journal of Optics, 2013 Article ID 756402, 9 pages (2013).
- [32] SH. Mujawar, AI. Inamdar, SB. Patil, PS. Patil, Solid State Ionics. 177 (2006) 3333.
- [33] RS Mahmood, W Abdulsatar, MA Fakhri, AIP Conference Proceedings 2213 (1) (2020) 020236.
- [34] R. Jose, V. Thavasi, S. Ramakhrisna, Journal of the American Ceramic Society. 92 (2009) 289.
- [35] R. A. Ismail, E. T. Salim, W. K. Hamoudi, Materials Science and Engineering C 33(1) (2013) 47-52.
- [36] F. Hashemzadeh, A. Gaffarimejad, R. Rahimi, Journal of Hazardous Materials, 286,64 (2015).
- [37] FG Khalid, AS Ibraheam, MA Fakhri, NH Numan, AIP Conference Proceedings 2213 (1) (2020) 020204.
- [38] L. Mitterhuber, E. Kraker, S. Defregger Energies, 12 (2019) 610.
- [39] J. A. Saimon. M. K. ABOOD, E. T. SALIM, Journal of Ovonic Research 15 (2) (2019) 109-115.
- [40] MA Fakhri, SFH Alhasan, NH Numan, JM Taha, FG Khalid, AIP Conference Proceedings 2213 (1) (2020) 020227.
- [41] A. M. Rabaa, J. B.-Ruíza, M. R. Joya, Materials Research, 19 (2016) 1980-5373
- [42] Ö. D. Coşkun¹, S. Demirel, Applied Surface Science, 277 (2013) 35.
- [43] MA Fakhri, MM Hassan, AIP Conference Proceedings 2213 (1) (2020) 020244.
- [44] E. T. Salim, J. Admon, M. K. Abood, Materials Research Express 6(4), (2019) 046420.
- [45] S. A. Zahra, J. Electron. Devices, 17 (2013) 1494-1499.
- [46] S. Basel, NH Numan, FG Khalid, MA Fakhri, AIP Conference Proceedings 2213 (1) (2020) 020228.
- [47] R. A. Ismail, B. G. Rasheed, E. T. Salm, Journal of Materials Science: Materials in Electronics 18(4) (2007) 397-400.
- [48] J. Nguu, B. Aduda, F. Nyongesa, R. Musembi, S. Njogu, and P. Mwathe, International Journal of Innovative Research in Advanced Engineering, 2(2) (2015) 2349-2163.
- [49] J. Sikula, J. Hlavka, V. Sedlakova, L. Grmela, P. Hoeschl, T. Zednicek, in Proceedings of the 17th Passive Components Symposium (CARTS Europe) (2003) 281-285.
- [50] SM Taleb, MA Fakhri, SA Adnan, AIP Conference Proceedings 2213 (1) (2020) 020234.
- [51] B. V. Zeghbroeck, University of Colorado, ISBN 10, 2007.
- [52] R. N. Singru, A. B. Zade and W. B. Gurnule Int. J. Chem. Sci 7 (2009) 1878-1884.
- [53] FG Khalid, AQ Raheema, ZS Alshakhli, MA Fakhri, AIP Conference Proceedings 2213 (1) (2020) 020229.
- [54] N. Bouhssira, S. Abed, E. Tomasella, J. Cellier, A. Mosbah, M. Aida, Applied Surface Science, 252 (2006) 5594-5597.
- [55] LZ Mohammed, MA Fakhri, AK Abass, AIP Conference Proceedings 2213 (1) (2020) 020231.