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Azo Dyes on the ligand(5-MeTAQ)Thin Films for Dye Sensitized Solar Cells Applications

Zeina mohammed kadam¹, Raheem Abed Jeber², Alaa Hussein Ali³

University of Al-Qadisiyah ,college of science¹
University of Al-Qadisiyah ,college of Dentistry²
University of Al-Qadisiyah , Diwaniya³

alaa61468@gmail .com

Abstract

This Include search the preparation of a new ligand from heterogeneous azo organic derivatives derived from Thiazolyl ligand (5-MeTAQ) and the identification of ligand (5-MeTAQ) were identification and analyzed by using ¹H-NMR, mass spectrum , XRD SEM and EDX. Thin films of ligand dye prepared of concentration of (0.05,0.1,0.3) M membrane pure and doped of 10% hexaferrite (SrFe₁₂O₁₉) of concentration (0.05M) was prepared of thin films with thickness (1000 ±15) nm the preparation of membranes by method spray pyrolysis. The study of optical properties of thin films pure and doped study spectral of absorbance and transmittance within the wavelength (200-800)nm the Results shows of the transmittance decrease of the molar concentration Increase and doped and the absorbance increase with increase molar concentration and doped and energy band gap decrease with increase molar concentration and doping the study of structure properties of the Thin films prepared through(XRD) where the results show membranes multiple crystallization and preferred trend of growth is (101) particle.

Keywords: Thiazolyl ligand(5-MeTAQ) , Spray pyrolysis, Strontium hexaferrite

1. Introduction

Thiazolyl azo compounds and their derivatives are compounds that are frequently used as organic reagents in spectrophotometry they also have applications in the medical and industrial fields this type of ligand contains an heterogeneous pentagonal ring in which two different atoms which are sulfur and nitrogen(1) The term thiazole, Also refers to a wide family of derivatives and its noted that the thiazole ring is a component of vitamin (B₁). Thiazole azo compounds and their derivatives are widely used as organic reagents And in the fields of solar energy (2). They are commonly used as reagents because of their properties and multi –use properties due to their high stability and rapid reaction with metal ions as well as their high sensitivity and selectivity,(3) azo Thiazole compounds are characterized by being heterogeneous compounds and contain nitrogen and carbon catalyst that contribute to the alignment with the transition element,(4) one of the most important uses of azo Thiazole compound is use in spectral mapping to estimate the very small quantities of transitional element as such thiazolyl compounds are use in pharmaceutical preparation,(5) such as anti fungal agents , the hetrocyclic of azo dye imidazole compounds of the important in in spectral



of determination of filed the trace amount elements of metal ions because of azo dye high selectivity,(6) In the research the preparation and spectroscopy of the new heterocyclic new azo dye ligand (5-MeTAQ) the present study report the preparation and spectral characterization of new azo imidazole ligand(5-MeTAQ) the term thin films is used to describe layer or several layers of atoms of ascertain substance whose thickness less than (1 μ m) thin film application in electronic resistances, (7) transistor and solar The thin films were prepared by thermochemical spraying method under optimum condition such as the distance between the spraying head and hot substrate is (30cm) the compressed air pressure was maintained at 4.5 Kg/cm².

The pressure adjustment process was done at 4.5 Kg/cm² and deposition This technique was used for the spraying process is versatile technique for deposition of ligand (5-MeTAQ) because of its cheapness and process control gives the films,(8).

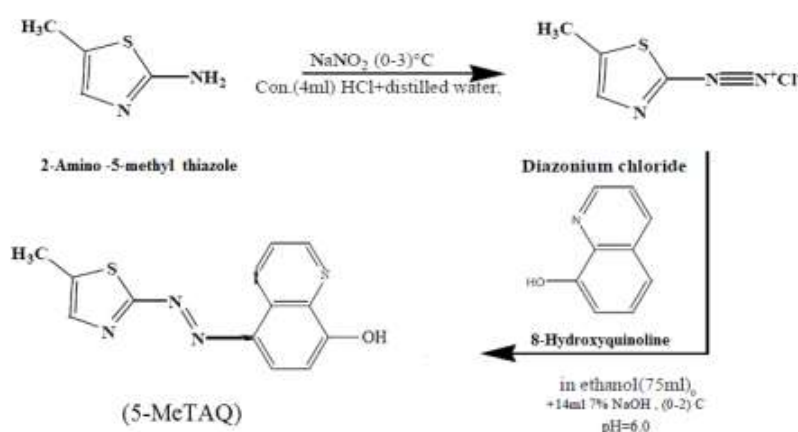
2- Experiment

2-1 Chemicals and method

Chemicals used High purity chemicals have been used the work Are the 2-amino-5-methyl thiazole, 8-hydroxy quinole, NaOH HCl, NaNO₂ and Strontium hexaferrite SrFe₁₂O₁₉ produced by (Sigma Fluka and Aldrich) company , In addition to use of ethanol and DMSO as a solvent azo dye ligand (5-MeTAQ) characterized by analytical data. ¹H-NMR spectra were recorded in DMSO-d₆ on Bruker 300 MHz spectrophotometer using TMS as an internal Reference ,mass spectrum was obtained using shimadzu Agilent Technologies, x-ray diffraction (XRD) technique using a shimadzu x-ray diffractometer with CuK_α($\lambda=1.5418\text{\AA}$) radiation for 2 θ values in the range of 10–80 $^{\circ}$,the electronic spectra of ligand and thin film ligand were Recorded on a shimadzu double beam UV-Vis spectrophotometer the range of (200-1100)nm in absolute ethanol solution , scanning electron microscopy (SEM) images of azo dye ligand using micrograph zeiss em 3200,energy dispersive-x-ray (EDX) of azo dye ligand .

2-2 Synthesis of ligand novel (5-MeTAQ)

Ligand was prepared above depending on the method suggested by Researchers(9) with some 1,2g (0.01mol) of 2-amino-5-methyl thiazole dissolved in a mixture of 5ml hydrochloric acid (30 ml) distilled water and 5ml ethanol the mixture with continuous stirring and not temperature above (5 $^{\circ}$ C) the mixture added to(0.9g) Sodium nitrite dissolved to 30ml distilled water added drop wise at (0-5 $^{\circ}$ C) The continuous rotation process is done with for (25min) the added of diazonium salt solution with continuous added drop wise with cooling at (0-5 $^{\circ}$ C) into 0.9g (0.012mol) of 8-hydroxy quinole was dissolved in mixture 50ml ethanol and 10 ml Sodium hydroxide . for coupling after had been stirring two hour at (0-5 $^{\circ}$ C) to PH=6.0 the precipitate



Scheme : (1)synthesis of azo dye ligand (5-MeTAQ)

2-3 Preparation of ligand novel (5-MeTAQ) thin films

Ligand thin films pure from a solution with different molar concentration (0.05M,0.1M,0.3M) in 100ml of deionized water and thin film distortion of 10% Strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) of concentration (0.05M) thin films were prepared by spray pyrolysis, The solution is sprayed at a rate of spraying (1 ml) every minute on a heated glass substrate at (130 ° C) using a compressed air device as a carrier gas, so that the nozzle to the substrate distance is about (45 cm) and the number of sprays (10) and when the time is stopped (1s).

3- Result and discussion

3-1 H-NMR Spectra of ligand novel (5-MeTAQ) ¹

In the ¹H-NMR of azo dye ligand (5-MeTAQ) used solvent DMSO and TMS internal reference the ¹H-NMR of ligand shows a signal peak back to solvent DMSO $\delta=2.523\text{ppm}$, singlet due to **H7** into benzene ring $\delta=7.427\text{ppm}$, singlet due to **H10** into benzene ring $\delta=7.561-7.590$, signal due to **H9** into benzene ring $\delta=7.739-7.747\text{ppm}$, signal due to **H5** into thiazole ring $\delta=7.767-7.775$ ppm signal due to **H4** into thiazole ring $\delta=7.837-7.845\text{ppm}$, singlet peak due to **H1** into thiazole ring $\delta=7.011\text{ppm}$, singlet due to OH $\delta=13.191\text{ppm}$. (10)

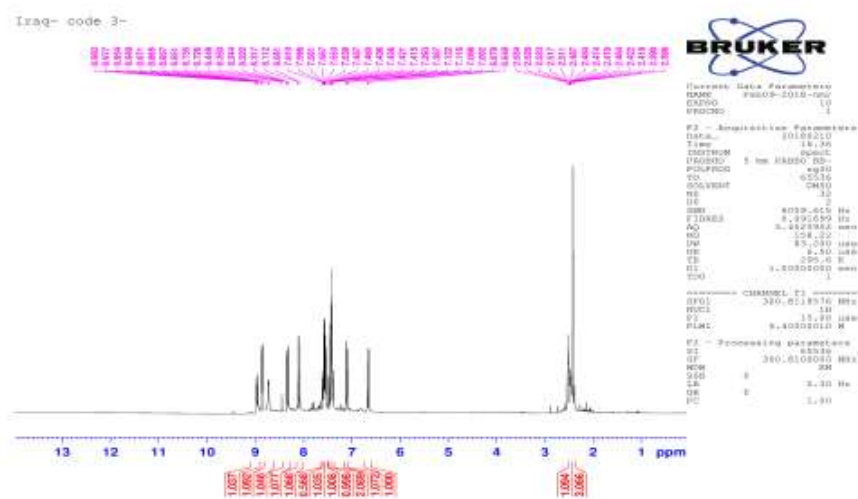


Figure.(1) ¹H-NMR spectrum of ligand (5-MeTAQ)

3-2 Mass spectra of ligand novel (5-MeTAQ)

Recorded of mass spectrum of ligand (5-MeTAQ) and molecular ion peaks the mass Spectra give signals when of ligand(5-MeTAQ) showed peaks to the molecular ions m/z^+ at.(11,12)

Table (1): Ligand State Mass Fragmentation Products (5-MeTAQ)

Fragment	m/z^+ Exact mass	Relative Abundance (%)
$[\text{C}_{13}\text{H}_{10}\text{N}_4\text{OS}]$	271.0	54.0
$[\text{C}_{13}\text{H}_{10}\text{N}_2\text{OS}]^+$	243.0	84.4
$[\text{C}_{11}\text{H}_7\text{N}_2\text{OS}]^+$	213.0	79.0

$[\text{C}_{10}\text{H}_4\text{N}_2\text{O S}]^+$	165.1	100
$[\text{C}_4\text{H}_4 \text{N}_2\text{OS}]^+$	129.1	2.0
$[\text{C}_3\text{H}_3\text{NS}]$	87.0	6.7
$[\text{C}_5\text{H}_3]^+$	64.0	2.8

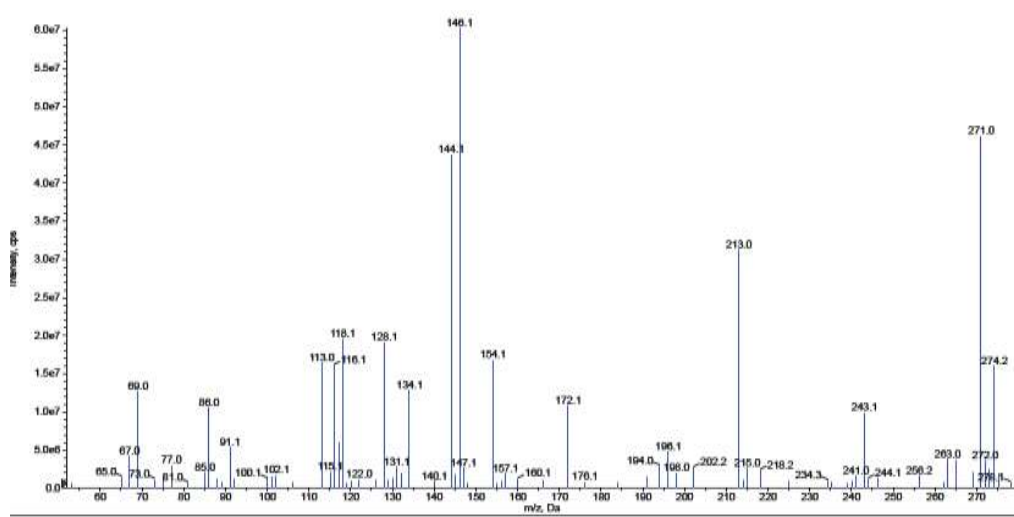


Figure.(2).mass spectrum fragmentation of ligand (5-MeTAQ)

3-3 Energy –dispersive -X-ray spectroscopy of ligand (5-MeTAQ)

For the element analysis or chemical characterized of ligand (13)

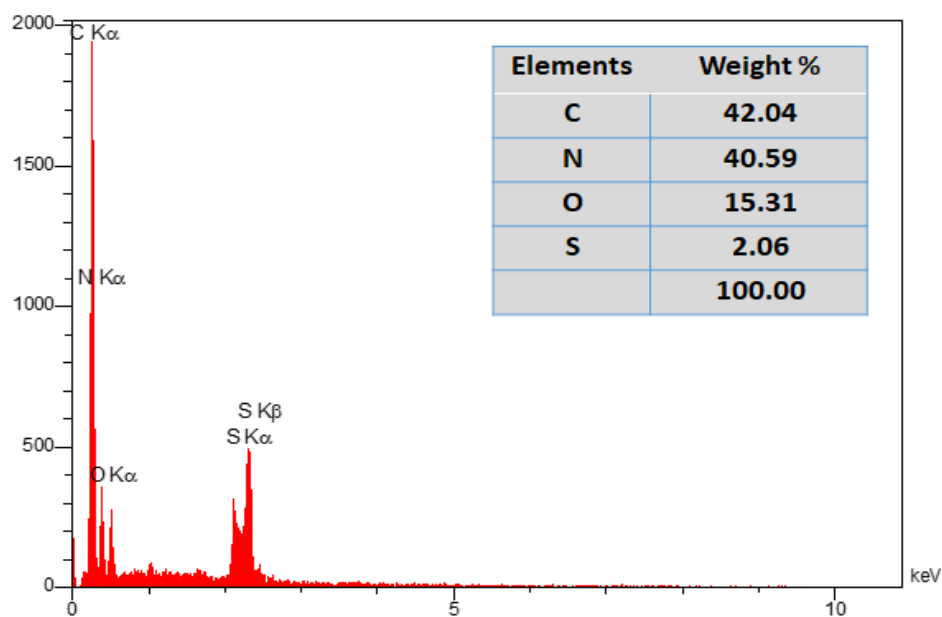


Figure (3). EDX of azo dye ligand (5-MeTAQ)

3-4 SEM analysis

The properties of the ligand novel (5-MeTAQ) like of surface morphology distribution of particles aggregation and shape of the particle study by scanning electron microscopy (SEM), of the ligand(5-MeTAQ) have Particles are differentiated with average size (35)um ,(14)

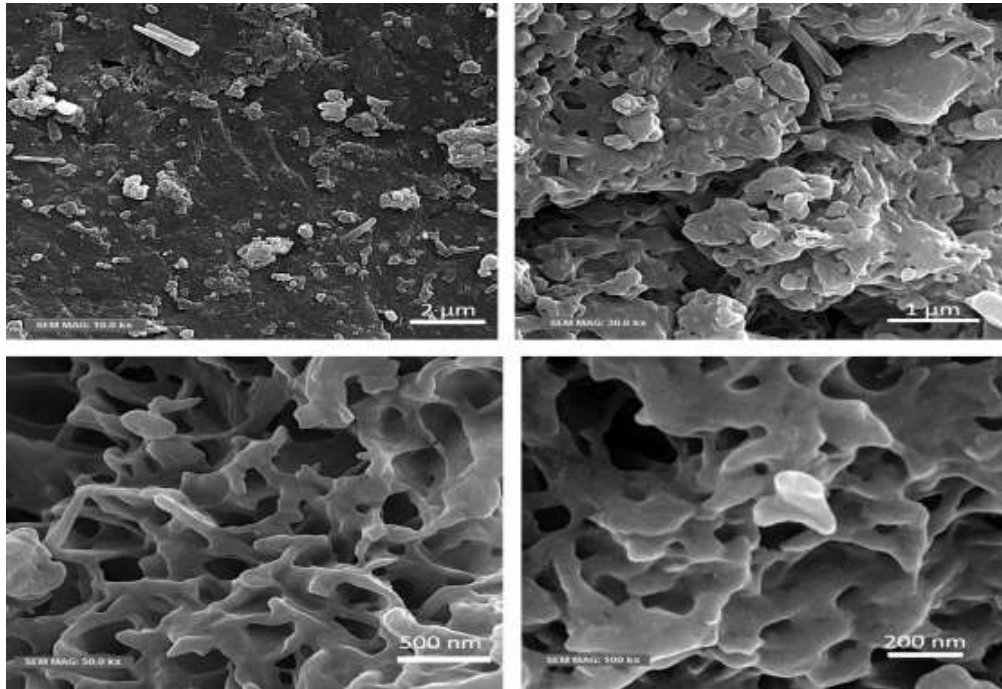


Figure (4): SEM of the ligand novel (5-MeTAQ)

3-5 X-ray Structure properties of thin films of ligand(5-MeTAQ)

The diffraction patterns of ligand(5-MeTAQ) films deposited at (0.05,0.1and 0.3)M and doping 10% hexaferrite of concentration (0.05M) precursor concentration at substrate temperature (130°C) in nature with (101) preferred orientation ,(15) the crystallite size (D) of X-ray using Debye –Scherrer's equation[1]:

$$D = \frac{k\lambda}{\beta \cos \theta}$$

D = Grain size in a particular orientation

λ = X-ray wavelength

θ = Diffraction angle corresponding to the particular orientation

$\beta \cos$ = Width at Half Maximum intensity (FWHM)

Table: (2) the crystallite size, FWHM ,hkl and 2theta of thin films of ligand (5-MeTAQ)

Molar Concentration (M)	2theta (degree)	FWHM (degree)	Crystallite Size D (nm)	hkl
0.05M	25.3	0.394	112	101
0.1 M	24.3	0.358	132	101
0.3M	26.2	0.325	143	101
0.05M Doping Strontium hexaferrite $\text{SrFe}_{12}\text{O}_{19}$	23.9	0.376	156	101

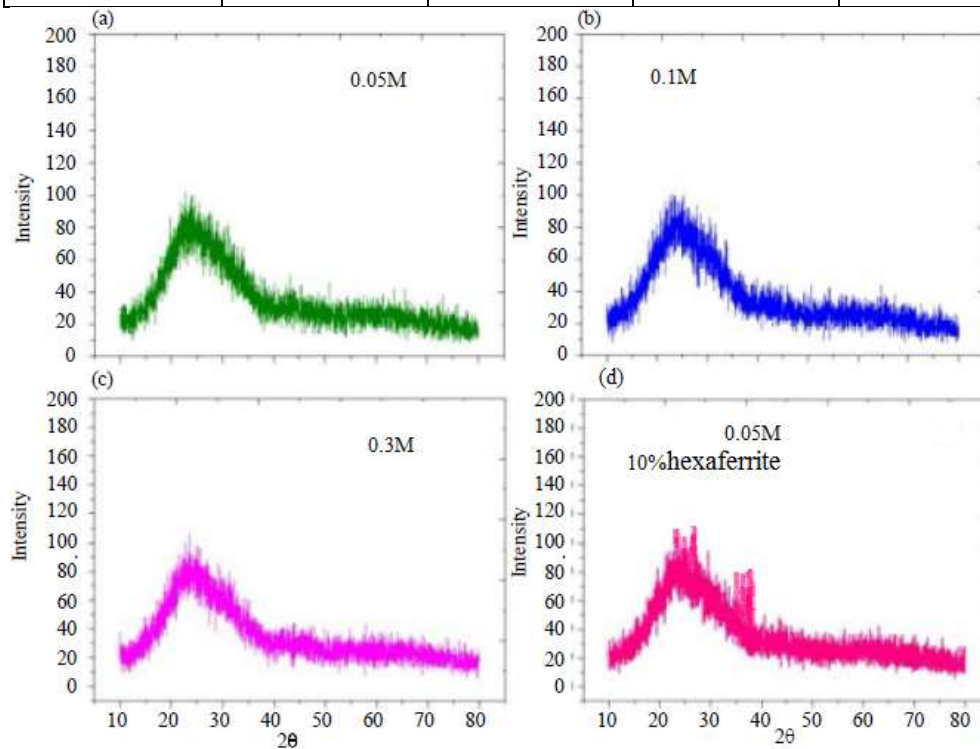


Figure:(5): the (XRD) thin films of ligand (5-MeTAQ)

3-6 Optical properties of thin films of ligand (5-MeTAQ)

Transmission and absorbance curves of ligand (5-MeTAQ) thin films recorded as a function of wavelength in the range (200-1100)nm ,the transmission goes down from 90% to 70% when concentration in creased from (0.05 to 0.3) M and doping 10% hexaferrite , all the films are high absorbance in UV :the reduction of the transmission at high molar concentration may be attributed to the increased scattering of photons by increased of the roughness of the surface morphology .(16)

The optical band gap was determine using equation(2)

$$\alpha h\nu = B(h\nu - E_g)^{1/2}$$

$h\nu$: the photon energy

E_g : the optical band gap

α : absorption coefficient

B : constant

$h\nu$ is the photon energy.

The energy band gap decreased with increased molar concentration and doping 10% hexaferrite ,the energy band gap decrease from (3.19 to 3.0 21) ev , the band gap decrease with increase molar concentration and doping due to decrease in strain values has been demonstrated earlier the correlation between the direct band gap and the compressive stress of the ligand thin films,(17)

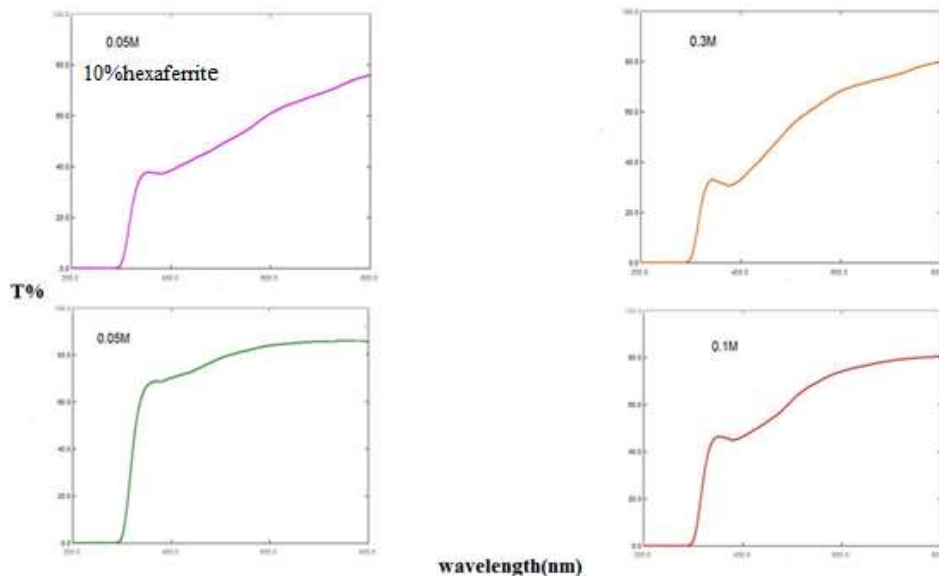


Figure (6).the transmission spectral thin films of ligand(5-MeTAQ)

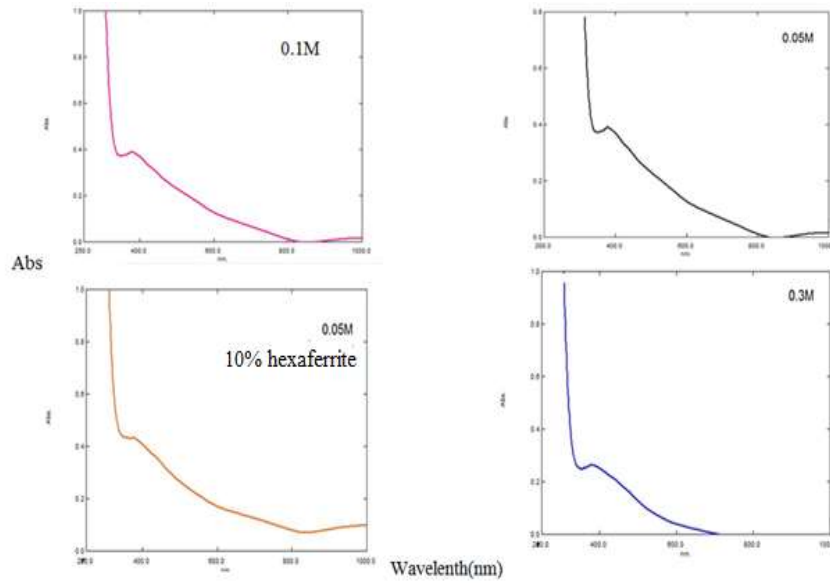
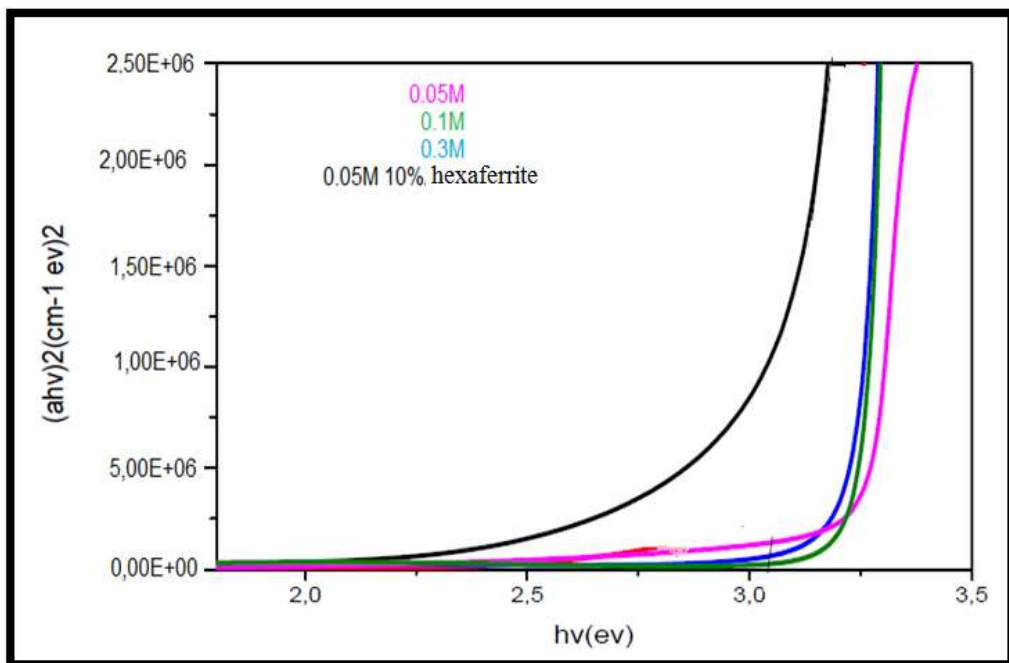


Figure (7).the absorbance spectral thin films of ligand(5-MeTAQ)



Figure(8).the energy band gap thin films of ligand(5-MeTAQ)

Table(3) energy band gap with different molar concentration and doping

Concentration (M)	Eg (eV)
0.05M	3.1 98
0.1M	3.142

0.3M	3.125
0.05M	3.056
10% hexaferrite doped	

3-7 Results of electrical measurements

The electrical measurements included measuring the conductivity of all the prepared pure and doped films with a ratio (and by measuring the resistance, the conductivity was calculated and the reciprocal of the resistance was equal as in the following law(18).

The electrodes for this measurement were first deposited, which are aluminum electrodes and the deposition was using a system of the type (Edward 306). This system contains a temperature heater (130 ° C) installed for the measurement process using a power supply connected to it in parallel, and after placing the membranes inside The system we notice the difference in the resistance value with the difference in concentration at the deformation and by means of a device containing copper wires tied to the poles of the membrane and the device used to measure the electrical conductivity of the type (Kiethly 619 electrometer) was found through the results of electrical conductivity measurements of the ligand membranes in the record that it has a high conductivity of Temperature (130 ° C) and this conductivity increases with the increase in concentration and when denatured with by 10% hexaferrite In the conduction beam, which leads to an increase in the conductivity and a decrease in the resistance of the prepared films. This can be explained by the approach of the Fermi plane more towards Delivery package. (19)

Table (4) the values of electrical conductivity and resistance of the pure and doped prepared films at different concentrations

Concentration (M)	Conductivity ($\Omega \cdot \text{cm}$) ⁻¹	Resistivity ($\Omega \cdot \text{cm}$)
0.05 M	2.24×10^{-4}	4.46×10^3
0.1 M	3.87×10^{-4}	2.56×10^3
0.3 M	4.48×10^{-4}	2.23×10^3
0.05 M 10% hexaferrite doped	6.53×10^{-4}	1.53×10^3

4- Conclusion

Based on the results of the spectroscopic and analytical diagnostic results of the heterogeneous ring dye of the ligand (5-MeTAQ) derived from thiazole ,The prepared complex was not affected by moisture, light and heat, in addition to its high melting temperatures, indicating its high stability, and the spectroscopy of ligand was diagnosed by analytical techniques Mass spectrum ¹H-NMR, FT-IR electronic spectra and EDX ,SEM, the ligand novel (5-MeTAQ) thin films were deposited by a simple and cheap method spray pyrolysis, The X-ray diffraction showed that films have a polycrystalline structure with an orientation the (101) The optical measurements have showed a decrease in the transmission T (%) with an increase in the molar concentration due to the surface roughness, SEM image shows the morphology thin films of ligand(5-MeTAQ) seems relatively homogeneous, The band gap values were decreased from (3.19 eV to 3.052) eV as molarity of increased for(0.05 to 0.3) M

and doping 10% hexaferrite. The reduced band gap is due to the impurity levels that are introduced into the band gap. Through the electrical measurements and the calculation of electrical conductivity, it was found that the conductivity increases with the increase in concentration and when the deformation is increased by (10% hexaferrite) and the electrical resistance decreases.

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