Influence of Graphene–Carbon Nanotube Loading on the Morphological and Electrical Transport Properties of NiO Hole Transport Layers

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ABSTRACT

Nickel oxide (NiO) has attracted considerable attention as an inorganic hole transport layer (HTL) because of its good chemical stability, wide optical bandgap, and compatibility with solution-based fabrication. However, the relatively low electrical conductivity of NiO can limit carrier transport through the layer. In this study, graphene carbon nanotubes (g-CNTs) were incorporated into NiO at different loadings to investigate their influence on the film morphology and electrical transport properties. NiO:g-CNT composites containing 3, 7, and 11 wt% g-CNT were prepared and compared with pristine NiO. The surface morphology of the films was examined using field-emission scanning electron microscopy, while Hall-effect measurements were used to evaluate carrier concentration, mobility, and resistivity. The addition of g-CNT clearly changed the electrical behaviour of the NiO films. The carrier concentration increased from 2.780×10^{14} cm^{-3} for pristine NiO to 7.481×10^{14} cm^{-3} at 3 wt% g-CNT, before showing some variation at higher g-CNT loadings. Meanwhile, carrier mobility increased steadily from 19.07 $cm^2 V^{-1} s^{-1}$ for pristine NiO to 170.50 $cm^2 V^{-1} s^{-1}$ at 11 wt% g-CNT. This increase was accompanied by a reduction in resistivity from 1177 to 60.95 Ω·cm and an increase in conductivity from 8.49×10^{-4} to 1.641×10^{-2} S cm^{-1}. The findings suggest that the improvement in conductivity at higher g-CNT loadings is mainly related to enhanced carrier mobility and the presence of additional conductive pathways within the NiO matrix. Overall, the results show that incorporating g-CNT can improve the electrical transport properties of NiO.

Keywords: Nickel oxide, graphene–carbon nanotube, hole transport layer, carrier mobility, electrical conductivity

INTRODUCTION

The growing demand for sustainable energy has driven the development of photovoltaic technologies that can convert sunlight directly into electricity. Among the emerging technologies, perovskite solar cells (PSCs) have gained considerable interest due to their promising optoelectronic properties, compatibility with solution-based processing, and potential for low-cost fabrication. Their performance, however, depends strongly on the properties of the individual functional layers, particularly the hole transport layer (HTL).

The HTL plays an important role in transporting holes from the perovskite absorber towards the corresponding electrode. Although organic materials are commonly used as HTLs, their sensitivity to environmental conditions, thermal instability, and concerns over long-term durability have encouraged the use of inorganic alternatives. Nickel oxide (NiO) is one of the promising inorganic HTL materials because of its p-type semiconducting behaviour, good chemical stability, wide optical bandgap, and suitability for inverted PSC structures. It can also be prepared using simple solution-based techniques such as the sol–gel method. Despite these advantages, pristine NiO generally has relatively low electrical conductivity, which may limit the movement of charge carriers through the HTL.

Several approaches have been explored to improve the electrical properties of NiO, including elemental doping, defect engineering, and the addition of conductive nanomaterials.[1][2] Among these approaches, carbon-based nanomaterials are particularly attractive because of their good electrical properties and their ability to provide additional pathways for carrier transport within a composite material. Carbon nanotubes (CNTs), for example, have a high aspect ratio that allows them to connect different regions of the host material and form conductive pathways. [3], [4], [5]. Graphenated-carbon nanotubes (g-CNTs) can provide additional conductive pathways, which may improve electrical transport in NiO.

Previous work has shown that incorporating a small amount of g-CNT into NiO can reduce electrical resistance and increase conductivity. [5], [6]. However, the effect of different g-CNT loadings on the electrical properties of NiO is not yet fully understood. Changes in g-CNT concentration may affect carrier concentration, mobility, resistivity, and conductivity, while also influencing the morphology and distribution of g-CNT within the NiO matrix.

For a p-type semiconductor, electrical conductivity can be expressed as a function of the hole carrier concentration and mobility:

$$\sigma = q\, p\, \mu \qquad (1)$$

where σ is electrical conductivity, q is the elementary charge, p is the hole carrier concentration and μ is the hole mobility. The present study investigates the influence of g-CNT loading on the morphological and electrical transport properties of NiO thin films. Pristine NiO is compared with NiO:g-CNT composites containing 3, 7 and 11 wt% g-CNT. Concentration-dependent changes in carrier concentration, mobility, resistivity and conductivity are evaluated using Hall-effect measurements and interpreted together with morphological observations. The conductivity measured here is similar to values reported for metal-doped NiO. The best layer (11 wt% g-CNT) reached 1.641×10^{-2} S cm^{-1}, close to the 0.024 S cm^{-1} reported for NiO doped with 8% Ag [7], and higher than that of Cu-doped NiO films made by the sol–gel method ($\approx 5.5 \times 10^{-3}$ S cm^{-1} for 6% Cu) [8] and ($\approx 4.3 \times 10^{-4}$ S cm^{-1} for 5% Cu) [9]. Still, fully optimized metal doping can do better sputtered NiO–Ag reached about 111 S cm^{-1} [10] because metal dopants raise the hole concentration directly, while g-CNT mainly improved carrier mobility.

METHODOLOGY

Glass substrates were cut into 1.5 cm × 2.0 cm pieces using a glass cutter. They were washed with tap water and detergent, rinsed in distilled water for 10 minutes, and cleaned in an ultrasonic bath. The substrates were then immersed in acetone for 10 minutes and ultrasonicated, followed by the same cleaning procedure using isopropyl alcohol.

Preparation of NiO

The NiO precursor was synthesised using nickel(II) acetate tetrahydrate ($Ni(CH_3COO)_2 \cdot 4H_2O$) via a sol–gel route. Initially, 0.625 g of nickel(II) acetate tetrahydrate was dissolved in a mixed solvent consisting of 15 mL of ethanol and 10 mL of isopropyl alcohol. Separately, 0.56 g of potassium hydroxide (KOH) was dissolved in 100 mL of deionised water at room temperature. The prepared KOH solution was then added dropwise to the nickel acetate solution until the pH reached 10. The solution was subsequently heated to 60 °C and continuously stirred for 2 h. After stirring, the mixture was allowed to stand undisturbed at room temperature until two distinct

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phases were observed, consisting of an upper supernatant and a lower precipitate. The supernatant was carefully removed, and the remaining mixture containing the precipitate was centrifuged using a Joanlab high-speed centrifuge at 1000 rpm for 10 min. Following centrifugation, the mixture separated again into a supernatant and precipitate, and the precipitate was retained for subsequent processing.

Preparation of NiO:g-CNT composites

For the preparation of the NiO:g-CNT composites, 0.625 g of nickel acetate tetrahydrate was used as the NiO precursor. g-CNT was incorporated at concentrations of 3, 7 and 11 wt%, calculated relative to the total mass of each composite. Accordingly, 0.0193, 0.0475 and 0.0773 g of g-CNT, respectively, were weighed using a Joanlab analytical balance. The resulting mixtures were synthesised using the same sol–gel procedure as that employed for pristine NiO.

Morphological characterisation

The surface morphology of pristine NiO and NiO:g-CNT composite films was examined using field-emission scanning electron microscopy (FESEM). The FESEM images were used to observe changes in the film surface as the g-CNT loading increased, particularly the distribution of g-CNT, surface uniformity, and the formation of agglomerates. These morphological changes were then compared with the electrical transport properties of the films.[11]

Hall-effect measurement

The electrical transport properties of the films were measured using the Hall-effect method with a Van der Pauw configuration.[12] The measurements provided the carrier concentration, carrier mobility, and electrical resistivity of each sample. The electrical conductivity was then calculated from the measured resistivity using the following equation:

$$\sigma = 1 / \rho \qquad (2)$$

where σ is electrical conductivity in S cm^{-1} and ρ is electrical resistivity in Ω·cm. The electrical parameters of pristine NiO were compared with those of the 3, 7 and 11 wt% g-CNT composites.

RESULTS

Morphological characteristics

FESEM was used to compare the surface morphology of pristine NiO with the NiO:g-CNT composite films. Pristine NiO showed a mainly granular surface with closely packed NiO grains. After the addition of g-CNT, elongated structures could be observed within the NiO matrix. As the g-CNT loading increased from 3 to 11 wt%, changes in the distribution and arrangement of these structures were observed across the film surface. The FESEM images were further compared to examine the dispersion of g-CNT and the presence of any agglomeration at higher loadings.

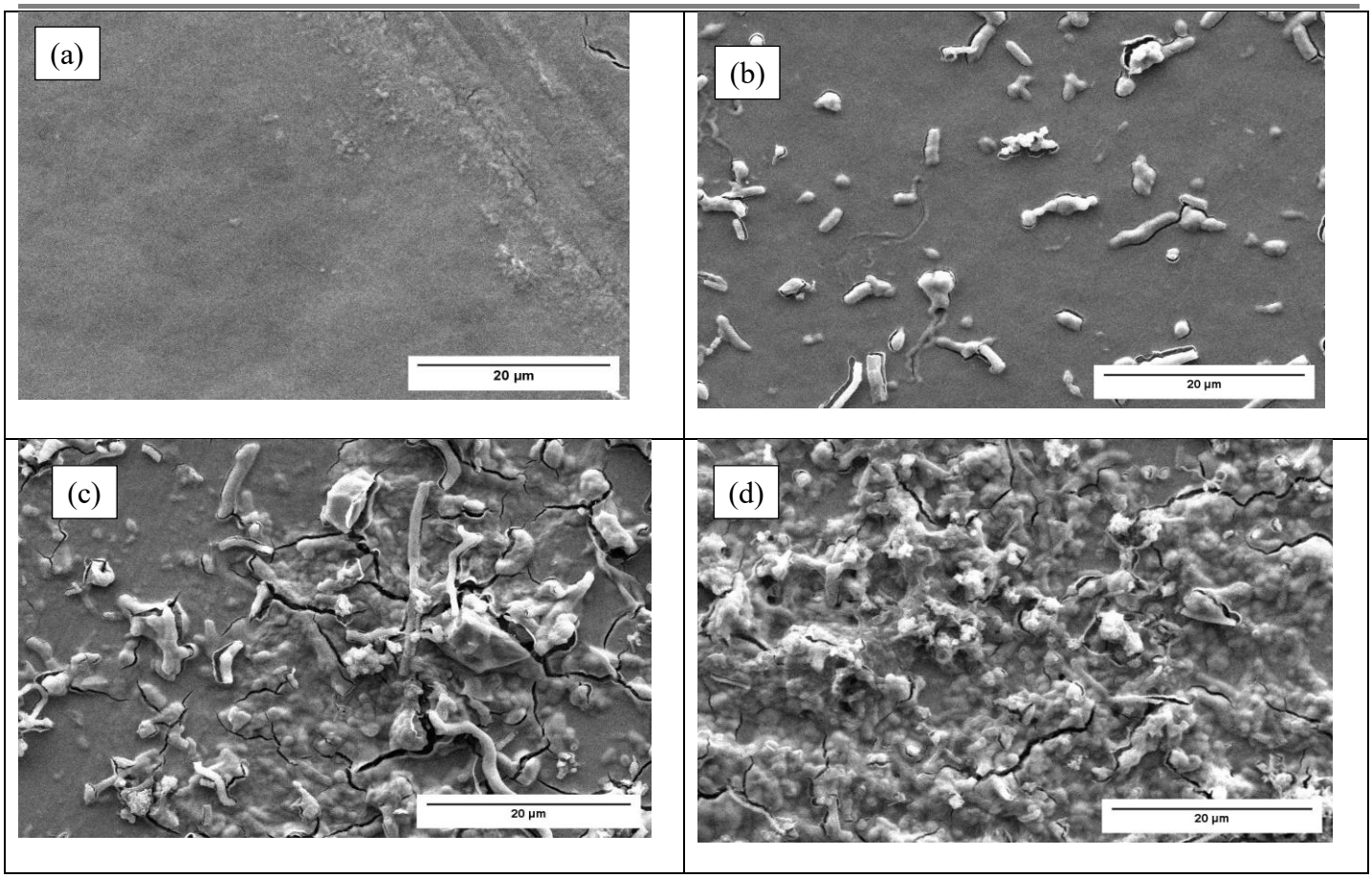

Figure 1: FESEM surface morphology of (a) pristine NiO and NiO:g-CNT composite thin films containing different g-CNT loadings (b) 3 wt% (c) 7 wt% and (d)11 wt%.

Elemental Composition

EDS analysis showed in Table 1 that the carbon content progressively increased from 24.0 at% in pristine NiO to 52.4 at% at 11 wt% g-CNT, confirming the increasing incorporation of g-CNT, while the Ni/O ratio remained relatively stable between 0.24 and 0.27. Enhanced optical and electrical properties of NiO-GO composite thin films on flexible PET substrates for optoelectronic applications [13].

Table 1: EDS elemental composition (atomic %) and Ni/O ratios of NiO and NiO:g-CNT at different g-CNT concentrations.

Sample	**Ni (at%)**	**O (at%)**	**C (at%)**	**Ni/O Ratio**
NiO (0 wt%)	15.1	55.7	24	0.27
NiO:g-CNT (3 wt%)	12.5	49.4	33.3	0.25
NiO:g-CNT (7 wt%)	9.6	39	48.1	0.25
NiO:g-CNT (11 wt%)	8.8	37	52.4	0.24

Electrical transport properties

Table 2 summarises the Hall-effect measurements for pristine NiO and the NiO:g-CNT composite films. Incorporation of g-CNT substantially changed all measured electrical transport parameters.

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Table 2: Electrical transport properties of pristine NiO and NiO:g-CNT thin films.

Sample	**Carrier concentration ($\times 10^{14}$ cm^{-3})**	**Mobility (cm^2 V^{-1} s^{-1})**	**Resistivity (Ω·cm)**	**Conductivity (S cm^{-1})**
NiO	2.780	19.07	1177.00	8.49×10^{-4}
NiO:g-CNT (3 wt%)	7.481	31.62	263.90	3.79×10^{-3}
NiO:g-CNT (7 wt%)	5.573	90.46	123.80	8.08×10^{-3}
NiO:g-CNT (11 wt%)	6.006	170.50	60.95	1.641×10^{-2}

Pristine NiO exhibited a carrier concentration of 2.780×10^{14} cm^{-3}, mobility of 19.07 cm^2 V^{-1} s^{-1} and resistivity of 1177 Ω·cm. After incorporation of 3 wt% g-CNT, carrier concentration increased to 7.481×10^{14} cm^{-3} and mobility increased to 31.62 cm^2 V^{-1} s^{-1}, while resistivity decreased markedly to 263.9 Ω·cm. Conductivity consequently increased from 8.49×10^{-4} to 3.79×10^{-3} S cm^{-1}.

At 7 wt% g-CNT, carrier concentration decreased to 5.573×10^{14} cm^{-3} relative to the 3 wt% sample, while mobility increased substantially to 90.46 cm^2 V^{-1} s^{-1}. Resistivity decreased further to 123.8 Ω·cm, corresponding to a conductivity of 8.08×10^{-3} S cm^{-1}. The 11 wt% g-CNT sample exhibited the highest mobility of 170.50 cm^2 V^{-1} s^{-1} and the lowest resistivity of 60.95 Ω·cm. Its conductivity reached 1.641×10^{-2} S cm^{-1}, despite its carrier concentration of 6.006×10^{14} cm^{-3} remaining below the maximum recorded at 3 wt%.

Conductivity increased continuously with g-CNT loading.[14] Relative to pristine NiO, conductivity was approximately 4.46 times higher at 3 wt%, 9.52 times higher at 7 wt% and 19.33 times higher at 11 wt%. The increase from 8.49×10^{-4} to 1.641×10^{-2} S cm^{-1} corresponds to an overall increase of approximately 1833%, while resistivity decreased by approximately 94.8%. The decrease in carrier concentration at 7 wt% can be understood from the EDS results, which show the Ni atomic percentage falling from 15.1 at% to 9.6 at% as the g-CNT loading rises, so that fewer nickel-vacancy acceptor sites are available to generate holes in the NiO host [15], while the carbon from g-CNT is not an ionized acceptor and thus mainly adds electrically inactive material [16], and agglomerated g-CNT bundles may further trap carriers at the interfaces [17].

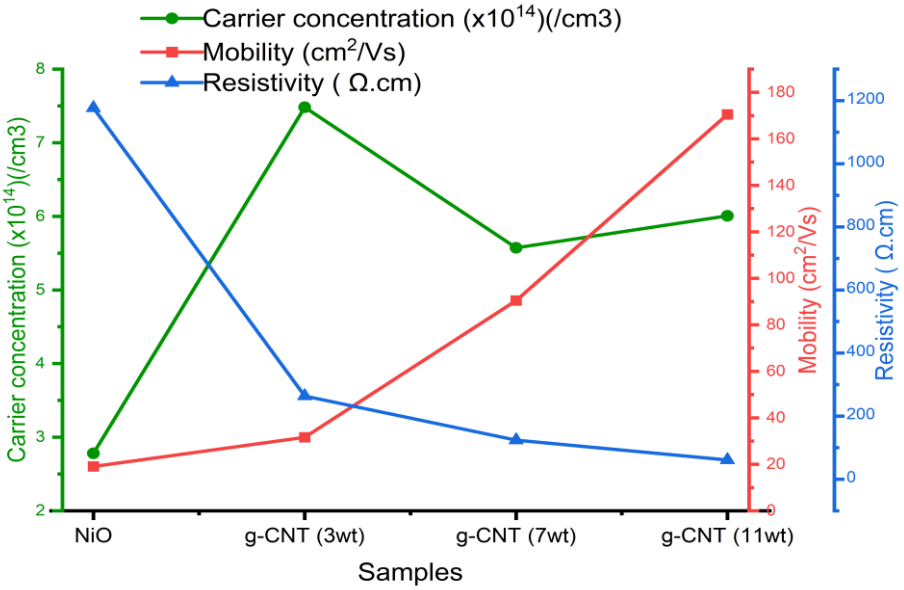

Figure 2: Variation in carrier concentration, mobility and resistivity of NiO thin films as a function of g-CNT loading.

DISCUSSION

The electrical properties of NiO improved as the g-CNT loading increased, although the carrier concentration did not follow the same trend. The carrier concentration increased from 2.780×10^{14} cm^{-3} for pristine NiO to its highest value of 7.481×10^{14} cm^{-3} at 3 wt%, before decreasing to 5.573×10^{14} cm^{-3} at 7 wt% and slightly increasing to 6.006×10^{14} cm^{-3} at 11 wt%. In contrast, carrier mobility increased steadily from 19.07 $cm^2 V^{-1} s^{-1}$ for pristine NiO to 31.62, 90.46, and 170.50 $cm^2 V^{-1} s^{-1}$ at 3, 7, and 11 wt%, respectively. At the same time, resistivity decreased from 1177 to 60.95 Ω·cm, while conductivity increased with g-CNT loading. These results show that the higher conductivity at increased g-CNT loading was mainly related to improved carrier mobility and additional conductive pathways rather than an increase in carrier concentration alone.

The electrical results can be linked to the changes observed in the FESEM images. Adding g-CNT to the NiO matrix provides additional pathways for carrier transport, which helps explain the increase in mobility and the decrease in resistivity as the g-CNT loading increases. However, adding more g-CNT also changes the surface morphology of the films. Although the 11 wt% sample showed the highest mobility and conductivity and the lowest resistivity, its surface became rougher and less uniform, with more visible agglomeration. This shows that the highest conductivity does not necessarily give the most suitable film for use as a hole transport layer. In comparison, the 7 wt% sample still showed a significant improvement in electrical transport while maintaining a better surface morphology than the 11 wt% sample. Therefore, 7 wt% g-CNT was considered a more suitable loading because it provided a better balance between electrical properties and layer quality.

CONCLUSION

The effect of different g-CNT loadings on the elemental composition, morphology, and electrical transport properties of NiO thin films was investigated using pristine NiO and samples containing 3, 7, and 11 wt% g-CNT. EDS analysis showed a progressive increase in carbon content from 24.0 at% in pristine NiO to 33.3, 48.1, and 52.4 at% for the 3, 7, and 11 wt% g-CNT samples, respectively, consistent with the increasing incorporation of g-CNT. Meanwhile, the Ni/O atomic ratio showed only a small variation from 0.27 to 0.24 across the investigated compositions. Hall-effect measurements revealed that carrier concentration increased from 2.780×10^{14} cm^{-3} for pristine NiO to its highest value of 7.481×10^{14} cm^{-3} at 3 wt%, but did not continue to increase at higher loadings. In contrast, carrier mobility increased steadily with g-CNT content, reaching 170.50 $cm^2 V^{-1} s^{-1}$ at 11 wt%. Correspondingly, resistivity decreased from 1177 to 60.95 Ω·cm, while conductivity increased from 8.49×10^{-4} to 1.641×10^{-2} S cm^{-1}. The continuous increase in carbon content and carrier mobility, despite the non-monotonic variation in carrier concentration, indicates that the conductivity enhancement at higher g-CNT loadings was predominantly associated with improved carrier mobility and the increasing contribution of conductive g-CNT pathways. Although the 11 wt% sample exhibited the highest conductivity and mobility, greater agglomeration and surface non-uniformity were observed. Therefore, among the investigated compositions, 7 wt% g-CNT provided a more favourable balance between enhanced electrical transport and film morphology for potential application as a NiO-based hole transport layer

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