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Electrical conductivity, mobility and carrier concentration in Nb_2O_5 films: Effect of NH_4OH molarity

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ABSTRACT

This work presents the effect of NH_4OH molarities on the electrical characteristics of Nb_2O_5 films prepared using the precipitation method. Hall Effect measurements proved the formation of n-type Nb_2O_5 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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Keyword: Nb_2O_5 thin films; electrical characteristics; Hall Effect measurements; Resistivity - temperature measurements.

1. INTRODUCTION

Nowadays, there has been rising attention in materials representing metal-insulator transitions (MITs) on account of their promising implementations in electronic apparatus [1-3]. NbO_2 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].

Recently, Niobium Pentoxide (Nb_2O_5) 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 TiO_2.

Nb_2O_5 presents an inorganic semiconductor. Its bandgap ranging 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 WO_3 and MoO_3 [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 Nb_2O_5 variations into a brownish-grey color [10]. Nevertheless, Nb_2O_5 system is complex particularly, as slight aberrations from the particular stoichiometry in Nb_2O_5 affect powerfully the material's physical properties. As an example, a minor oxygen lack arrives at the conversion to n-type semiconducting from insulating behavior [26-28].

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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 Nb_2O_5 thin films have originated implementations in photonics for 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 Nb_2O_5 films depending on the capability to deposit rise fineness films employing comparatively unpretentious and low-cost procedures. Generally, Nb_2O_5 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

Nb_2O_5 suspension was prepared by adding HF and ammonium hydroxide to Nb_2O_5 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 1(A). The activation energy (Ea) was estimated using log σ vs. 1000/T curve, where its value could be found from the following equation (1) [44-46]:

$$Ea=0.86*slope \quad (1)$$

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 1(B).

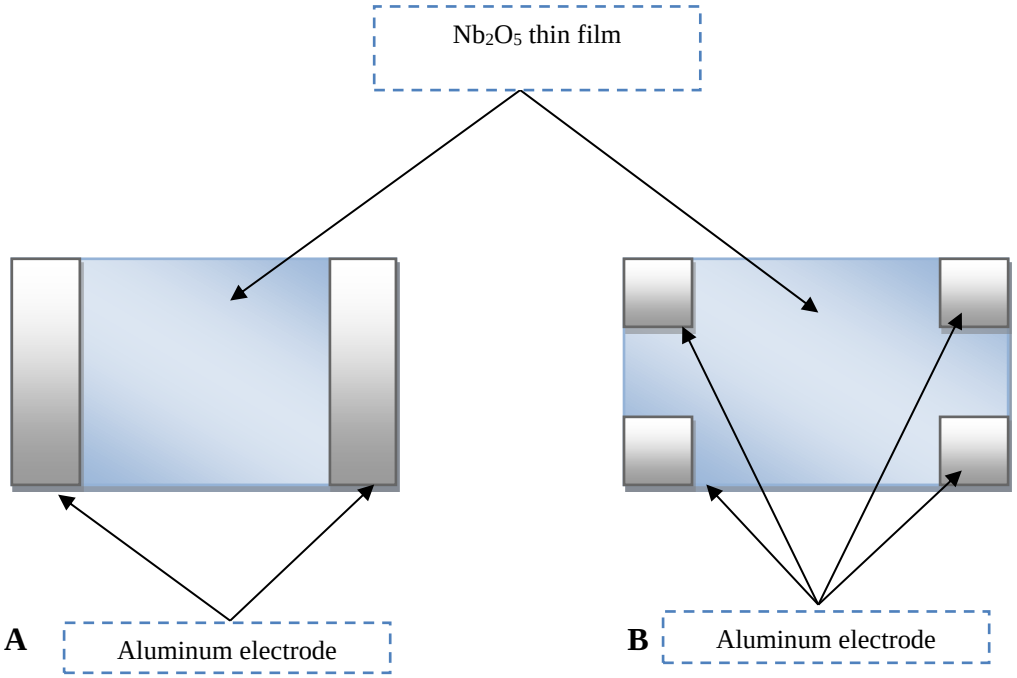

Figure 1. Aluminum electrode deposition for A: R-T; B: Hall effect measurements

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 O-2 (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 defect, 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].

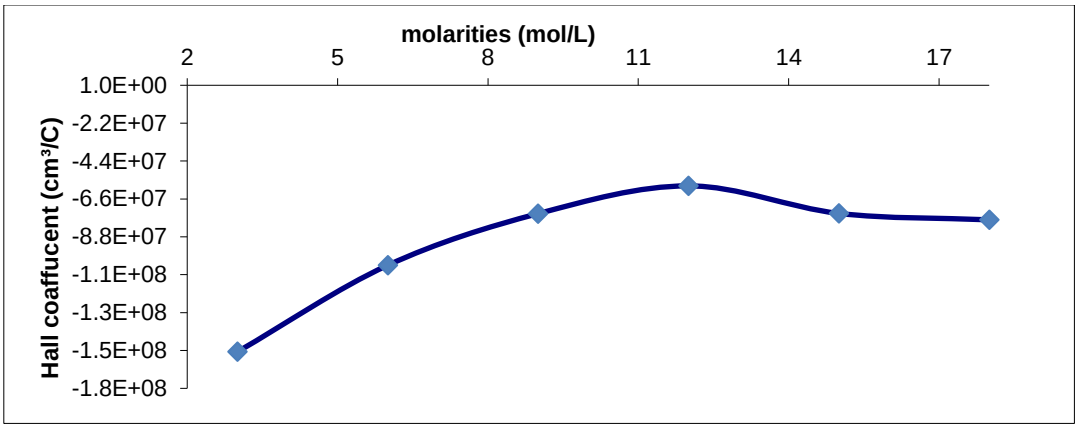

Figure 2. Hall coefficient for Nb_2O_5 films with the ammonium hydroxide molarities

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

$$n = \frac{1}{eR_H} \tag{2}$$

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 Nb_2O_5 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 Nb_2O_5 remaining a very small material amount within the suspension.

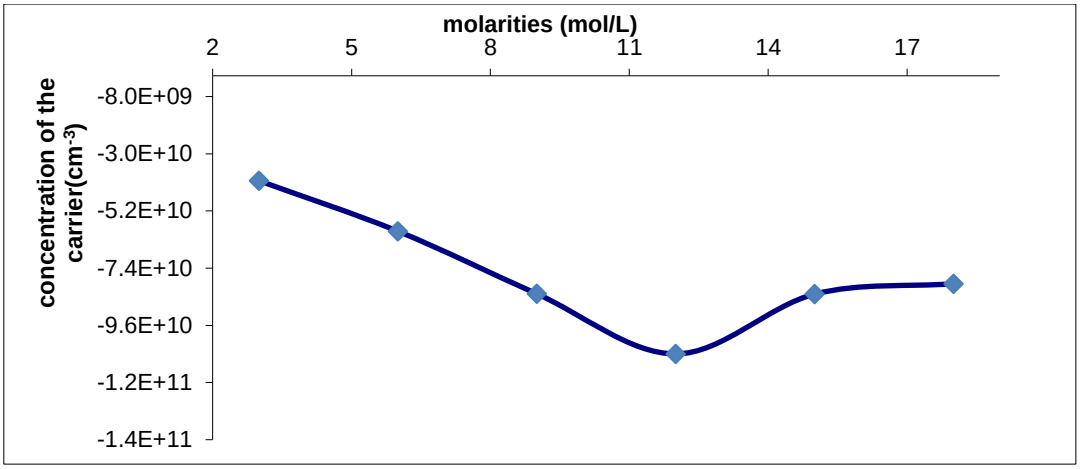

Figure 3. Nb_2O_5 films carrier cons. with the ammonium hydroxide molarities

Electrical mobility of the carrier affected also by the morality 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 Figure 4, similar results could be shown in other work [52, 53].

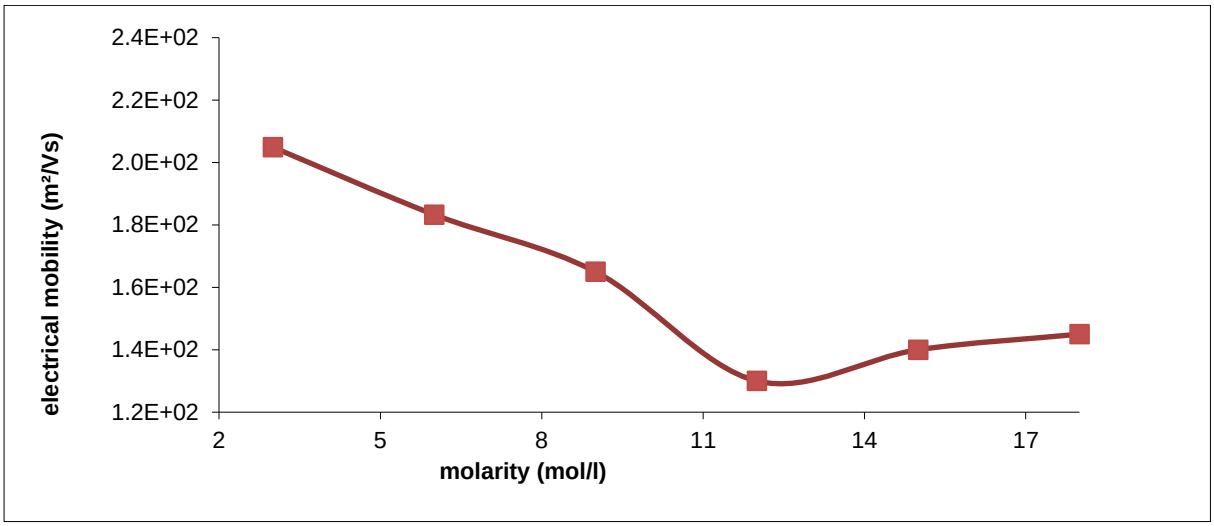

Figure 4. Electrical mobility for Nb_2O_5 thin films with the ammonium molarities

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.

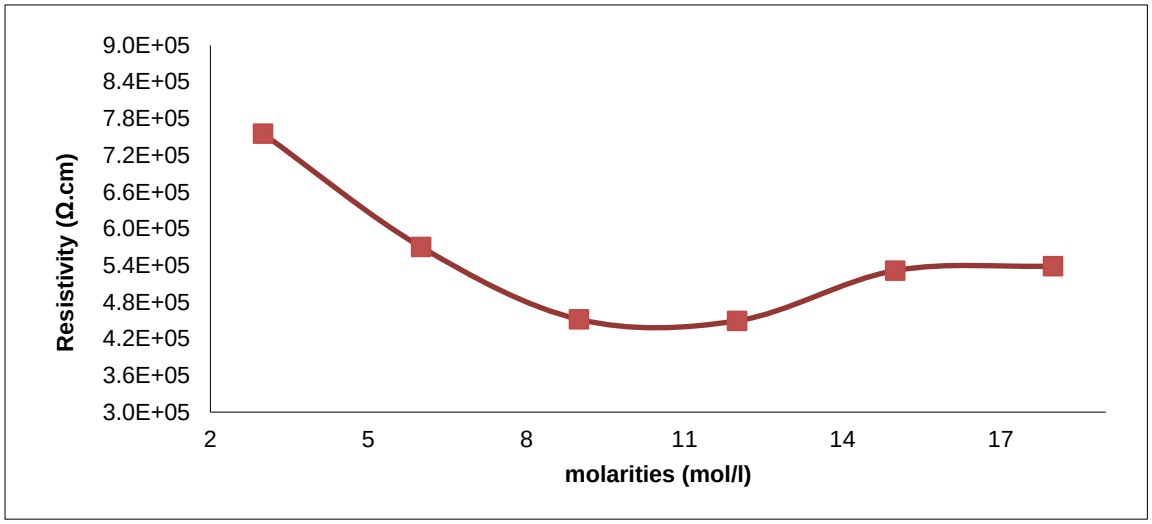

Figure 5. The electrical resistivity of Nb_2O_5 film with the ammonium molarities

Figure 6 shows the investigation study of the resistivity of the Nb_2O_5 film at a wide temperature range of (40-200C°) 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 decrease as the molarity of the ammonium hydroxide increase to reach minimum at 12mol/l; it increase back as ammonium molarity increase further beyond (12) mol/l.

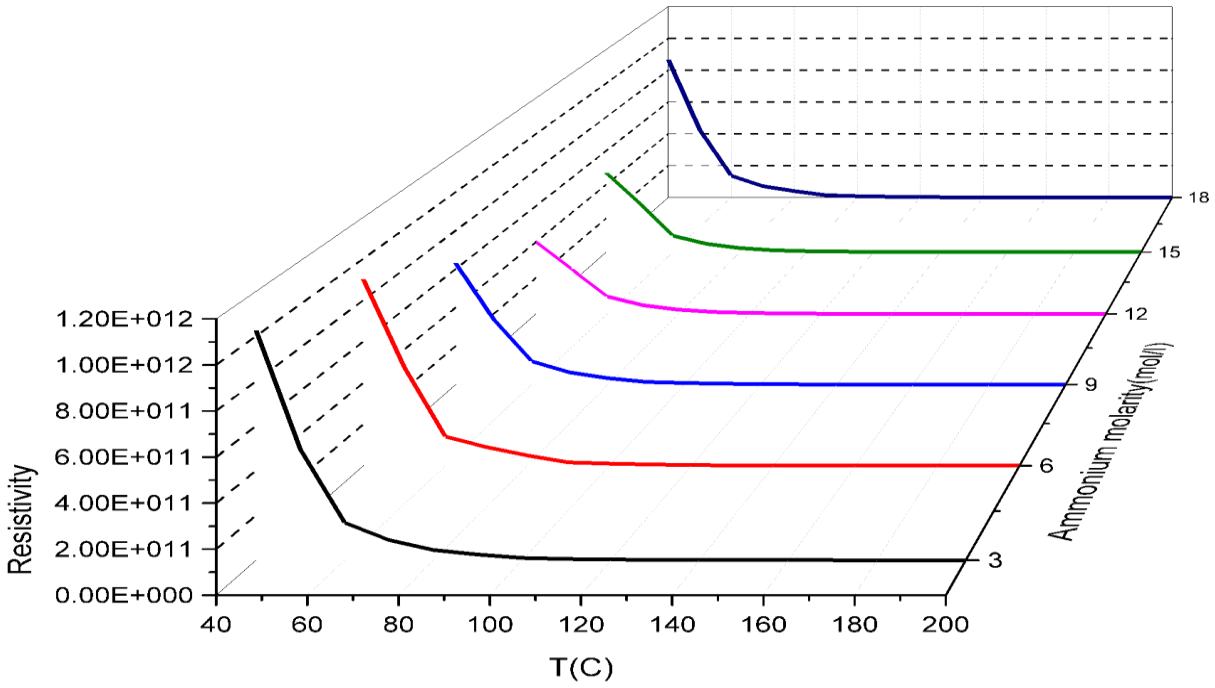

Figure 6. Resistivity of for Nb_2O_5 films with ammonium molarities at different temperatures

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. And resistance, in turn, is inversely proportional to D. It is interesting to point out that along with the decrease of

resistance. This is because, increasing the molarity of the precursor, the thickness (D) of the resulting film is also increased, and resistance, in turn, is inversely proportional to D.

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.

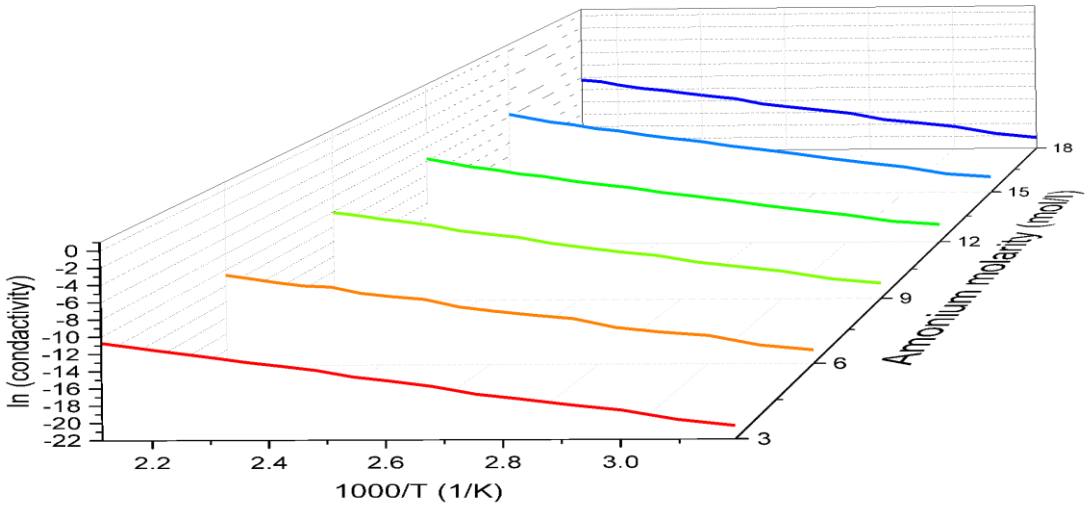

Figure 7. The conductivity with 1/T for Nb_2O_5 thin films at different molarities

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

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

where σ = electrical conductivity at specific temperature, Ea 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 Figure 7. Typical semiconducting behavior could be recognized. The calculated energy of activation is found in Figure 8 with the hydroxide molarities.

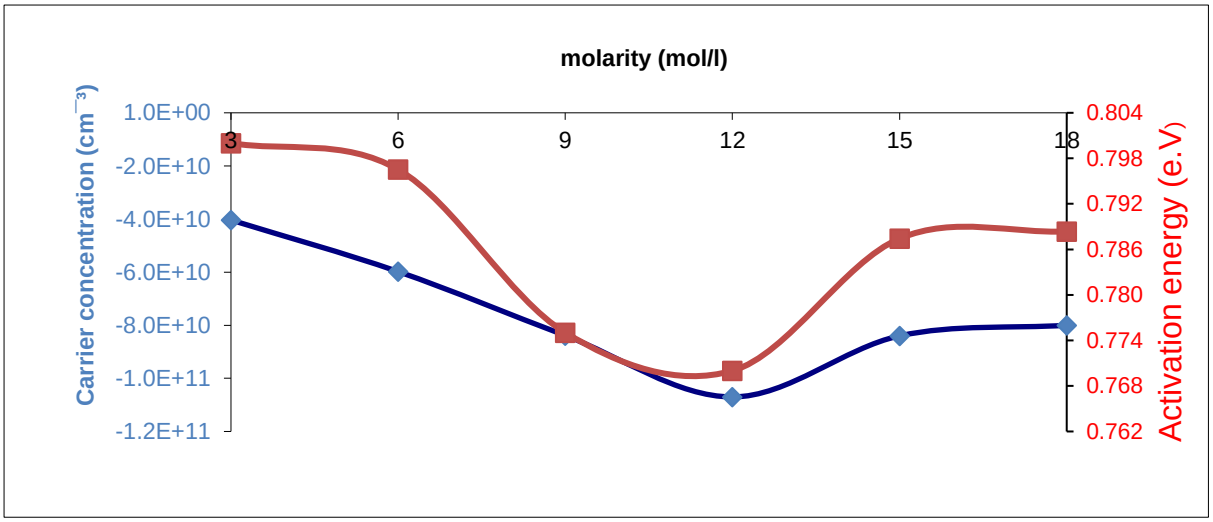

Figure 8. Activation energy, a carrier concentration for Nb_2O_5 thin at different molarities

The inverse relation between the energy of activation and hydroxide molarities up to 12mol/L could be recognized 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]:

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

where n is the free carrier concentration (electrons), N_C is the effective electrons concentration at the conduction band, E_C and 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 Nb_2O_5 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.

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