





Green, cost-effective synthesis of NiMnO₃ nanoparticles and their use in supercapacitor and photodegradation applications

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Abstract

Nickel manganese oxide nanoparticles (NiMnO₃ NPs) have been synthesized by a solution combustion method, varying the metal-to-fuel ratios. The obtained compounds were characterized by several techniques, including X-ray diffraction, FT-IR, FESEM and HR-TEM. The optical band gap decreased with increasing crystallite size. The BET analysis revealed a decrease in surface area with increasing crystallite size. Crystallite size dependent electrochemical investigations of the NiMnO₃ NPs were carried out using a three-electrode system. The NiMnO₃ NPs were observed to be a better supercapacitor material with a specific capacitance of 65.45 F/g at 1 A/g. The specific capacitance decreased linearly with the cumulative current densities, which is the characteristic behavior of electrochemical supercapacitors. The degradation of RB increased with the enhancement in the crystallinity of the sample. The proportion of RB dye degradation was found to be 99.89% in two hours. The enhancement of photodegradation and the supercapacitor property of the NiMnO₃ NPs are attributed to the reduction in the band gap due to increase in crystallite size.

Introduction

Cu, Co and Ni mixed metal oxides, with a spinel-type structure, are more stable than their single oxides. Among them, Ni-Mn-based materials have sufficient active sites and facilitate the transfer of charges [1]. Mixed nickel manganese oxide matches two forms of oxide structure in the crystal: spinel (NiMn₂O₄) and ilmenite (NiMn₂O₃); within the spinel structure, Ni²⁺, Mn²⁺, Mn³⁺ and Mn⁴⁺ ions can co-occur and be located between the tetrahedral and octahedral spinel sites. At the same time, Ni²⁺ and Mn⁴⁺ are octahedrally located in the ilmenite structure [2]. Stoichiometric nickel manganese oxides have been synthesized by different approaches, such as the hydrothermal [3], thermal decomposition [4], combustion [5], electro co-precipitation [6], solvothermal and annealing methods [7], [8].

Green nanotechnology is a new topic of study that studies the synthesis of nanoparticles by using natural fuels [9]. This issue has critical implications in a variety of industries, including medicines, nuclear energy, fuel energy, electronics and biotechnology. Metal and metal oxide nanoparticles synthesized by green technology are finding new uses in the biomedical field, including diagnostics, wound healing, tissue treatment, immunotherapy, regenerative medicine, dentistry and biosensing platforms [10]. Green synthesized NPs have also found application in the field of energy storage devices, like supercapacitors and lithium-ion batteries (LIBs) [11].

Fuel has an important role in the quality of the final product, acting not only as a reducer but also as a complexing agent and a microstructural template. Chelating compounds in fuels prevent metal ions from precipitating and maintain compositional homogeneity among all the ingredients, which aids in the formation of strong coordinating bonds [12]. As a result, fuels are commonly utilized in the synthesis of metal oxides, as they promote a gel network following solvent evaporation, which keeps the metal cations uniformly fixed in their positions throughout burning [13].

Due to their high theoretical power, low toxicity, low price, thermal stability and increased natural abundance, Mn-based oxides are attractive as anode materials for LIB applications [14]. Ni-Mn-based oxide materials are of great interest because of their greater pseudo capacitive reaction, higher specific capability and conductivity when used as an electrode material for supercapacitors [15]. Ni- and Mn-based materials have abundant catalytic sites and facilitate charge transfer reactions and adsorption; hence, they have been used as catalysts in the oxygen evolution reaction [16]. Mixed 3D transition metal oxides (NiMnO₃) have more oxidation states and could, therefore, be used as a catalyst for the decomposition of ozone [17]. The potential of nickel and manganese to form oxides with varying oxidation states and their high oxygen storage capacity makes it possible to use nickel manganese oxides in the combustion of volatile organic compounds [18]. An effective photocatalyst was therefore found in Mn-based oxides through optimal electronic properties and electrochemical stability against electrochemical corrosion and photo corrosion [19]. The redox reaction occurs at the outermost layer of the nickel-based materials. From this perspective, the material should be as thin as feasible. However, if the material is too thin and aggregation is avoided at the same time, the loading mass decreases. As a result, the emission of Ni-Mn into the environment is prevented [20].

Semi-conductive nanomaterials exhibit UV-vis activity, are non-toxic, physiologically inactive, chemically stable, water-insoluble and cost-effective. Metal oxide photocatalysts absorb photons equal to or more than their band gap energy, resulting in photo-generated charge carriers. These charge carriers are very reactive, producing damaging species like OH⁻ ions and superoxide radicals that may oxidize and decolorize hazardous organic pollutants [21], [22]. Semiconducting transition metal oxides nanoparticles and their composites play an important role in the removal of organic pollutants from waste water [23], [24]. In the present research, NiMnO₃ NPs were synthesized via a solution combustion method and their supercapacitor and photocatalytic properties were examined. We have studied crystallite size varied photodegradation and supercapacitor behaviour. In this study, we observed that the photodegradation of RB dye and capacitance property increases with the increase in the crystallite size of the NiMnO₃ NPs. The reason behind the enhancement of the photodegradation and supercapacitor properties is the reduction in the band gap due to the increase in the crystallite size.

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Materials

The seeds of *Tamarindus indica* were obtained from a local market in Bengaluru, Karnataka, India. Nickel nitrate hexahydrate and manganese nitrate hexahydrate with analytical reagents grade were procured from SD Fine-Chem Limited and were used without additional purification....

Structural analysis

The crystal structure of the NiMnO₃ NPs was studied using PXRD measurements, as shown in Fig 1. The XRD pattern reveals a highly crystalline structure with nine prominent peaks, a key feature of NiMnO₃ NPs. All these peaks indicate the rhombohedral structure of the NiMnO₃ NPs [27], with the R3 space group, and lattice constants are $a = c = 4.93 \text{ \AA}$, $b = 5.34 \text{ \AA}$. The peaks obtained correspond to the planes (101), (112), (220), (020), (220), (231), (130) and (211), and the lattice constants measured ...

Conclusion

NiMnO₃ NPs were prepared by a solution combustion technique using tamarind seed powder as a fuel. The obtained samples were synthesized at different metal to fuel ratios. The band gap of the synthesized nanomaterials decreased with an increasing metal to fuel ratio due to an increase in the crystallite size. The surface area analysis confirmed the porous nature of the materials, which could be used in energy conversion and energy storage applications. The electronic absorption spectra and...

CRedit authorship contribution statement

H.S. Sumantha: Writing – original draft, Methodology, Conceptualization. **B.L. Suresha:** Writing – review & editing, Validation, Supervision, Conceptualization....

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper...

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