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# Characterization of hydrogen storage behavior of the as-synthesized p-type NiO/n-type CeO<sub>2</sub> nanocomposites by carbohydrates as a capping agent: The influence of morphology

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## ABSTRACT

This study proposes a p-type/n-type heterojunction system for electrochemically hydrogen storage. The electrochemical investigation was done as a simple method to evaluate storage capacity. The p-type NiO/n-type CeO<sub>2</sub> mesoporous nanocomposites were prepared via a facile thermal decomposition way that is better than the carbohydrates as a green capping agent. For the first time, the electrochemical hydrogen storage behavior of this nanocomposite was evaluated by chronopotentiometry technique in a potassium hydroxide aqueous solution (6 M KOH) under 1 mA current. The electrochemical measurements display that the hydrogen storage capacity is largely dependent on the design of the nanostructures. Sample No. 2 with the plate-like architecture has higher hydrogen storage capacity than sample No. 1 having particle architecture. The plate-like architecture increases the storage capacity by reducing the diffusion pathway, increasing the surface area, and buffering the volume change during cycling. The hydrogen storage capacity for sample No. 1 and 2 was obtained  $\approx$ 5500 and 6850 mAh/g, respectively.

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## Introduction

Global demand for energy is increasing rapidly as the population and economy is growing. In the past, humans used to apply wood as an energy source. After the Industrial Revolution, fossil fuels were used as the main source of energy. The number of fossil fuel reserves are not known. Hence, this source of energy has confronted with limitations [1]. Today, hydrogen is known as an energy carrier and can be employed to produce electricity. Hydrogen can be used for vehicular

applications because of its potential as a source to resolving the global energy problems [2,3]. Due to the gravimetric energy density of hydrogen, it is a promising candidate for applications in future [4,5]. The typical routes of hydrogen storage are consist of compressed gas, chemisorption in the form of metal hydrides under high pressure, and liquefied hydrogen [6–9]. These traditional routes have major problems such as (1) having a safe and effective storage problem, and (2) not being economical due to unfavorable temperature conditions and high pressure [10–13]. Hence, development of a safe, simple, and cheap storage route is essential for its widespread usage

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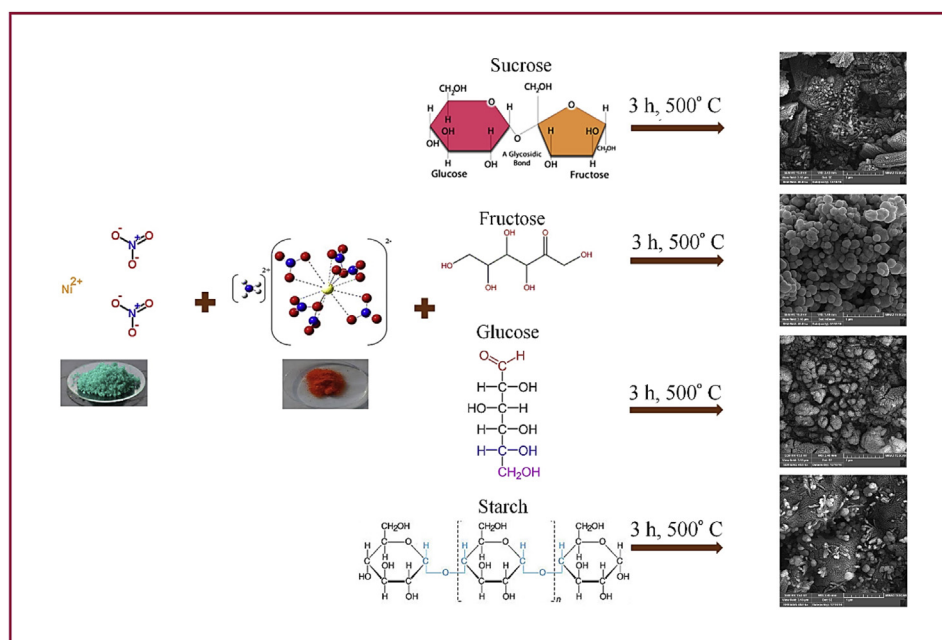
[14]. The electrochemically hydrogen storage route has been known as an effective and stable storage method at ambient temperature and pressure [15–18]. In hydrogen storage, porous materials have more stable and effective performance. Many porous materials such as zeolites, aluminum silicates, metal oxides, aluminum phosphate, activated carbon, active alumina, carbon nanotubes, silica gel, mineral resins and polymers, metal organic framework (MOF) and composite organic metals as the adsorbent for hydrogen adsorption utilized. The number of these adsorbents are currently used in the industry [18]. The most important adsorbents for hydrogen adsorption are zeolites, MOFs, activated carbon, nanostructures, and nanocomposites. In 1997, Dylon and colleagues applied carbon nanotubes as hydrogen adsorbers in hydrogen storage process [12]. The results showed that the hydrogen absorption potential for carbon nanotubes is 10–5% by weight.

The hydrogen storage process occurs on the surface of porous materials during electrochemical decomposition of an aqueous solution [19]. One of the most important challenges in this process is finding the effective hydrogen storage

materials. Metal oxides based nanocomposites are widely researched with the objective of replacing expensive noble metals due to their unique characteristics [20–22]. To enhancement the selectivity and catalytic performance of these nanocomposites, various parameters can be changed such as addition of doping agent and appropriate support with certain foreign cations [23,24]. In this process, materials should possess two characteristics: 1) high activity and 2) high surface area. Nickel oxide (NiO) is a p-type semiconductor with a band gap about 3.5 eV. This oxide has attracted a lot of attention due to its wide applications such as electrode materials for lithium-ion batteries and fuel cell [25], electrochemical supercapacitors [26], catalysts [27], and electrochromic films [28]. Ceria ( $\text{CeO}_2$ ) is an n-type semiconductor with the fluorite structure and band gap around 3.2 eV  $\text{CeO}_2$  has been extensively researched due to its high dispersion properties [29,30]. One of the features of ceria is the mobility of oxygen atoms that can leave easily the ceria lattice without losing its structural integrity [31]. Hence, in ceria lattice, oxygen vacancies are important in its efficiency as a doping matrix and also in hydrogen storage process (occupied via hydrogen). Theoretically, the formed p–n heterojunction structures could facilitate the transfer of electron-hole pairs and create a potential driving force. The chemical interaction between  $\text{CeO}_2$  and NiO is favorable for the hydrogen storage performance due to the enhancing number of surface sites of the sample and also, interstitial sites during the redox processes. In this work, we have prepared p-type NiO/n-type  $\text{CeO}_2$  nanocomposite as a newly developed class for electrochemically hydrogen storage. To the best of our knowledge, it is the first time that p-type NiO/n-type  $\text{CeO}_2$  mesoporous nanocomposites are synthesized by carbohydrate sugars as a capping agent. Also, there is no report on the p-type NiO/n-type  $\text{CeO}_2$  mesoporous nanocomposites in electrochemically hydrogen storage process.

**Table 1 – Preparation conditions of the synthesized p-type NiO/n-type  $\text{CeO}_2$  nanocomposite.**

Sample No	Capping agent	Decompose Time (h)	Decompose Temperature ( $^{\circ}\text{C}$ )
1	Fructose	3	500
2	Fructose	3	600
3	Fructose	3	700
4	Fructose	3	800
5	Fructose	2	500
6	Fructose	4	500
7	Glucose	3	500
8	Sucrose	3	500
9	Starch	3	500



**Scheme 1 – Effect of various capping agents on the morphology of as-synthesized p-type NiO/n-type  $\text{CeO}_2$  nanocomposite.**

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