

RESEARCH PAPER

Water Radiolysis by Gamma –Irradiation for High Quality Synthesis of Nickel Oxide Nano Sheet

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ABSTRACT

A simple, time effective and cost effective, method was reported for synthesis of nickel oxide nanostructure. The NiO nanostructure was synthesized by gamma-irradiation method from NiSO₄ precursor. The solution was irradiated by ⁶⁰Co gamma-ray source at the dose rate of 40 Gy/min for determined time. The synthesized powders are characterized by means of X-ray powder diffraction (XRD), Energy-dispersive X-ray spectroscopy (EDX), Scanning electron microscopy (SEM), and Fourier transform infrared (FT-IR) spectroscopy. The XRD and FTIR results confirmed the formation of high pure phase of NiO nanostructure. Results of scanning electron microscope (SEM) images showed that product has flake-like nanostructure morphology with average particle size of 60 nm. The mechanism of product formation through water radiolysis was discussed. Three main stages can be considered for the radiolytic reactions of water in different time scales. (a) The physical stage (1 fs after the initial radiation interaction), (b) Physico-chemical stage (10⁻¹⁵–10⁻¹² s), (c) The chemical stage (10⁻¹²–10⁻⁶ s).

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INTRODUCTION

Due to wide range applications of the bulky and nanoscale forms of nickel oxide in various fields such as catalysis, fuel cell electrodes, gas sensors, p-n heterojunctions, electrochromic films, battery cathodes, magnetic materials, smart windows, electrochemical supercapacitors, dye-sensitized photocathodes and photovoltaic devices many attentions have been regarded to its synthesis [1,2]. So the NiO is one of the most important transition metal oxides. Synthesis of NiO at nanoscale with different morphologies such as one-, two-, and three-dimensional nanostructures (particle, wire, tube, rod, sphere, and sheet) has recently found interesting research area in nanotechnology. Physico-chemical properties of materials depend on their sizes, in the other words at nano-scale

size because of enhancing their effective surface area and maximizing their contact surfaces a novel physico-chemical properties would be appeared [3-5]. Many methods such as sol-gel method, ultrasonic radiation, pyrolysis by microwave, hydrothermal synthesis, precipitation-calcination, carbonyl, laser chemical, mechanochemical processing, microemulsion, flame spray pyrolysis and solid-state, have been used for the synthesis of NiO nanostructures[6]. The synthesis of NiO nanostructures by the gamma-irradiation technique has many advantages compared with other methods. For example, the synthesis could be carried out at room temperature under atmosphere pressure, with great success [7]. So in the current work, gamma-irradiation and subsequent heat-treatment method for the

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preparation of NiO nanostructures was reported.

MATERIALS AND METHODS

All of the reactants were analytical-grade, and used without further treatment. NiSO₄·6H₂O Isopropyl alcohol, Tetramethyl ethylene diamine were obtained from Aldrich. Distilled water was used for solutions preparing. The obtained product were dried at 80 °C (Behdad Drying Oven, 01 145, IRAN) and then characterized by X-ray diffraction (Phillips, STOE XD-3A), Scanning electron microscopy (TESCAN VEGA3 SB), and Fourier transform infrared (FT-IR) spectroscopy of the sample was recorded with a KBr pellet on a BOMEM MB spectrometer ranging from 400 to 4000 cm⁻¹.

Synthesis of NiO

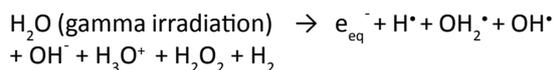
A 0.01 M NiSO₄·6H₂O solution was prepared in distilled water and isopropyl alcohol (3:1). The role of isopropyl alcohol can be indicated as: a) trapping and removal of some oxidative radicals and b) adjusting of solution dielectric constant for Ni(OH)₂ precipitation and prevent form its dissolution. The Tetramethyl ethylene diamine was added to solution in 1:1 ratio relative to Ni²⁺ ions (as complexing agent). Then the solution was irradiated by ⁶⁰Co gamma-ray source at the dose rate of 40 Gy/min. After the centrifuge of solution the obtained precipitate was dried at 80 °C for 2 h,

then the heat treatment of hydroxide was carried out at 400 °C with temperature rate of 10 °C/ min.

RESULT AND DISCUSSION

Synthesis mechanism

The mechanism of water radiolysis by gamma irradiation can be written as[8] :



The oxidative radicals such as OH₂[•] and OH[•] are trapped by isopropyl alcohol and removed from media (Fig. 1). Three main stages can be considered for the radiolytic reactions of water in different time scales [8]:

(a) The physical stage (1 fs after the initial radiation interaction): The formation of ionized (H₂O⁺), excited water molecules (H₂O*) and sub-excitations electrons (e⁻) take place in this stage. It consists the energy deposition followed by fast relaxation processes.

(b) Physico-chemical stage (10⁻¹⁵–10⁻¹² s): Numerous processes occur, including ion-molecule reaction, dissociative relaxation, auto ionization of excited states, thermalization of subexcitation electrons (solvation of electrons), hole diffusion, etc.

(c) The chemical stage (10⁻¹²–10⁻⁶ s): The species react in the tracks and then diffuse in solution.

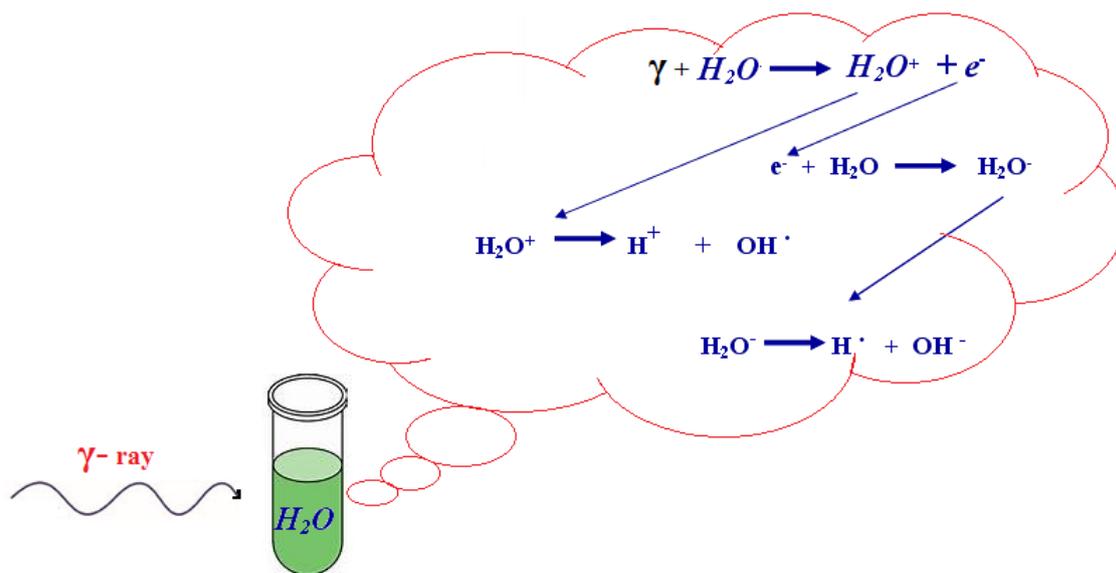
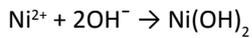


Fig. 1. Mechanism of water radiolysis by gamma radiation

They can thus react with each other and also with surrounding molecules (in the solute).

The produce of OH⁻ in the solution causes the increase in pH and the deposition of Nickel ions as Hydroxide:



XRD

Fig. 2 is XRD patterns of the prepared NiO material annealed at 400 °C. As can be seen the main diffraction peaks are observed around 2θ = 37.2, 43. 2, 62.8, 75.3 and 79.3° which are attributed to (111), (200), (220), (311) and (222) planes (Miller indices) which they could be indexed as diffraction peaks of the nickel oxide (ICDD Reference code: 01-073-1523)[9]. The XRD spectrum with clear and sharp peaks (and no impurity peaks) confirm the phase purity of the obtained product.

Morphology

To obtain the insight information about the surface morphology and particle size of the obtained NiO, the SEM analysis was performed. Fig. 3 shows the SEM images of sample in two different magnifications. As can be seen the SEM images reveal that the product consisted

the large spherical nanostructures with average size of about 50 nm. The agglomeration of these spherical structures lead to honeycomb outward morphology. Carefully and detailed look at the images indicates that each fine spherical nano structures consist of large number of sub-nano sheets with average width and length of 5 and 10 nm respectively. The fine-sheets are interconnected to each other and have given a porous and flower-like appearance to the spherical nanostructures. Base of on the synthesis conditions the porous structure and sheet-like morphology of nano structures is result of hydrogen bubbling during radiolysis of water molecules. In other words the H₂ bubbles act as dynamic template for fine-sheet growth and nanostructure formation.

The EDX analysis was performed to determine of the elements and their percentage in the sample. The EDX spectra of obtained NiO is shown in Fig. 3(inset). From the atomic and weight percentages of the elements in EDX spectra the NiO formation can be confirmed.

FTIR

The FTIR spectrum of the as-prepared and temperature annealed samples is shown in Fig 4. The spectrum was recorded in the range of 4000cm⁻¹ – 400 cm⁻¹. The broad absorption band at (can

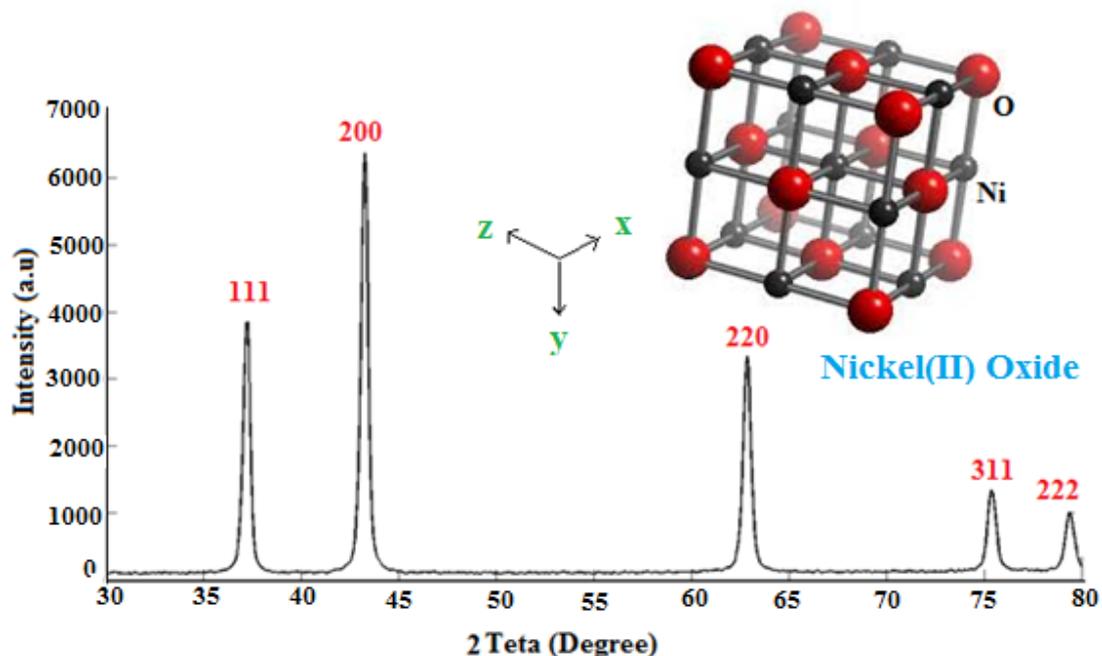


Fig. 2. XRD pattern of NiO nanostructure

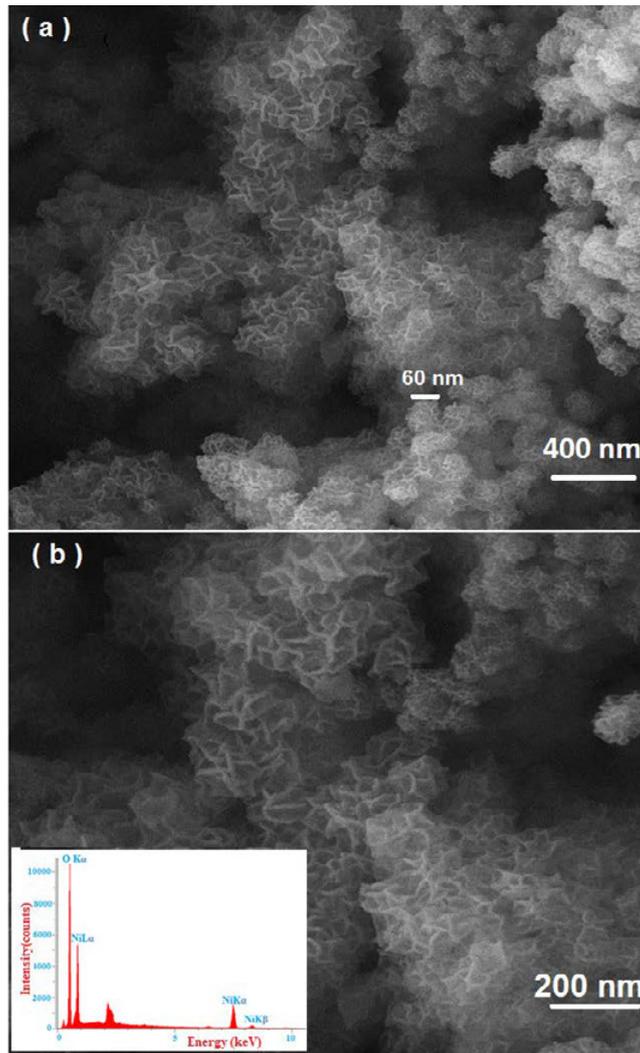


Fig. 3. (a) and (b) SEM images and EDX spectra (inset) of NiO

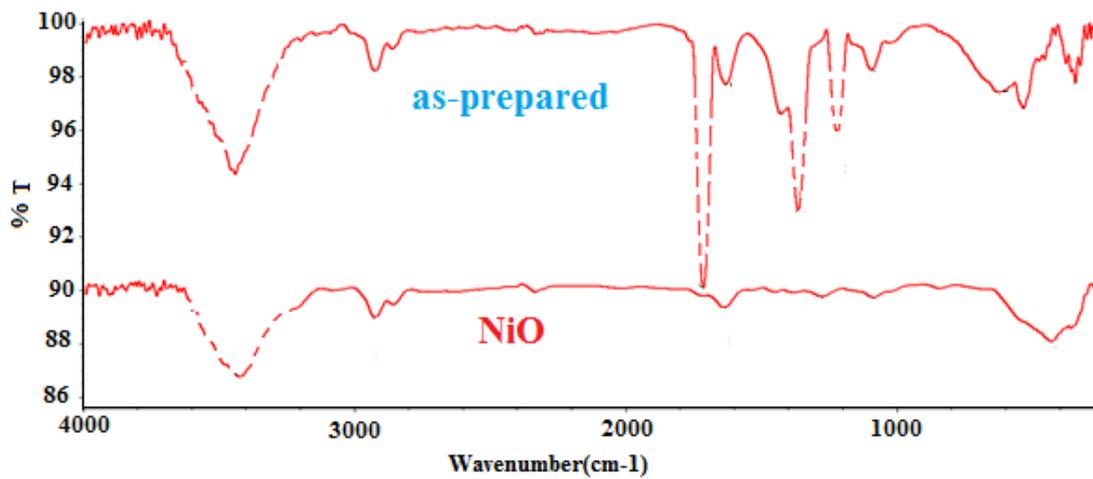


Fig.4. FT-IR spectra of as prepared and NiO samples

be seen in two samples) 3440 cm^{-1} is attributed to the band O–H stretching vibrations[10]. As can be seen from the figure, the intensity of this band is reduced in heat annealed sample indicating which the despite drying this sample contained traces of water. The two bands at 1631 and 1093 cm^{-1} to O–H bending vibrations associated with Ni atoms. The sharp peaks at 1260 cm^{-1} can be assigned to asymmetric S=O stretching vibration mode of SO_4^{2-} anions intercalated in the structure of the deposit in the as prepared sample (The $\text{NiSO}_4 \cdot 5\text{H}_2\text{O}$ reagent was used to prepare the 0.01M Ni^{2+} solution) [11]. Also in as prepared sample the contact of trapped water in the deposition with CO_2 could lead to the formation of carbonate species characterized by the C=O peak at 1715 cm^{-1} . The peak is observed at 440 cm^{-1} , is correspond to Ni–O stretching mode [12].

CONCLUSION

Nanostructured particles of NiO have been successfully synthesized through radiolysis.

method using $\text{NiSO}_4 \cdot 6\text{H}_2\text{O}$ as precursor and isopropyl alcohol as radical trapping agent.

For confirm the NiO nano structures formation the product was characterized with X-Ray Diffraction (XRD), scanning electron microscopy (SEM) and Fourier transform infrared (FT-IR). The SEM results confirm the nano scale of particle with spherical shape. The presence and interconnection of sub-nano sheets created a porous flower-like morphology. The mechanism of base generation through water radiolysis and nickel hydroxide formation was discussed. In summary this simple, time effective and cost effective, synthesis method will be useful for industries for the preparation of pure phase nickel oxide nanostructure.

CONFLICT OF INTEREST

The authors declare that there are no conflicts of interest regarding the publication of this manuscript.

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