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Electrical and impedance studies of $(MoO_3)_x$ – $(V_2O_5)_{1-x}$ for x= 0, 0.2, 0.4, 0.6, 0.8 Composites

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composite bulk samples, thermal evaporation, ac, dc electrical characteristics, impedance characterization.

ABSTRACT

$(MoO_3)_x$ – $(V_2O_5)_{1-x}$ pellets were prepared with the composition of x = 0, 0.2, 0.4, 0.6, 0.8 and these pellets are coated with silver by thermal evaporation technique with an lower oxygen partial pressure of $2x10^{-5}$ mbar. The ac, dc electrical characteristics were studied. The dc electrical conductivity of the results showed that V_2O_5 decreases with increasing MoO_3 in$(MoO_3)_x$-$(V_2O_5)_{1-x}$, is attributed to the increasing concentration of oxygen vacancies and vanadium interstitials. The variation of a.c. conductivity with frequency of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x=0, 0.2, 0.4, 0.6, 0.8. At lower frequencies conductivity varies slowly and at high frequencies the curves approximate to a square law. The impedance characteristics were studied for $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x=0, 0.2, 0.4, 0.6, 0.8*pellets and both sides of pellets were coated with the silver. The impedance in V_2O_5 is purely due to the bulk grains and decreasing with increasing temperature. Which indicates that the increase in grainsize with increasing temperature.*

1. INTRODUCTION

Transition metal oxides with layered type of structures have a strong anisotropy in most of its structural and physical properties. These metal oxides possesses either two-dimensional Vander Waal's bonded layered structure (V_2O_5, MoO_3 etc.) or three dimensional frame work tunnel structures (V_6O_{13}, $LiCoO_2$ etc.). The versatility of these compounds is exploited in traditional applications such as electric switching (varistors), temperature switching (thermistors), chemical processing (catalysts) and electronic circuits (capacitors, insulators, resistors) [1]. Vanadium oxide is widely used in elctrochromic, photochromic and thermo chromic devices. MoO_3 also exhibit photo chromic, thermo chromic and gasochromic properties. The interaction between V_2O_5 and MoO_3 is unique due to their similar ionic radii and nearly- identical structures in their higher oxidation state. MoO_3 - V_2O_5 have been used as catalysts for the selective oxidation of benzene and other hydrocarbons [2]. The present investigations are undertaken to study the electrical properties of V_2O_5 and $(MoO_3)_x – (V_2O_5)_{1-x}$ composites.

II. EXPERIMENTAL

Pure V_2O_5 powder (purity of 99.96 %) and MoO_3 powder (purity of 99.99 %) SIGMA ALDRICH is used to prepare different composites of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x=0, 0.2, 0.4, 0.6, 0.8. Various compositions of MoO_3, V_2O_5 powders have been taken in the mortar and grind well to get fine powders of the mixed oxide and they were sintered at 450 °C. For the preparation of pellets for both electrical and impedance studies. $(MoO_3)_x$ -$(V_2O_5)_{1-x}$ composite pellets (with $0 \le x \le 1$) were deposited by silver with thermal evaporation in a vacuum coating unit supplied by vacutech systems. The vacuum coating unit was initially evacuated to a base pressure of 5 X 10^{-5} mbar. Then argon gas was introduced into the chamber to clean the chamber. Oxygen gas introduced into the chamber and the flow was controlled by needle valve. The Molybdenum boat was used to evaporate the material. The films were deposited at a substrate temperature of 300K and pressure is fixed at 2 x 10^{-4} mbar.

III. RESULTS AND DISCUSSION

AC Electrical Conductivity

Electrical conduction in transition metal oxides can take place by two parallel processes: (1) by band conduction and (2) by hopping conduction in the localized states. The former tends to occur at higher temperatures, where carriers excited beyond the mobility edges into non-localised states dominate the transport, while the latter may be due to carriers excited into localized state band edges [3].

It is well known that the total measured a.c. conductivity, $\sigma_{a.c.}$, may be represented in a wide variety of amorphous semiconductors and insulators by the experimental relation

$$\sigma_{a.c.} = \sigma_{total} – \sigma_{d.c.}$$

where $\sigma_{d.c.}$ is the d. c. conductivity and $\sigma_{a.c.}$ is the true a.c. conductivity which is frequency, f, dependent. The fig. 1 shows the variation of a.c. conductivity with frequency of $(MoO_3)_x$-$(V_2O_5)_{1-x}$ for x=0, 0.2, 0.4, 0.6, 0.8.

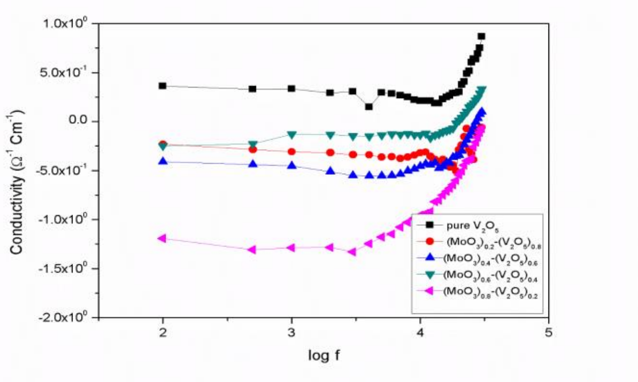

Fig. 1 AC electrical conductivity of $(MoO_3)_x$- $(V_2O_5)_{1-x}$for x=0, 0.2, 0.4, 0.6, 0.8.

At lower frequencies conductivity varies slowly and at high frequencies the curves approximate to a square law. The a. c. conductivity obeys the power frequency law [4]

$$\sigma_{a.c.} \alpha\ \omega^s$$

The a.c. conductivity is decreased with increasing MoO_3in $(MoO_3)_x$-$(V_2O_5)_{1-x}$.

DC Electrical Conductivity

Electrical conduction in transition metal oxides is due to the electron hopping mechanism between the oxidation states of the metal atoms. The V^{4+} and V^{5+} are the two centres responsible for the electrical conduction in V_2O_5 [5] Under the application of an electric field the barrier between V^{4+} and V^{5+} centres may be lowered and the transfer of charge between V^{4+} and V^{5+} centres may be increased [6]. Fig. 2 gives the plot of ln σ vs 10^3/T of $(MoO_3)_x$-$(V_2O_5)_{1-x}$ for x=0, 0.2, 0.4, 0.6, 0.8. The d.c. electrical conductivity of V_2O_5 decreases with increasing MoO_3 in $(MoO_3)_x$-$(V_2O_5)_{1-x}$, is attributed to the increasing concentration of oxygen vacancies and vanadium interstitials [7].

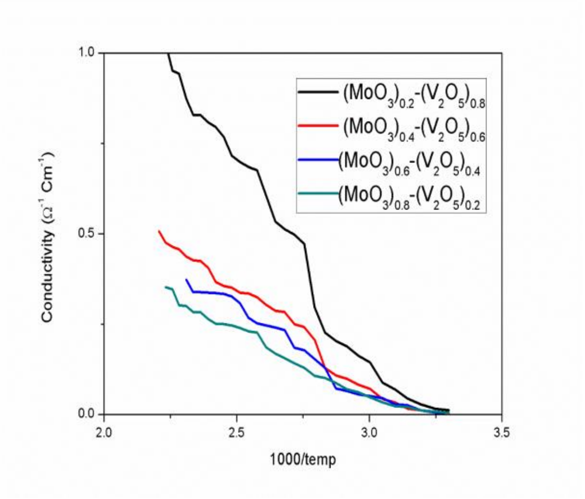

Fig. 2 DC electrical conductivity of $(MoO_3)_x$- $(V_2O_5)_{1-x}$ for x=0, 0.2, 0.4, 0.6, 0.8.

The number of V^{4+} ions is induced by the presence of Mo^{6+} ions and decrease with increase in the MoO_3 content. The interference of the non-periodic coulmbic field produced by Mo^{6+} will dominate with increase in molybdenum content in V_2O_5. In turn, this interference may lead to decrease in the number of V^{4+} ions when compared with the number of Mo^{6+} ions which is responsible for decrease in electrical conductivity with increase in Mo concentration of $(V_2O_5)_{1-x}$ - $(MoO_3)_x$composite. More over the electron density in d band decreases with increasing MoO3 content [8].

Another reason is due to increase in MoO_3 content the grain size of V_2O_5 crystallites in the composite is decreasing which leads to the decrement in electrical conductivity and enhancement in the ionic conductivity. The interference of the Coulomb fields may not be significant at lower concentration of molybdenum in $(V_2O_5)_{1-x}$-$(MoO_3)_x$composite.

Impedance

Impedance spectroscopy is one of the powerful tools to analyse and separate the contributions of grains, grain boundary and interfaces of the material to the electrical properties over a wide frequency range [9]. Impedance spectra of the materials can be explained using Cole-Cole or Nyquist plots where real part of impedance (z^1) was plotted against imaginary part of impedance ($-z^{11}$).The plots consists of one or two semicircles depending on the material properties. A spike at very low frequencies corresponds to grain boundary contribution and at high frequency corresponds to bulk grains contribution to the impedance. Material can be treated as series combination of grain and grain boundary.The grain or grain boundary can be imagined as parallel combination of resistance and capacitance [10].

The cole - cole plots of pure V_2O_5 *and* $(MoO_3)_x$ – $(V_2O_5)_{1-x}$ *composites with* $0<x<1$ *for various temperatures were shown in figs.* 3.1 to 3.9.

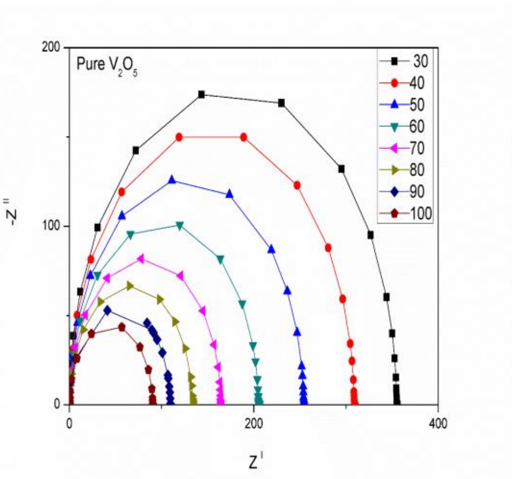

Fig. 3.1 Impedance spectra of Pure V_2O_5at different temperatures

Fig. 3.1 shows the semicircle arcs of V_2O_5 huge pattern [12]changes but not its shape when the temperature is increased. It is observed that the real part of impedance is decreasing with increasing the substrate temperature. Moreover in the present case the semicircle corresponding to low frequency were not present. So this indicates that the contribution of electrodes and their grain boundaries of impedance is almost nil. The impedance in V_2O_5 is purely due to the bulk grains and decreasing with increasing temperature. Which indicates that the increase in grainsize with increasing temperature.

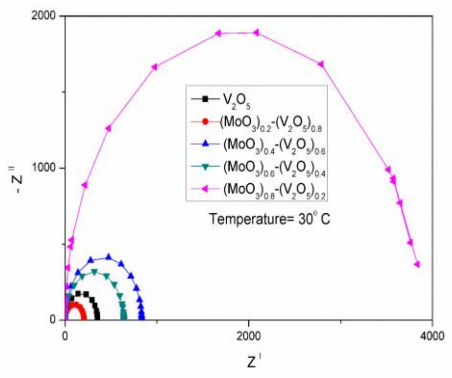

Fig. 3.2 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 30 ^{0}C.

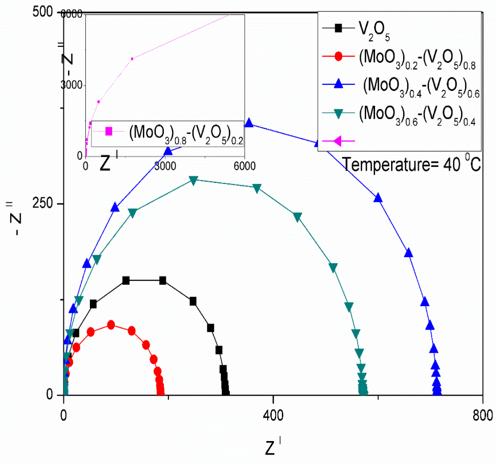

Fig. 3.3 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 40 ^{0}C.

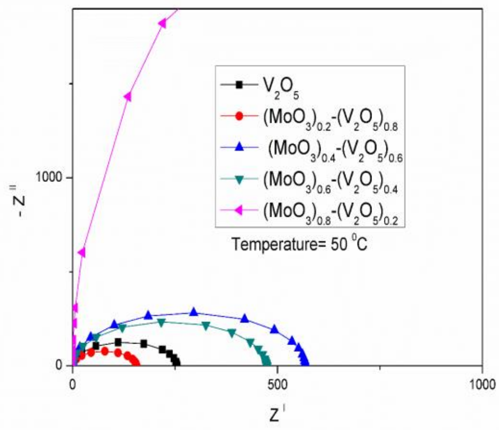

Fig. 3.4 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 50 ^{0}C.

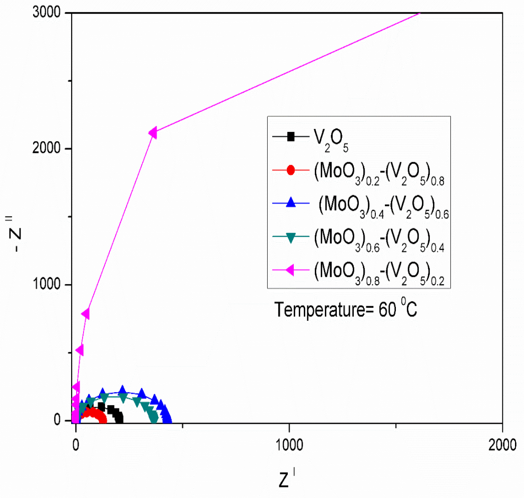

Fig. 3.5 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 60 ^{0}C.

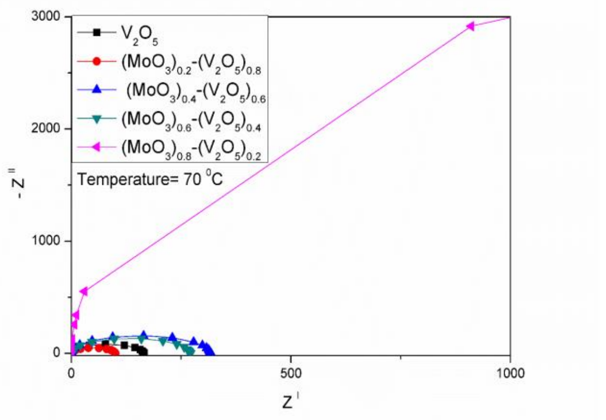

Fig. 3.6 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 70 ^{0}C.

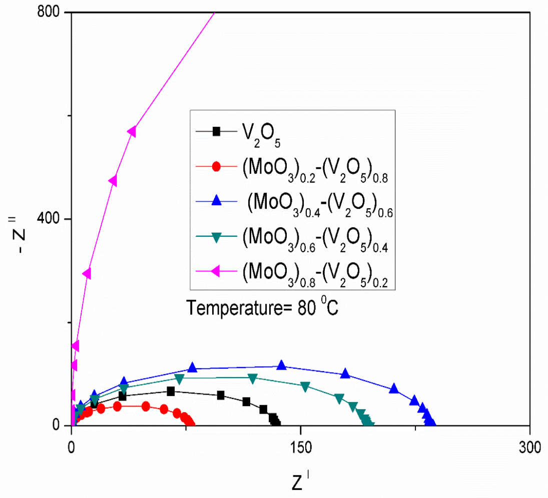

Fig. 3.7 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 80 ^{0}C.

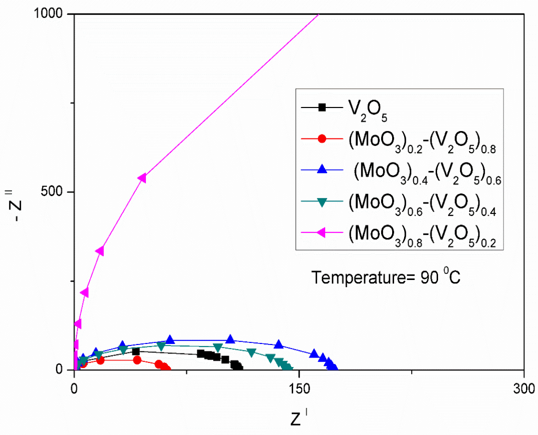

Fig. 3.8 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 90 ^{0}C.

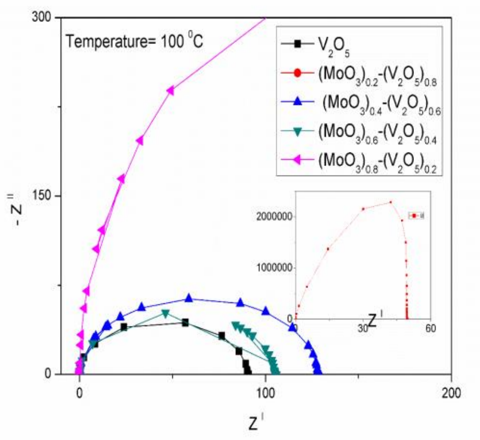

Fig. 3.9 Impedance spectra of $(MoO_3)_x$ - $(V_2O_5)_{1-x}$ for x = 0, 0.2, 0.4, 0.6, 0.8 at a temperature of 100 ^{0}C.

The equivalent circuit configuration for the impedance plan plot is the resistance R_P (bulk resistance) and in the terms of complex elements: constant phase elements (capacity of the fractal interface CPE). The impedance of CPE is

$$Z_{CPE} = \frac{1}{Q(jw)^{\alpha}} \text{------ (1)}$$

where Q indicates the value of capacitance of the CPE element and α is the fractal exponent. Further, (1) shows that as $\alpha \rightarrow 1$, ZCPE → 1/jQω, involving a pure capacitance, and as $\alpha \rightarrow 0$, $Z_{CPE} \rightarrow 1/Q$, a pure conductance. The real and imaginary components of the whole impedance of this circuit were calculated according to the following expressions:

$$Z^I = \frac{[R_P^2 Q W^{\alpha} \cos\left(\frac{\alpha\pi}{2}\right) + R_p]}{[\left(1 + R_P^2 Q W^{\alpha} \cos\left(\frac{\alpha\pi}{2}\right)\right)^2 + \left(R_P^2 Q W^{\alpha} \sin\left(\frac{\alpha\pi}{2}\right)\right)^2]} \text{--(2)}$$

$$-Z^{II} = \frac{[R_P^2 Q W^{\alpha} \sin\left(\frac{\alpha\pi}{2}\right)]}{[\left(1 + R_P^2 Q W^{\alpha} \cos\left(\frac{\alpha\pi}{2}\right)\right)^2 + \left(R_P^2 Q W^{\alpha} \sin\left(\frac{\alpha\pi}{2}\right)\right)^2]} \text{ ----(3)}$$

From fig 3. 2 for x = 0.2 at room temperature the impedance is 208. It is decreasing when compared to V_2O_5 which is due to the increment in the grain size and this behaviour is not fixed and showing oscillating variations with increasing MoO_3 content[11]. The impedance is further decreasing with increasing temperature which is due to the enhancement in the grain size. Further increasing the temperature to 50 ^{0}C the similar behaviour in case of pure and composites was observed except for high content of MoO_3. Moreover it is observed that the shape of the impedance curve changes from perfect semicircle to enlarged semicircle. So, in all cases at high temperatures, it is observed that for x = 0.8 the semicircle is changes to linear. At 100 ^{0}C it was observed that in case of x = 0.2 the imaginary part ($-Z^{11}$) is increasing enormously.

IV. CONCLUSION

The a.c. and d.c. electrical conductivities were decreased with increasing MoO_3in $(MoO_3)_x$-$(V_2O_5)_{1-x}$.Impedance properties f*or x = 0.2* at room temperature is 208. It is decreasing when compared to V_2O_5 which is due to the increment in the grain size and this behaviour is not fixed and showing oscillating variations with increasing MoO_3 content. The impedance is further decreasing with increasing temperature which is due to the enhancement in the grain size. So, in all cases at high temperatures, it is observed that for x = 0.8 the semicircle is changes to linear.

Conflict of interest statement

Authors declare that they do not have any conflict of interest.

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