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A Green Pathway to Advanced Cobalt Oxide Materials: Dye Integration and Phytochemical Modification

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Abstract

This study investigated the impact of natural dyes derived from three plant sources (Eupatorium odoratum, Ageratum conyzoides, and Pueraria phaseoloides) on the bandgap energies of cobalt oxide (CoO) thin films thin films were synthesized using a chemical bath deposition technique at 80-85°C and subsequently annealed at 100, 150, and 200 °C. The incorporation of these natural dyes significantly influenced the bandgap energies of the CoO films, with values ranging from 3.80 eV to 4.12 eV depending on the dye and annealing temperature with the as-grown films exhibiting bandgap energies ranging from 3.80 eV to 4.00 eV. Moderate heat treatment typically increased the bandgap of the films, but at higher temperatures, the behavior became more complex, with some dyes demonstrating a substantial reduction in bandgap. These findings demonstrate the potential of utilizing natural dyes to effectively tailor the electronic structure of CoO, offering a sustainable and eco-friendly approach to materials modification.

Keywords: Bandgap, chemical bath deposition, Cobalt oxide, natural dyes, phytochemicals

1 Introduction

Cobalt oxide (CoO) is a significant p-type semiconductor with direct optical band gaps ranging from 1.48 eV to 2.19 eV, making it a material of interest for applications in gas sensing, solar energy absorption, and environmental purification [1]. CoO thin films have also found extensive use in lithium-ion battery electrodes, catalysts, ceramic pigments, field-emission materials, and magnetic applications [2]. Its unique electronic and optical properties, coupled with its abundance and low cost, make it an attractive candidate for technological advancements. However, the intrinsic limitations of CoO, such as its relatively low conductivity and limited light absorption range, hinder its full potential.

For optoelectronic applications, a wide band gap is essential to achieve high transmittance in the visible and solar spectral ranges [3, 4]. Researchers have explored various strategies, including doping, nanostructuring, and composite formation to improve its quality. Doping these materials enhances their electrical conductivity, metallic properties, and infrared reflection capabilities for specific plasma wavelengths. Various metal oxides, including zinc, cadmium, tin, lead, and their alloys, are often doped with foreign elements to achieve these properties. Such doping improves chemical and mechanical stability, which is a key advantage for these semiconductors [5, 6]. In recent years, there has been growing interest in utilizing natural dyes as a sustainable and eco-friendly approach to modify the properties of semiconductor materials. Natural dyes, derived from plants, offer a rich source of organic compounds with diverse functional groups, which can interact with the semiconductor surface, leading to significant changes in its electronic and optical properties.

Traditional methods of producing cobalt oxide coatings, such as chemical vapor deposition and sputtering, are costly and require complex setups [7]. Conversely, chemical bath deposition offers a cost-effective and scalable alternative for producing high-quality, large-area metal oxide films suitable for solar control, decorative coatings, and optoelectronic applications [8, 9, 10, and 11] demonstrated the surface reactivity and structural stability of cobalt oxides, highlighting facile interconversion between CoO and Co_3O_4 and their ability to undergo hydroxylation under ultra-high vacuum conditions, crucial for partial oxidation catalysis. [12] Studied CoO thin films deposited at varying pH levels, revealing that films with lower pH had high transmittance and potential for antireflection coatings, while those with higher pH exhibited properties suitable for solar cells and anti-dazzling applications.

This study investigated the impact of natural dye integration on the optical properties of CoO thin films, incorporating natural dyes extracted from *Eupatorium odoratum*, *Ageratum conyzoides*, and *Pueraria phaseoloides*, to enhance the light absorption properties of CoO and potentially tune its bandgap. This novel and green approach not only provides an environmentally friendly pathway but also offers a sustainable and cost-effective alternative to conventional methods of semiconductor modification.

2 Material and Methods

Fresh leaves of *Eupatorium Odorata*, *Ageratum Conyzoides*, and *Pueraria Tuberosa* were harvested and thoroughly washed. These were labeled as Dyes A, B, and C, respectively. Ten grams of each plant material were immersed in 100 mL of distilled water and extracted at 80-85°C for one hour. The resultant dye extracts were used for subsequent experiments. A reaction bath containing 10 mL of 0.2 M $CoCl_2$, 5 mL of thiourea, 20 mL of distilled water, and 0.4 mL of a specific dye extract was prepared for each film deposition. Thoroughly cleaned glass slides were vertically immersed in the respective bath and heated at 80-85°C for one hour. After deposition, the slides were rinsed with distilled water, air-dried, and annealed at 100°C, 150°C, and 200°C. The optical properties of the cobalt thin films were characterized using a spectrophotometer. The absorbance and transmittance data were used to calculate the optical absorption coefficient using Lambert's law [13]

3 Results and Discussions

Analysis of Phytochemical Content in Plant Leaf Extracts

The summary of the Phytochemical Contents of the dyes used is as presented in Fig. 1. The three plant species, *Eupatorium odoratum*, *Ageratum conyzoides*, and *Pueraria phaseoloides*, were found to contain a diverse range of phytochemicals. These compounds are known for their potential biological activities, including antioxidant, anti-inflammatory, and antimicrobial properties capable of fighting bacteria [14].

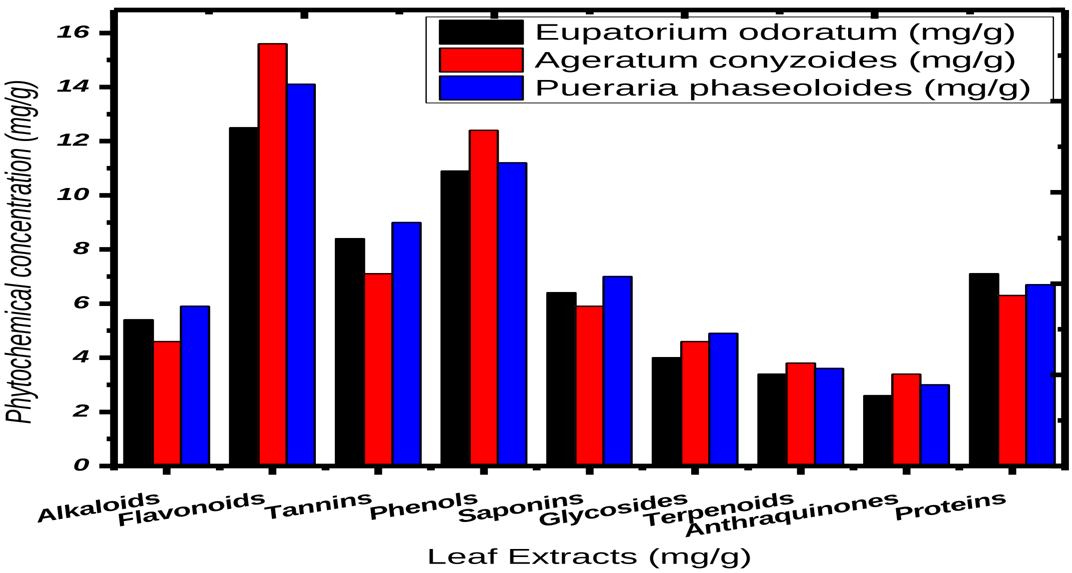

Figure 1. Comparative Chart of Phytochemical Composition in Plant Leaf Extracts

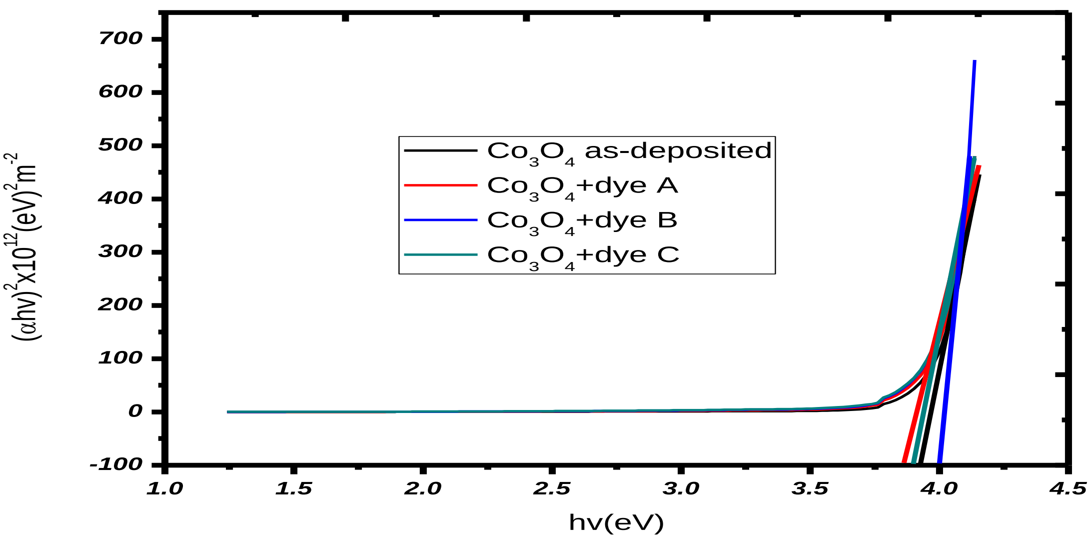

Figure 2. Plot of $(\alpha hv)^2$ Versus *hv* for As-grown and Dye Samples of CoO Thin Films

Fig. 1 reveals that all three plant species, Eupatorium odoratum, Ageratum conyzoides, and Pueraria phaseoloides, contain significant levels of flavonoids and phenols, both of which are known for their potent antioxidant properties [15]. Ageratum conyzoides exhibited the highest concentrations of both these compounds. Additionally, Pueraria phaseoloides was found to have the highest levels of tannins and alkaloids. Tannins are associated with astringent properties and antioxidant activity [16], while alkaloids are known for their diverse biological activities, including analgesic and anti-inflammatory effects [17]. The bandgap energies of the films with and without dye, annealed and unannealed are as displayed in Figs. 2 to 5. The band gap of the CoO thin films generally increased with increasing annealing temperature. This is in line with that obtained by [18, 19, and 20].

However, there are notable exceptions, particularly with CoO+dye B and CoO+dye C at higher temperatures (200°C). The presence of the dyes significantly impacts the band gap values compared to the as-deposited CoO film. Film CoO+dye A showed a decrease in band gap with increasing annealing temperature, suggesting a potential reduction in band gap due to the dye incorporation. This is trend to that obtained by [21] and indicates that annealing improves the films. Film CoO+dye B had an initial increase followed by a sharp decrease at 200°C.

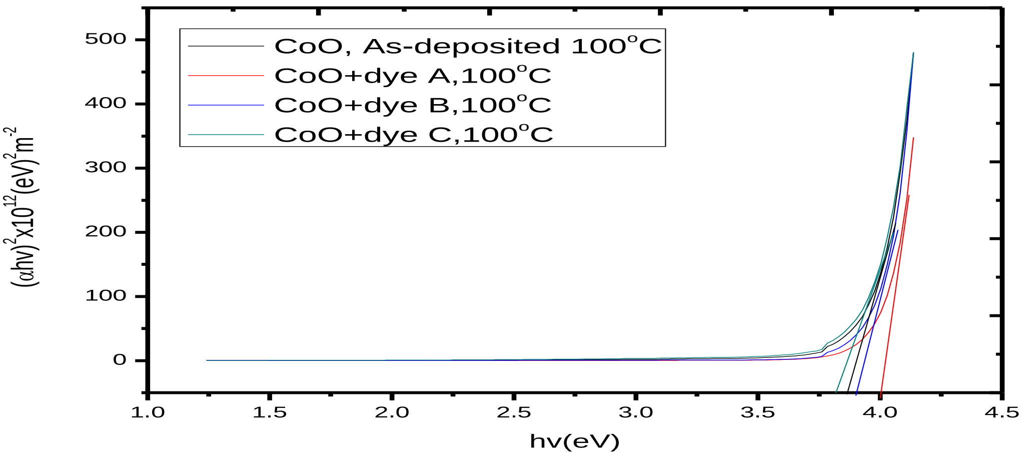

Figure 3. Plot of $(\alpha hv)^2$ versus hv for As-grown and Dye Samples of CoO Thin Films Annealed at 100°

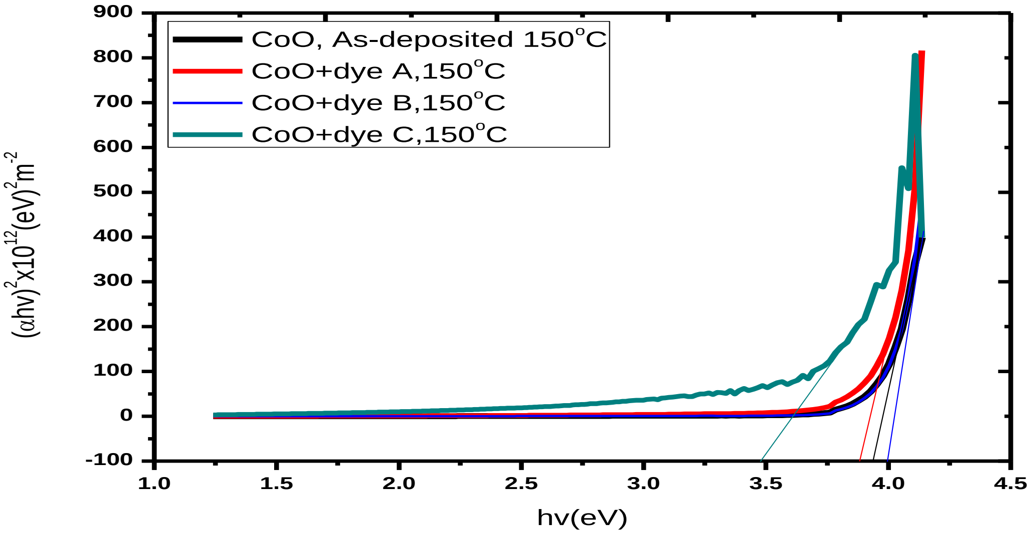

Figure 4. Plot of $(\alpha hv)^2$ versus hv for As-grown and Dye Samples of CoO Thin Films Annealed at 150°C

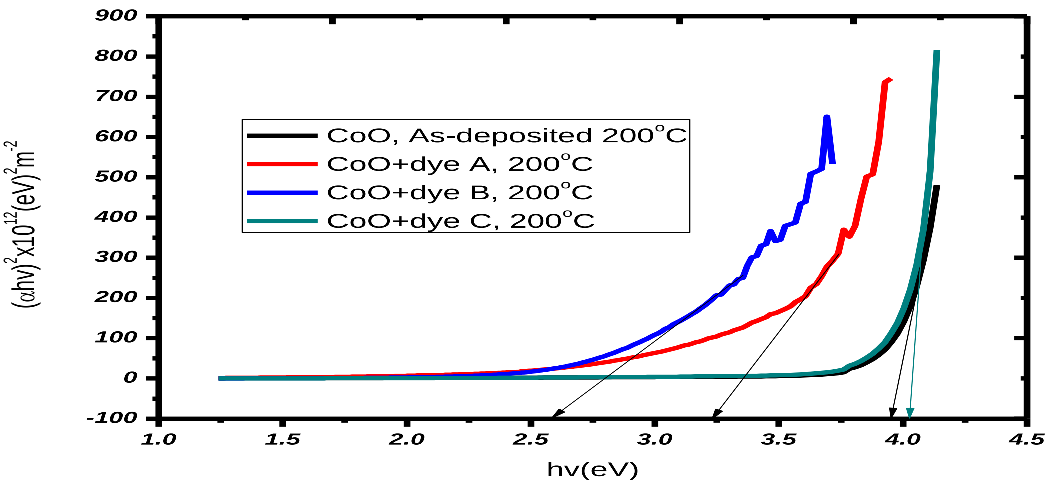

Figure 5. Plot of $(\alpha hv)^2$ versus hv for As-grown and Dye Samples of CoO Thin Films Annealed at 200°C

For Film CoO+dye C, it was an initial slight increase followed by a significant decrease at 200°C. At room temperature in Fig. 2, Dye A exhibited the lowest band gap (3.80 eV), suggesting strong light absorption, while Dye B showed the highest (4.0 eV), indicating weaker interaction with CoO. Dye C displayed an intermediate band gap (3.88 eV). Annealing at 100°C as displayed in figure 3 generally increased band gaps due to improved molecular order within the dye-CoO interface. However, higher temperatures (150–200°C) in Figs. 4 and 5 led to diverse responses, likely due to dye degradation or structural reorganization. Notably, Dye B exhibited a significant band gap narrowing to 2.60 eV at 200°C probably due to its high Phenols and tannins content functioning as primary photoreceptors [22], while Dye C's band gap increased to 4.12 eV, suggesting stabilization at elevated temperatures. The presence of tannins and saponins in the extracts, with their strong chelating and amphiphilic properties [23], facilitated strong dye-CoO interactions. Meanwhile, flavonoids and phenols contributed significantly to light absorption and charge transfer processes. These findings suggest that the developed dye-sensitized CoO composites hold promise for applications in solar cells, optoelectronics, and photocatalysis, where tunable optical properties are crucial.

4 Conclusions

This research explores a sustainable approach to enhancing cobalt oxide (CoO) materials by incorporating natural dyes extracted from three plant species. Using a solution growth technique, CoO thin films were successfully doped with these dyes. The study found that these plant-based dyes, rich in bioactive compounds, significantly influenced the optical properties of CoO, particularly the bandgap energy. While moderate heat treatment generally increased the bandgap, higher temperatures led to more complex behavior, with some dyes causing significant narrowing. These findings demonstrate the potential of utilizing natural dyes to tune the properties of CoO, opening promising avenues for the development of advanced CoO-based materials for applications such as solar cells and other optoelectronic devices.

For large-scale applications in mainstream optoelectronics and photovoltaics, further optimization and standardization of the materials and processes are essential. For niche applications where sustainability and cost-effectiveness are paramount, this method may be feasible on a smaller scale. Future research should focus on deeply understanding how dyes interact with CoO using techniques like FTIR, Raman spectroscopy, and XPS to identify chemical bonds and interactions between dyes and CoO, examining the CoO film's microstructure and morphology changes due to dye incorporation and annealing using TEM and AFM

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Disclosure of Conflict of Interest:

The authors declare that they have no conflicts of interest.

Data Availability Statement:

The data used in this study are available upon reasonable request from the corresponding author.

Statement of informed consent:

All participants gave their informed consent.

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