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Optical Characteristics of Nickel Oxide Thin Films Doped with Dye Extract from Three Natural Dyes

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ABSTRACT
Nickel Oxide (NiO) has displayed useful properties for application in solar cells, sensors, optoelectronics, and coatings. Modifying NiO with natural dyes has the potential to enhance its performance in these areas. This study deposited Nickel Oxide thin films with dye extract from Eupatorium Odorata, Ageratum conyzoides, and Pucro Pueraria using a chemical bath technique. The dye extraction was carried out at a constant temperature. The thin films are characterized using RBS, XRD, and SEM, and the optical properties of the films are investigated using various analytical methods. Transmittance and Reflectance data were used to calculate the film's optical constants such as refractive index, extinction coefficient and film thickness. Results showed that transmittance decreases with the doping of dyes in the UV region and is high in the VIS-NIR region, while reflectance is low in all spectral regions. The films show very high absorbance in the UV region, moderate in the VIS region and low in the NIR region. The band gap value was found to be 4.00eV, 3.83eV, 3.95eV, and 3.85eV for the as-grown NiO doped with Dye extract from leaves of Eupatorium Odorata, Ageratum conyzoides and Pucro Pueraria respectively. The extinction coefficient of the layer doped with dyes is higher compared to the as-deposited layer. The incorporation of dyes into the film matrix altered the real dielectric spectra of the as-deposited films and could be deployed in optoelectronics, coatings, or other fields.

Keywords:
Nickel Oxide,
Thin Film,
Eupatorium Odorata,
Ageratum conyzoides,
Pucro Pueraria.

INTRODUCTION

The exploration of non-toxic, earth-abundant inorganic materials that are easy to fabricate has become increasingly crucial in the advancement of solar energy technology, as highlighted by Ukpai et al. (2021). Addressing the soaring demand for energy, transitioning toward solar energy conversion into electricity can significantly help satisfy global power needs while lessening environmental harm. A promising way to lower the high costs of traditional electricity generation is by deploying materials like polycrystalline thin films or non-silicon-based compounds in solar cells. Such materials offer feasible alternatives because of several advantages: Non-toxicity and environmental safety, Abundance and commercial viability, Ease of fabrication, Non-silicon solar materials and Cost reduction.

Thin film is a layer of material that ranges from fraction of a nanometer to several micro-meters in thickness. Uhuegbu and Chidi (2007) defined thin films as

crystalline or non-crystalline materials developed two-dimensionally on a substrate's surface by physical or chemical methods. The methods of thin film deposition range from the very simple and cheap methods to the very expensive and complicated ones. In general, the classification of thin-film deposition techniques depend on the ways in which the atomic-molecular-ionic cluster of species are created, transported and condensed (deposited) on substrates from the solution or vapour phase of the coating materials. Nickel is a material that has wide application in thin films and that is essential for modern technologies based on the fabrication of nanostructured materials (Potocnik et al., 2013). Nickel oxide (NiO) is a transition metal oxide with excellent chemical and thermal stability (Chen et al., 2000; Osuwa and Onyejiuwa, 2013). It has potential applications in such areas as electro-chromic display devices, anti-ferromagnetic layers, and solar thermal absorber and as cathode material for alkaline batteries (Yadav et al., 2016; Gupta and Ahmad, 2018). Some other interesting electronic properties of NiO thin film include its wide band gap range of 3.6 - 4.0 eV and its p-type conductivity which make it a favorable material for electronic device applications (Krishnakumar et al., 2007).

In this study, the grown films were characterized using UV-VIS spectrophotometer to determine the optical and solid state properties. The surface and compositional characterization of the films were also carried out. The utilization of the dyes which are natural and locally available is a way towards producing green energy and reducing the climatic environmental problems associated with fossil fuel production.

MATERIALS AND METHODS

All measuring instruments, including beakers, syringes, and glass cylinders, were thoroughly cleaned with detergent, rinsed with distilled water, and dried before use. Glass slides were degreased by immersion in concentrated hydrochloric acid for 24 hours, then removed, washed with detergent, rinsed thoroughly with distilled water, and air-dried. Fresh leaves of Eupatorium odoratum, Ageratum conyzoides, and Pueraria species were harvested and thoroughly washed to remove dust and contaminants from which natural dyes were extracted. Molar solutions of the solid substances were prepared by dissolving the compound in distilled water to achieve the desired molarity. Specifically, 10 mL of 0.2 M nickel chloride ($NiCl_2$), 5 mL of distilled water, and 5 mL of ammonia solution were combined in a 1000 mL beaker and stirred continuously using a magnetic stirrer. For the deposition of nickel oxide thin films doped with natural dyes, three separate reaction baths were prepared, each containing 10 mL of 0.2 M $NiCl_2$, 5 mL of ammonia, 20 mL of distilled water, and 0.4 mL of dye extract from Eupatorium odoratum, Ageratum conyzoides, and Pueraria respectively. Five prepared glass slides were vertically immersed into the reaction bath, supported by synthetic foam to ensure stability. The bath was maintained at a temperature between 80°C and 85°C for 1 hour to facilitate the deposition of nickel oxide thin films. After the deposition period, the slides were removed, rinsed with distilled water, and hung to air dry on pegs allowed to dry at room temperature, thereafter annealed at 100 °C, 150 °C, and 200 °C, respectively. To evaluate the optical properties of the films, UV-Vis spectrophotometer was used to measure their absorbance, from which additional parameters were, derived (Nworie et al., 2024)

RESULTS AND DISCUSSION

Morphological and Optical Results for As-grown and Dye Doped NiO Thin Films is displayed in figures 1 to 16. The SEM image of the as-grown NiO thin film is presented in Figure 1, while Figures 2, 3, and 4 displays the SEM images of the films doped with dyes A, B, and C, respectively.

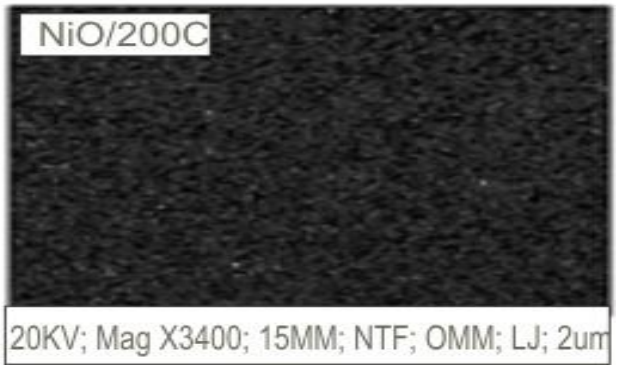

Figure 1: SEM Image of As-Grown NiO Thin Films

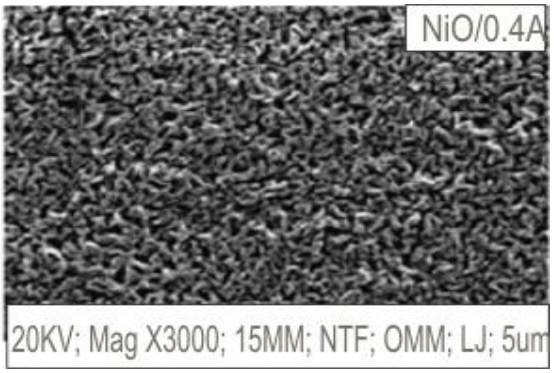

Figure 2: SEM Image of NiO Thin Films Doped with Dye A

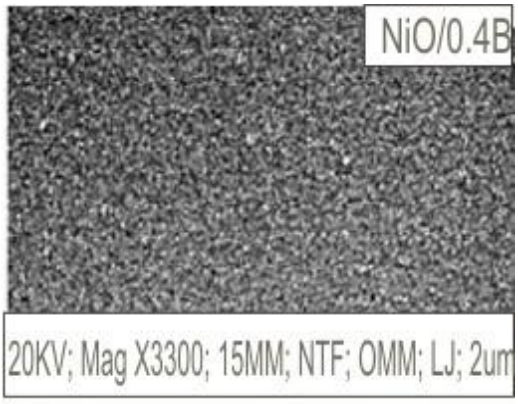

Figure 3: SEM Image of NiO Thin Films Doped with Dye B

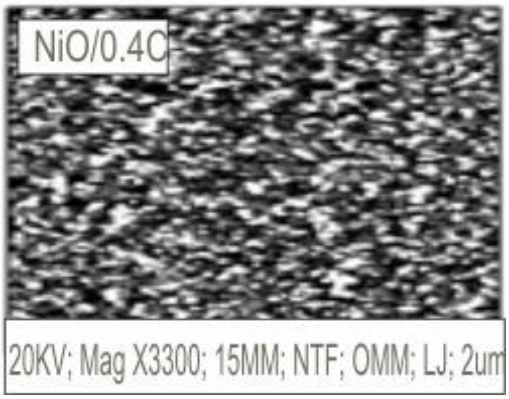

Figure 4: SEM Image of NiO Thin Films Doped with Dye C

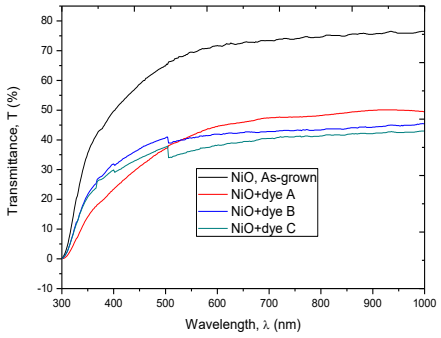

Figure 5: Trans. Vs λ for as-grown NiO + Dye

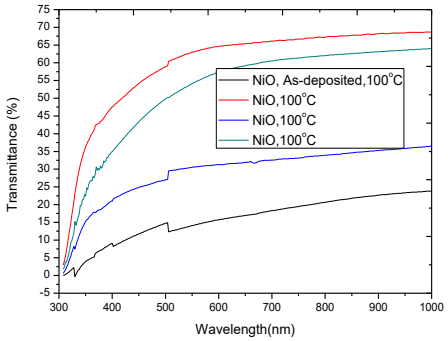

Figure 6: Trans. Vs λ for NiO + Dye, Annealed

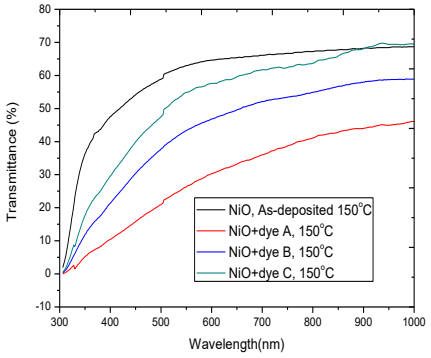

Figure 7: Trans. Vs $\lambda$for NiO + Dye, Annealed at 150°C

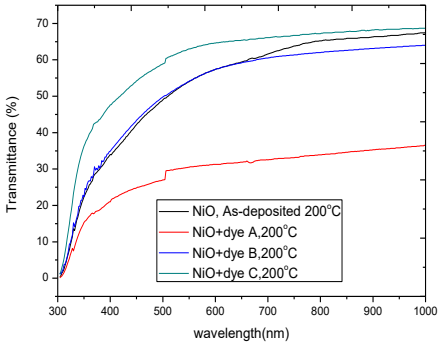

Figure 8: Trans. Vs $\lambda$for NiO + Dye, Annealed at 200°C

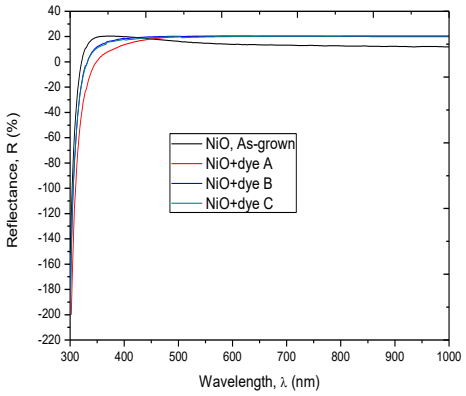

Figure 9: Reflect. Vs $\lambda$for as-grown NiO + Dye

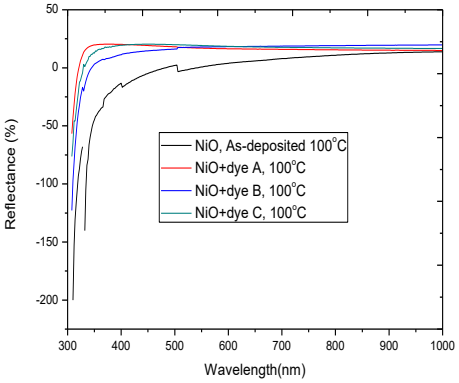

Figure 10: Reflect. Vs $\lambda$for NiO + Dye, Annealed at 100°C

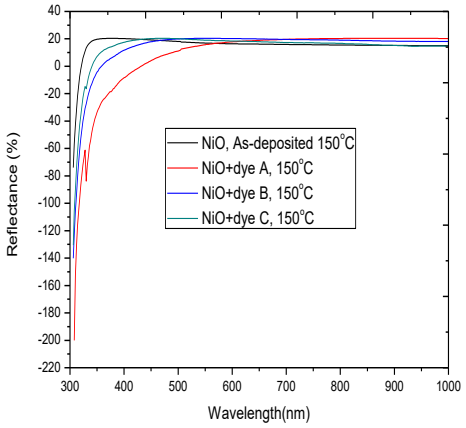

Figure 11: Reflect. Vs $\lambda$for NiO + Dye, Annealed at 150°C

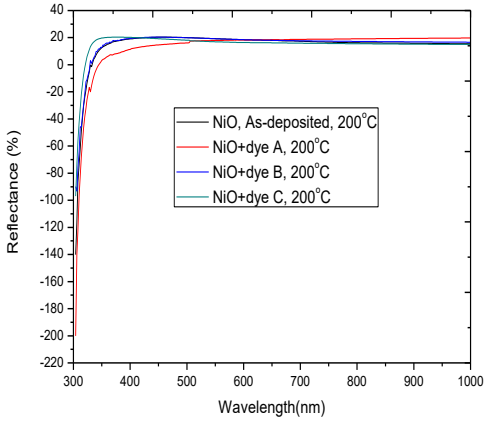

Figure 12: Reflect. Vs $\lambda$for NiO + Dye, Annealed at 200°C

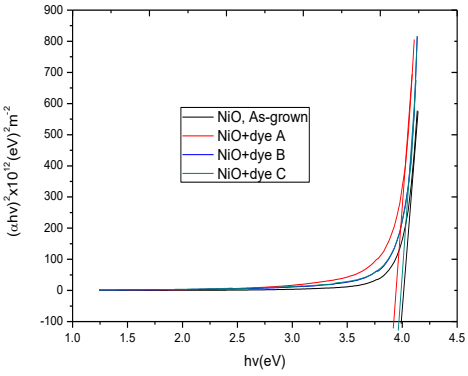

Figure 13: $(\alpha hv)^2$ Versus hv for as-grown NiO + Dye

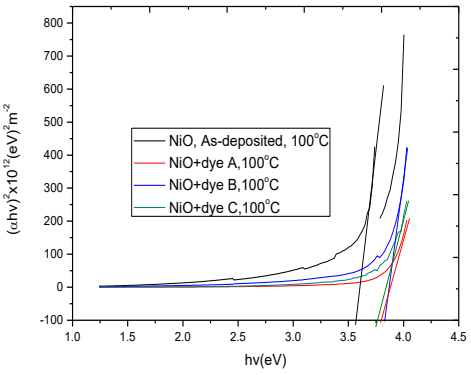

Figure 14: $(\alpha hv)^2$ Versus hv for as-grown NiO + Dye, Annealed at 100°C

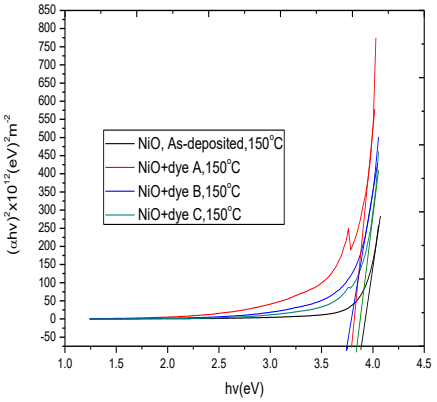

Figure 15: $(\alpha hv)^2$ Versus hv for as-grown NiO + Dye, Annealed at 150ºC

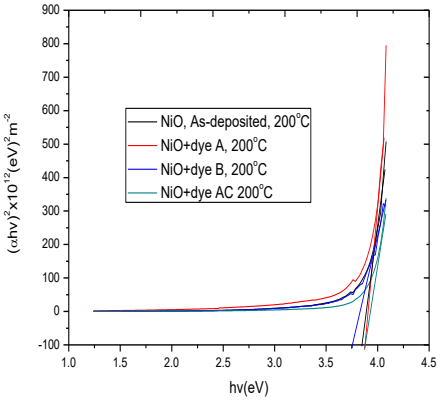

Figure 16: $(\alpha hv)^2$ Versus hv for as-grown NiO + Dye, Annealed at 200ºC

Figure 5 presents the transmittance spectra of the as-grown dye-doped NiO thin films as a function of wavelength. Figures 6, 7, and 8 shows the transmittance versus wavelength for samples doped with dyes and annealed at 100 °C, 150 °C, and 200 °C, respectively.

The reflectance spectra as a function of wavelength for the as-grown, dye-doped, and annealed samples at 100 °C, 150 °C, and 200 °C are illustrated in Figures 9, 10, 11, and 12, respectively. Figures 13, 14, 15, and 16 display the plots of $(\alpha hv)^2$ versus photon energy for the as-grown, dye-doped, and annealed samples at 100 °C, 150 °C, and 200 °C, respectively.

Discussion

The scanning electron microscopy (SEM) images of both as-grown and dye-doped NiO thin films demonstrate homogeneous, smooth, and well-adherent surfaces, free from pinholes and cracks (Figures 1–4). The micrographs reveal nanoparticles of varying sizes interconnected to form a continuous, flower-like network, indicative of nanostructured film morphology. This ordered network structure suggests favorable optical properties by enabling efficient light trapping, a desirable characteristic for applications in solar cell design (Soga, 2006). Similar nanoscale morphologies have been previously observed by others (Ukoba et al., 2018).

Figures 5 through 8 present the transmittance spectra of undoped NiO and NiO films doped with dye extracts derived from Eupatorium odoratum (dye A), Ageratum conyzoides (dye B), and Pueraria (dye C), annealed at 100 °C, 150 °C, and 200 °C, respectively. The transmittance values vary considerably: undoped NiO exhibits transmittance ranging from 0 to 75%, while films doped with dyes A, B, and C show reduced transmittance maxima of approximately 48%, 45%, and 42%, respectively. The decrease in transmittance upon dye doping, especially notable for dye C, indicates that the optical transparency of these films can be effectively modulated by the incorporation of natural dyes and by adjusting deposition parameters.

The transmittance values observed in this study are comparable to those reported for NiO thin films prepared via other techniques such as glancing angle deposition (GLAD) (Potocnik et al., 2013), spray pyrolysis (Vigneshkumar et al., 2016), and chemical bath deposition (Ezema et al., 2008) and Ishiwu et al., (2024). However, the doped films display transmittance below 50% in the visible region (400–700 nm), which is the spectral range relevant to human vision and peaks near 500 nm (Adaogbu et al., 2014). This relatively low visible transmittance disqualifies the dye-doped NiO films from applications requiring high optical transparency such as window coatings. Conversely, the as-grown undoped NiO films maintain sufficient transmittance suitable for such applications.

Additionally, these films exhibit low transmittance in the ultraviolet-visible (UV-VIS) region but moderate to high transmittance in the visible to near-infrared (VIS-NIR) region. This optical behavior suggests potential applications in UV protection, as the films can effectively filter harmful UV radiation while allowing visible and near-infrared light to pass through. This property is advantageous for protective coatings, such as UV-blocking eyeglasses to prevent sunburn or ocular damage, and for poultry house roofing and walls, where UV shielding is necessary to protect sensitive chicks while maintaining heat and visible light transmission. These results concur with previous findings reported by Ezema et al. (2008), Chikwenze and Nnabuchi (2010), and Augustine et al., (2018).

Reflectance spectra shown in Figures 9–12 indicate that reflectance increases within the wavelength range of 300 to 400 nm and remains relatively constant between 400 and 1000 nm. The maximum reflectance of about 20% is higher than values reported by Vigneshkumar et al. (2016) for spray-pyrolyzed NiO films, possibly due to

enhanced light scattering linked to the nanostructured morphology of the films in this study.

The optical band gap (Eg) of the films was estimated by extrapolating the linear portion of the Tauc plot, namely the $(\alpha hv)^2$ versus photon energy (hv) graphs (Figures 13–16). The as-grown undoped NiO film exhibited an Eg of approximately 4.00 eV, while doping with dyes A, B, and C produced band gaps of 3.83 eV, 3.95 eV, and 3.85 eV, respectively. Upon annealing at 100 °C, the band gaps shifted to 3.62 eV, 3.79 eV, 3.82 eV, and 3.75 eV for undoped and dye-doped films, respectively. Similar trends were observed at annealing temperatures of 150 °C and 200 °C, with band gaps maintaining a narrow range and showing slight variations corresponding to the specific dye used.

The observed narrowing of the band gap from 4.00 eV to around 3.85 eV upon dye doping and annealing correlates well with previous reports where energy band gap reductions were found in NiOx thin films deposited by combined electrochemical and sol-gel methods (Jiménez-González and Cambray, 2000) as well as films deposited by chemical bath and spray pyrolysis techniques (Boukhachem et al., 2014). Notably, the band gap values observed here are higher than those reported by Vigneshkumar et al. (2016) (3.25 eV) and Guziewicz et al. (2011) (3.56–3.71 eV), which could be attributed to differences in deposition methods, dopant effects, and film microstructure. The relatively constant energy band gap values within the doped films suggest that doping with natural dyes can be used as a viable strategy to tailor the optical band gap properties of NiO thin films through careful control of deposition and annealing conditions. This tunability could be leveraged in optimizing NiO films for specific applications such as electrochromic devices, solar cells, and UV protective coatings.

CONCLUSION

Nickel oxide (NiO) thin films doped with natural dyes from Eupatorium odoratum, Ageratum conyzoides, and Pueraria were successfully fabricated, exhibiting nanostructured morphologies and tunable optical properties. Dye doping reduced visible light transmittance and narrowed the band gap, making the films promising for UV-blocking and optoelectronic applications while maintaining environmental friendliness.

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