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DFT Study on the Role of Oxygen Vacancies in the Structural and Electronic Properties of Spinel $NiCo_2O_4$

Nurzaihani Batrisyia Said [1], Nur Hamizah Mohd Zaki[2*], Nursyahirah Nordin[1], Fatin Nabilah Sazman[1], Nur Amirah Syafiqah Amirul[1], Nafisha Balqis Khairuddin[61], Ainnur-Sherene Kamisan[1], Oskar Hasdinor Hassan[2], Mohamad Fariz Mohamad Taib[1], Muhd Zu Azhan Yahya[3]

[1]*Faculty of Applied Sciences, Universiti Teknologi MARA, 40450 Shah Alam, Malaysia*
[2]*Ionic Material& Devices (iMADE) Research Laboratory, Institute of Science (IOS), Universiti Teknologi MARA, 40450 Shah Alam, Malaysia*
[3]*Faculty of Defence Science & Technology, Universiti Pertahanan Nasional Malaysia, 57100 Kuala Lumpur, Malaysia*

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ABSTRACT

Nickel cobaltite (NCO) has been widely studied as an electrode material for supercapacitors due to its good redox properties and conductivity. Oxygen vacancies are introduced to enhance the electrical conductivity of NCO by increasing the electron charge carrier and facilitating electron transport. In this study, the effects of oxygen vacancies on the structural and electronic properties of NCO were investigated using density functional theory (DFT) by using Cambridge Serial Total Energy Package (CASTEP). An oxygen atom was removed from the structure to introduce an oxygen vacancy, resulting in a slight lattice volume expansion and local bond rearrangement that induces lattice distortion. These changes were mainly due to the reduction of Co^{3+} to Co^{2+}. The formation energy was calculated as 4.43 eV, showing that the vacancy can form under suitable conditions. A decrease in band gap from 1.158 eV to 0.746 eV was observed, indicating improved conductivity. From the density of states, more Co 3*d* states and fewer O 2*p* states were found near the Fermi level. This suggests that electron transport can be enhanced by the presence of oxygen vacancies. Overall, the performance of NCO as a supercapacitor electrode can be improved by introducing oxygen vacancies.

INTRODUCTION

Supercapacitors (SCs) have emerged as promising energy storage devices for electric vehicles and portable electronic equipment due to their superior power density, rapid charge-discharge behavior, eco-friendly operation, safety, and long cycle lifespan (Khedulkar et al., 2023). The increasing demand for efficient and suitable energy storage technologies has driven significant advancements in supercapacitor development, particularly in the design of high-performance electrode materials that play a key role in determining energy

[1*] Corresponding author. *E-mail address*: *hamizahzaki@uitm.edu.my*

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storage efficiency. Metal oxides have been extensively studied as electrode materials owing to their high specific capacitance (100-2000 Fg^{-1}), which originates from fast and reversible surface redox reactions (S. George et al., 2023). Among them, transition metal oxides such as manganese dioxide (MnO_2), nickel oxide (NiO), cobalt oxide (Co_3O_4) vanadium oxide (V_2O_5), and ruthenium oxide (RuO_2) have shown promising results (Durukan, Yuksel, & Unalan, 2016; Dong et al., 2014; Hassan et al., 2013). Ruthenium dioxide RuO_2, in particular, exhibits the highest specific capacitance (1580 Fg^{-1}) and outstanding electrochemical performance. However, its toxicity, scarcity, and high cost limit its large-scale practical applications, thereby prompting the search for alternative materials that combine environmental friendliness with excellent electrochemical characteristics (Yakuphanoglu, 2009).

NCO has attracted considerable interest as an alternative electrode material due to its low cost, natural abundance, environmental compatibility, and superior electrical conductivity compared to Mn and V-based oxides (Zhou et al., 2022; Mir et al., 2024). Additionally, the presence of mixed-valence states in NCO enhances its redox activity, making it a suitable candidate for high-performance energy storage systems. Nevertheless, NCO is still constrained by its relatively low intrinsic conductivity 10^6S/cm and slow ion and electron transport, especially under high charge-discharge conditions (Zhang et al., 2023). These limitations reduce its energy storage efficiency and cycling stability. Recent studies have shown that the introduction of oxygen vacancies can improve the electrical conductivity and electrochemical performance of metal oxides by modifying their electronic structure (Hsu et al., 2006; Zulfiqar et al., 2021). Despite these promising findings, the detailed effects of oxygen vacancies on the structural and electronic properties of NCO remain insufficiently understood. More in-depth theoretical investigations are necessary to guide the design of defect-engineered NCO with improved electrochemical characteristics.

This study aims to investigate the effect of oxygen vacancies on the structural and electronic properties of NCO through first-principles calculations based on density functional theory (DFT). Oxygen atoms are systematically removed from the NCO lattice to identify the most energetically favorable vacancy configurations and to enhance electrical conductivity and charge storage capability. The outcomes are expected to provide theoretical insights into optimizing NCO as a high-performance electrode material for a supercapacitor.

METHODOLOGY

First-principles calculations were carried out using the Cambridge Serial Total Energy Package CASTEP), implemented in Material Studio, based on density functional theory (DFT) and pseudopotential methods. The crystal structure of NCO was modified by removing one oxygen atom to simulate an oxygen vacancy. Before further analysis, the new structure with the oxygen vacancy was optimized. Geometry optimization was performed using the Perdew-Burke-Ernzerhof (PBE-sol) exchange-correlation functional for solids. All calculations were conducted using a conventional cubic cell containing 56 atoms, consisting of 8 Ni atoms, 16 Co atoms, and 32 O atoms for the bulk, and 31 O atoms for the defective structure. A kinetic energy cutoff of 380 eV was applied, and the Brillouin zone was sampled using a 4×4×4 Monkhorst-Pack *k*-point grid. To improve the accuracy of the electronic structure, the Coulomb Hubbard U correction was applied, with values of of U_{Co}=4 eV, U_{Ni}= 5 eV (Wu et al., 2020). This correction was used to better represent the strong on-site interactions among the 3d electrons of Co and Ni. It also helped to reduce self-interaction errors and provided a more accurate prediction of the band gap and defect-related properties. Two antiferromagnetic configurations of the NCO spinel structure were investigated: a pristine lattice and a defective lattice containing a single oxygen vacancy, created by removing an oxygen atom coordinated to three Co atoms and one Ni atom. Both configurations were evaluated under identical computational conditions, as illustrated schematically in Fig 1. NCO is a cubic spinel crystalline structure that belongs to the F*d*3m space group (No.227) and is commonly referred to as a normal cubic spinel. This structure follows the general spinel formula AB_2O_4 and is characterized by a face-centred cubic (FCC) arrangement. The spinel structure exhibits two distinct oxidation states, where divalent Co^{2+} ions occupy the tetrahedral sites, while trivalent Co^{3+} ions are located at the octahedral sites, as noted in reference (Zaki et al., 2018).

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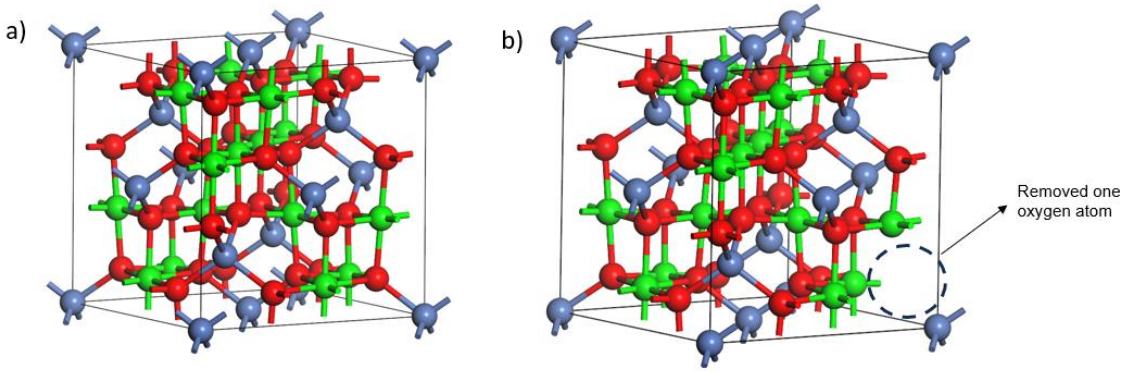

Fig.1. (a) shows the NCO spinel structure. (b) pristine (defect-free) lattice and lattice with an oxygen vacancy. Blue, red, and green spheres represent Ni, O, and Co atoms, respectively.

RESULTS

Structural properties

The oxygen vacancies are defects that play a vital role in influencing the properties of NCO on structural performance (Wei et al., 2022). Oxygen vacancies significantly influence the structural properties of NCO by introducing expansion in its crystal lattice volume with local distortion of the Co-O bond length. These vacancies disrupt the ideal arrangement of atoms, leading to local structural defects that alter the material's symmetry and stability. These results are consistent with the findings of Liu et al. (2016), which indicate that oxygen removal can lead to either lattice volume expansion or contraction. Table 1 shows the comparison of lattice parameters and volumes between pristine NCO and its oxygen vacancies. The lattice constant for pristine NCO is calculated as 8.084 Å, and increases to 8.104 Å after introducing a single oxygen vacancy. Similarly, the volume increases from 528.309 Å^3 to 532.229 Å^3. This observed expansion is attributed to the removal of an oxygen atom, which weakens the local Co-O bonds and reduces lattice cohesion, allowing neighboring atoms to relax outward (Zheng et al., 2022). Oxygen vacancies also alter the charge distribution, leading to the reduction of Co^{3+} to Co^{2+}. Because Co^{2+} has a larger ionic radius than Co^{3+}, this change further contributes to the expansion of the lattice (Sun et al., 2016). These structural relaxations help minimize the system's total energy, indicating that the defect structure becomes more energetically stable. In NCO, oxygen vacancies are expected to enhance supercapacitor performance by facilitating ion and electron transport, increasing lattice flexibility, and optimizing electrochemical processes. These vacancies can also generate additional pathways for ion migration, providing valuable insights into improving the material's energy storage capability and broadening its potential for energy-related applications (Wang et al., 2023). Moreover, the presence of oxygen vacancies has been reported to improve the electrical conductivity and electrochemical activity of transition metal oxides like Co_3O_4 (Hsu et al., 2006; Zulfiqar et al., 2021). In the case of NCO, such vacancies are expected to enhance its performance as a supercapacitor electrode by promoting better ion and electron transport. Therefore, analyzing the lattice change due to oxygen vacancy formation offers valuable insight into improving the material's energy storage properties.

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Table 1. The details on lattice parameter and volume of NCO

An example of a column heading	Calculated (NCO)	Calculated (O vacancy)	Experimental (Sun et al., 2016)	Theoretical (Shi et al., 2016)
Lattice constant (Å)	8.084	8.114	8.114	8.152
Volume (Å^3)	528.309	532.229	534.201	541.742

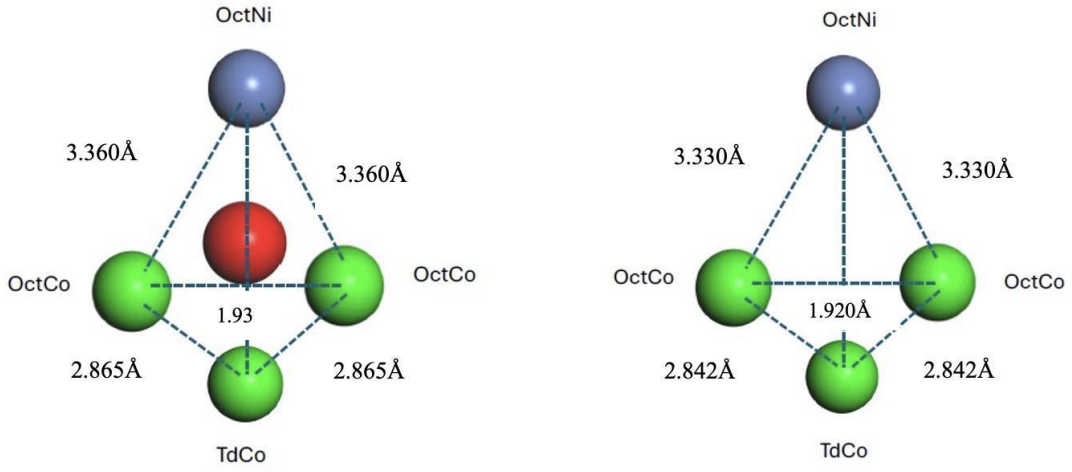

Fig. 2 (left) shows interatomic distances between metal atoms of pristine NCO; (right) oxygen vacancies of NCO

Formation and Electronic properties of NCO with one O vacancy

Oxygen vacancies have been shown to improve the electrical conductivity and electrochemical activity of various metal oxides, such as Co_3O_4, and are also believed to enhance the electrical performance of NCO (Sun et al., 2016). To evaluate the stability of an oxygen vacancy in NCO, the formation energy was calculated using the following equation;

$$E_{form} = E_o - E_i + 1/2\mu_{O2} \quad (1)$$

where is the E_{form}, formation energy, E_o is the total energy of the structure with one oxygen atom removed, E_i is the total energy of the pristine NCO structure, and μ_{O2} is the chemical potential of an oxygen molecule (Shi et al., 2016). The calculated formation energy of the oxygen-deficient NCO structure is 4.43 eV, indicating that the generation of oxygen vacancies is energetically feasible under appropriate synthesis or operating conditions. This value is in close agreement with the formation energy of 4.23 eV reported by Wu et al. (2021), suggesting good consistency between the present calculations and previous theoretical studies. Compared with the pristine structure, the introduction of an oxygen vacancy provides valuable insight into the role of oxygen defects in modifying the electronic structure and associated properties of NCO.

The electronic properties of NCO are significantly influenced by the presence of oxygen vacancies, which act as critical defects in altering its conductivity characteristics. Oxygen vacancies have been widely reported to enhance room-temperature antiferromagnetism (AFM) in materials like Co-doped ZnO and increase electrical conductivity and electrocatalytic activity in Co_3O_4 (Guruvammal et al., 2016). Similarly, in NCO, oxygen vacancies are known to improve its electronic properties, making it a more effective electrode material.

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Fig. 3 shows the band diagram for NCO, calculated along specific symmetry directions within the Brillouin zone at the points G(0.000, 0.000, 0.500), F(0.000, 0.500, 0.000), Q(0.000, 0.500, 0.500), Z(0.000, 0.000, 0.500) for (left) band structure of pristine NCO, (right) band structure of oxygen vacancies of NCO. Compared to the pristine structure, the oxygen-vacancy system exhibits noticeably narrower bands near the Fermi level, indicating increased carrier localization and modified electronic dispersion. This band flattening suggests enhanced electronic conductivity due to defect-induced states, which can facilitate charge transport across the lattice. The calculated band gap of pristine NCO is found to be in good agreement with the experimentally reported value of 1.490 eV (Saafi et al., 2021), validating the reliability of the present DFT+U approach. Upon the introduction of oxygen vacancies, the band gap is significantly reduced due to the emergence of defect states near the Fermi level, resulting in a value of 0.746 eV, which further enhances electrical conductivity. This band-gap value has been reported in other studies, such as 0.911 eV obtained by (Chang et al.,2021). Such electronic structure modulation is advantageous for electrochemical energy storage, as it promotes faster electron transfer and improves charge storage kinetics. In addition to electronic enhancement, oxygen vacancies are expected to increase the electrochemically active surface area and create additional ionic diffusion pathways, collectively contributing to improved supercapacitor performance (Abdelrahim et al., 2024). These findings highlight the critical role of oxygen defects in tailoring both the electronic and electrochemical properties of NCO, thereby reinforcing its suitability as a high-performance supercapacitor electrode material (Adel et al., 2024).

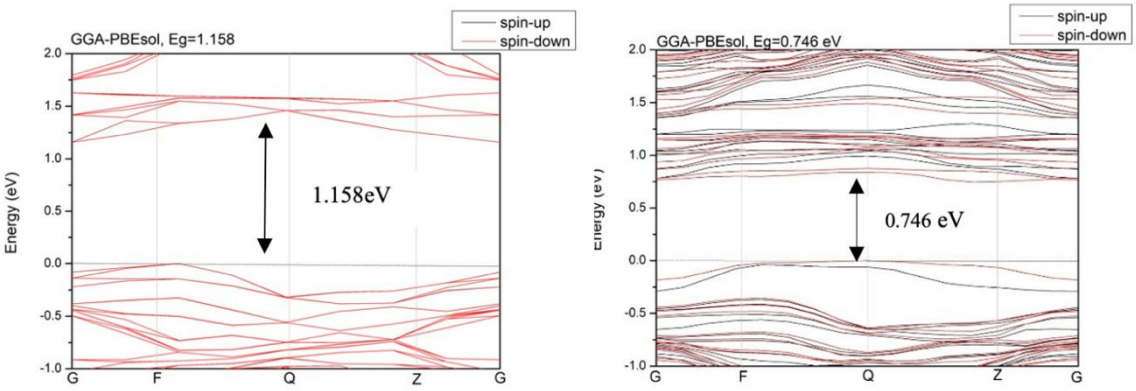

Fig. 3 left panel corresponds to the band structure of pristine NCO, while the right panel represents the band structure oxygen-deficient NCO.

Fig. 4 a) illustrates the partial and total density of states (DOS) of pristine NCO and NCO OV. In pristine NCO, the O 2*p* states exhibit strong hybridization with the Co 3*d* orbitals, and a narrow band gap is observed near the Fermi level, which reflects its semiconducting nature. Upon the introduction of oxygen vacancies, a significant increase in the Co 3*d* states near the Fermi level is observed, accompanied by a narrowing or near closure of the band gap. This indicates that oxygen vacancies generate defect states, thereby enhancing the conductivity of the material (Yuan et al., 2018). Furthermore, the spin asymmetry shows a magnetic configuration in NCO and NCO OV. These findings demonstrate that oxygen vacancies play a crucial role in tailoring the electronic structure of NCO, leading to improved conductivity and making it a more promising candidate for high-performance supercapacitor applications. Fig 4b) shows the charge density in the NCO pristine structure, while Fig 4c) shows the charge density after introducing an oxygen vacancy, the interaction along the Ni–(vacancy) direction weakens, whereas the remaining Ni–O bonds show relatively stronger interactions. The electron localization function (ELF) map of NCO further shows increased electron localization around the neighboring oxygen atoms, suggesting enhanced local bonding

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character around the remaining O sites. Oxygen vacancy has important effects on the magnetic and electrical properties of NCO. Oxygen vacancy has important effects on the magnetic and electrical properties of NCO (Wu et al., 2020).

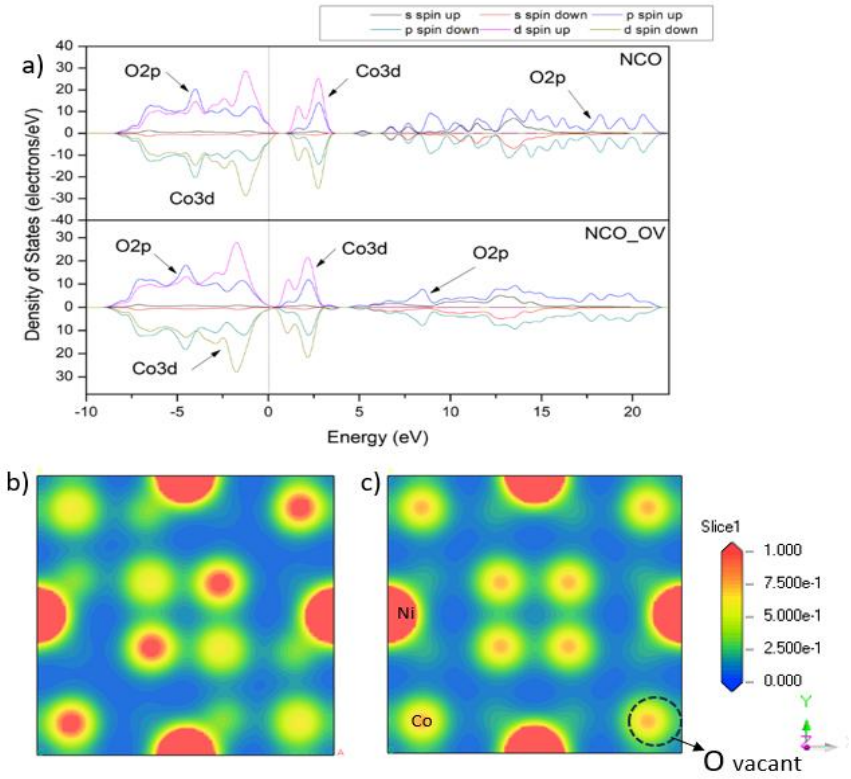

Fig. 4. a) Partial and total density of states (DOS) of NCO and NCO OV. b) charge density of NCO. c) charge density with O vacant.

CONCLUSIONS

In this study, the effect of oxygen vacancies on the structural and electronic properties of NCO was studied using DFT. When oxygen atoms were removed from the crystal, the structure become slightly expanded and distorted. This change happened because some of the strong metal-oxygen bonds were broken, and some Co^{3+} ions were converted into larger Co^{2+} ions. These changes helped make the structure more stable and allowed easier movement of ions. The formation energy of the oxygen vacancy was calculated to be 4.43 eV, showing that this defect can form under suitable conditions. The electronic properties were also affected, where the band gap became narrower (from 1.158 eV to 0.746 eV), meaning that material became more conductive. The partial density of states showed that more Co 3*d* electrons were present near the Fermi level, while the O 2*p* contribution decreased, which also pointed to better conductivity. In summary, the introduction of oxygen vacancies was found to modify both the structural and electronic properties of NCO. These defects enhance its conductivity, thereby improving its potential for supercapacitor applications.

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Conflict of Interest Statement

The authors agree that this research was conducted in the absence of any self-benefits, commercial or financial conflicts and declare the absence of conflicting interests with the funders.

Authors' Contributions

Nurzaihani Batrisyia conducted the research and prepared the initial draft of the manuscript. Nur Hamizah Zaki revised the manuscript and conceptualized the central research idea. Norsyahirah Nordin, Fatin Nabilah Sazman, Nur Amirah Syafiqah, and Nafisha Balqis contributed to writing the theoretical framework. Sherene and Oskar Hassdinor supervised the research progress. Fariz Taib designed the research methodology and anchored the review. Zu Azhan provided revisions and approved the final version of the manuscript for submission.

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