



Research article

Utilizing of neodymium vanadate nanoparticles as an efficient catalyst to boost the photocatalytic water purification



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ABSTRACT

Neodymium Vanadate (NdVO₄) nanostructures were successfully synthesized *via* a modified solid state method in the presence of ligand. These nanoparticles were further used as a photocatalyst. Primarily the best structural formations and smallest crystallite sizes of the systems were identified and optimized by changing the calcination time, calcination temperature and molar ratio of the ligand. The cationic (Methyl Violet (MV)), and anionic (Eosin Y (EY) and Eriochrome Black T (EBT)) dyes were used as a model to evaluate the photoactivity under UV-Vis irradiation. Several operational factors were examined to improve the photocatalytic efficiency include type of dye, type of light source, pH and dye concentration. As a result, the best efficiency in 5 ppm Eriochrome Black T at pH = 11 was achieved in the presence of 0.05 g NdVO₄ nanocatalysts.

1. Introduction

The organic dyes (cationic and anionic dyes) are one of the most important environmental pollutants that are used in various industries such as textile, paper printing and pulp, rubber, plastics, leather, cosmetics, etc (Şahin et al., 2013; Elijah and Nwabanne, 2014). Aquatic environmental deterioration is a serious issue due to rapid urbanization and economic development; hence, the reductions of the hazardous contaminants from waste water are urgently required. Electroactive nanocatalysts are recently achieved a great attention in order to remove colorants from aquatic environments. M_x - orthovanadates (M: Na, Ca, Ba, Sc, Er, Nd ...) are the inorganic compounds which consider for this application (Mahapatra et al., 2007). Among these compounds, neodymium vanadate is an important category of rare earth (RE) orthovanadates that has been synthesized by various techniques such as hydrothermal (Wu et al., 2005), solid-state reaction (Errandonea et al., 2014), sonochemical (Monsef et al., 2018), precipitation (Vosoughifar, 2016), sol-gel (Au et al., 1996) and microwave (Mahapatra et al., 2008a). NdVO₄ powders are renowned as promising materials in photocatalytic, electrical, catalytic, laser, photoluminescence, optic, phosphors, polarizer properties (Mahapatra et al., 2008b; Yuvaraj et al., 2014; Selvan et al., 2009; Yao et al., 2010; Verma et al., 2016; Deng et al., 2008; Nguyen et al., 1997). Rare earth orthovanadates, especially in nanosized state, are highly promising for biomedical applications due to their high stability and nontoxicity (Lyadov and Kurilkin, 2016). Moreover, transition metal vanadates have been also reported for

various academic and industrial applications; for example cobalt vanadate, copper vanadate, nickel vanadate, iron and zinc vanadate (Ghiyasiyan-Arani et al., 2016a, 2016b, 2016c, 2017, 2018; Ghiyasiyan-Arani and Masjedi-Arani, 2016; Mazloom et al., 2016).

In recent years, nanoscale neodymium vanadate has been achieved a great interest in its effective photocatalytic properties. It has been found that NdVO₄ as a wide-bandgap semiconductor with a zircon-type structure and I41/amd space group shows a photocatalytic activity for degradation of organic contaminants which is comparable or even higher than that of the commercial TiO₂ (Dragomir et al., 2013). But due to its wide band gap (> 3 eV), NdVO₄ is a UV active material. However, the previous research indicates under visible light, approximately 66% of RhB can be degraded by the catalysis of NdVO₄ nanowires over 6 h of illumination. This degradation can be attributed to the electron transitions of 4f electrons, the regular VO₄ tetrahedra. The regular VO₄ tetrahedra and Nd³⁺ of NdVO₄ play a crucial role in the photocatalytic process (Jing et al., 2011). In the current study, neodymium vanadium oxide was synthesized *via* a traditional solid state method in the presence of H₂acacen ligand as a Schiff base capping agent. This method is known as a facile, low-cost, green and solvent-free. In the literatures, the NdVO₄ powders have been prepared by SSR (Solid state reaction) method, where the particles are in the range of 1–5 µm (Dragomir et al., 2013); however, in this work, nanoscaled NdVO₄ ceramics are formed (~80 nm) and optimized by changing experimental conditions. These nanoceramics play a potential role as an impressive semiconductor in photocatalysis process for degradation of

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Table 1
Preparation conditions of NdVO₄ by solid state method.

Sample No.	Solvent	Schiff-base ligand	Molar ratio (Nd:H ₂ acacen)	Calcination temperature (°C)	Time(h)	Product
1	–	–	–	700	6	NdVO ₄
2	–	H ₂ acacen	1:0.5	700	6	NdVO ₄
3	Methanol	H ₂ acacen	1:0.5	700	6	NdVO ₄
4	Methanol	H ₂ acacen	1:1	700	6	NdVO ₄
5	Methanol	H ₂ acacen	1:2	700	6	NdVO ₄
6	Methanol	H ₂ acacen	1:2	700	4	NdVO ₄
7	Methanol	H ₂ acacen	1:2	700	2	NdVO ₄
8	Methanol	H ₂ acacen	1:2	600	2	NdVO ₄ /Nd ₂ O ₃
9	Methanol	H ₂ acacen	1:2	500	2	NdVO ₄ /Nd ₂ O ₃ /V ₂ O ₅

organic dyes. Eriochrome Black T, Methyl Violet, and Eosin Y were selected as pollutants; Eriochrome Black T is an azo dye that is used for tintinction silk and wool (Vaiano et al., 2017), Eosin Y and Methyl Violet are anionic and cationic dyes used in ink and textile industries (Oyelude et al., 2017; Mendhulkar Vijay et al.).

Our study aims to provide optimal frameworks in the synthesis of pure, uniform and nanoscale catalysts by applying Schiff-base ligand and also changing the synthesis conditions. Stability and recyclability of NdVO₄ nanoparticles is an important factor in water treatment. Effect of different parameters on photoactivity of the samples such as effect of type of dye, type of irradiation source, pH and dye concentration was investigated to improve efficiency of catalyst function.

2. Experimental

2.1. Materials and method

All reagents for the preparation of NdVO₄ nanoparticles such as neodymium nitrate hexahydrate (Nd(NO₃)₃·6H₂O), ammonium metavanadate (NH₄VO₃), methanol, ethylenediamine (en) were purchased from Merck Company and used as received without further purification. Bis (acetyl acetanato) ethylene diamine Schiff base (H₂acacen) was prepared to consider the published procedure (Motahari et al., 2015). Briefly, the stoichiometric amounts of acetylacetonate solution and ethylenediamine were heated in 40 ml ethanol under reflux for 4 h. The product was separated and washed with ethanol and dried overnight in an oven at 60 °C.

NdVO₄ nanostructures were synthesized by a general solid state reaction in the air using Nd(NO₃)₃·6H₂O and NH₄VO₃ as the starting materials in the presence of the as-prepared H₂acacen Schiff-base. In a typical experiment, 0.25 g of Nd(NO₃)₃·6H₂O and 0.067 g NH₄VO₃ (Nd:V/1:1) were grinded carefully, and then various amounts of H₂acacen were added. The admixture was further milled by zirconia ball-mill for 30 min using 5 drops methanol as a solvent and homogenizing agent. The reaction admixture was dried and calcined in an alumina crucible at 500, 600 and 700 °C. The preparation conditions are summarized in Table 1.

2.2. Characterizations

The phase structure of the products was identified using X-ray powder diffraction measurements (Philips-X'pertpro (by Ni filter and Cu Ka lines as a radiation source (λ = 1.54 Å). Fourier transform infrared (FT-IR) spectra of the samples diffused in KBr pellets were obtained using Nicolet Magna- 550 spectrometer at room temperature. The morphology and particle sizes of the samples were observed using a

LEO-1455VP scanning electron microscope (SEM) and transmission electron microscope (TEM) by a Philips EM208 operating at an accelerating voltage of 200 kV. The energy dispersive spectrometry (EDS) was used in order to determine the elements of compounds with an accelerating voltage of 20 kV. Solid UV–Visible diffuse reflectance spectrometer (DRS) was performed with a Shimadzu UV-3101PC, UV/Vis spectrophotometer. The magnetic properties of the NdVO₄ were studied by a vibrating sample magnetometer (VSM, Meghnatis Kavir Kasha Co., Kasha, Iran).

2.3. Photocatalytic measurement

To inquire the photocatalytic activity of the NdVO₄ nanoparticles, a series of control experiments was carried out. Photodecolorization experiments were conducted in a photoreactor using a 400 W mercury lamp and 400 W Osram lamps as the irradiation sources at room temperature. Experimentally, an exact amount of the NdVO₄ sample (0.05 g) was added in to the organic pollutant solutions (Eriochrome Black T, Methyl Violet, Eosin Y) in different conditions. The above suspension was transferred into the reactor, stirred and aerated in the dark for 30 min to attain adsorption-desorption equilibrium between the catalyst and dye substrates. This suspension was subjected to light irradiation where 5 ml of the solution was taken at certain times. At the end of the experiment, the samples were centrifuged to separate the catalysts. Concentration and absorbance of dye solutions was determined by UV–vis spectrophotometer. The amount of dye degradation (D.P.) with standard deviation at the sampling time was calculated by means of Eq. (1). In this equation, C₀ and C_t are the absorbance quantity of the sample at 0 and t minute, respectively (Error bars in the Fig. 11 are standard deviations in average percentage destruction).

$$D. P. (t) = \frac{C_0 - C_t}{C_0} \times 100 \quad (1)$$

3. Result and discussion

3.1. Phase compositions and morphological observations

3.1.1. X-ray diffraction patterns

The XRD patterns were used to identify the phase structures of NdVO₄ powders (Fig. 1(a-c)). The results indicate that all the samples are highly pure and crystalline. The diffraction peaks can be ascribed to the tetragonal phase of NdVO₄ (JCPDS 15-0769; space group: 141/amd). In order to study the effect of temperature and time on the lattice structure of NdVO₄, the respective diffractograms (Fig. 2(a-d)) indicate that the samples (Samples 8 and 9) at reduced calcination temperatures

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