

# Sol Gel Synthesis of $\text{CeVO}_4$ using (*Senna occidentalis* (L.) Link) Leaves Extract: Characterization and Photocatalytic under Visible Light Irradiation

Eka Mardika Handayani<sup>1,2</sup>, Muryanto Muryanto<sup>3</sup>, Oman Zuas<sup>4</sup>, Yoki Yulizar<sup>2\*</sup>

<sup>1</sup>Center for Research and Human Resource Development, National Standardization Agency of Indonesia Kawasan PUSPIPTEK, Serpong 15314, Tangerang, Indonesia

<sup>2</sup>Department of Chemistry, Faculty of Mathematics and Natural Sciences, Universitas Indonesia, Depok 16424, Indonesia

<sup>3</sup>Research Centre for Chemistry, National Research and Innovation Agency (BRIN), Kawasan PUSPIPTEK, Serpong Tangerang Selatan, Banten, 15314, Indonesia

<sup>4</sup>Research Centre for Testing Technology and Standard (PRTPS), Research Organization for Energy and Manufacture (OREM), National Research and Innovation Agency (BRIN), Building 417, Kawasan PUSPIPTEK, Serpong Tangerang Selatan, Banten, 15314, Indonesia

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Corresponding Author:

Yoki Yulizar

[yokiy@sci.ui.ac.id](mailto:yokiy@sci.ui.ac.id)

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**Abstract:**  $\text{CeVO}_4$  as visible light driven photocatalyst was synthesized using *Senna occidentalis* (L.) Link leaves extract (SOE) by sol-gel method which can increase the degradation of malachite green (MG) under visible irradiation. The products were characterized using Fourier Transform Infrared Spectrophotometer (FTIR), X-ray Diffraction (XRD), and Field Emission Scanning Electron Microscope Energy Dispersive Spectroscopy (FESEM-EDS). UV-Vis Diffuse Reflectance Spectrophotometer (UV-Vis DRS) absorption spectrum indicated that the band gap of the  $\text{CeVO}_4$  is about 2,76 eV, and XRD patterns of the samples were indexed to tetragonal zircon type  $\text{CeVO}_4$  structure. Photocatalytic activity of the synthesized  $\text{CeVO}_4$  nanoparticles was evaluated for degrading the MG under visible light irradiation, showing as an active catalyst on photodegradation of the MG up to 77% under experimental condition of this study.

**Keywords:**  $\text{CeVO}_4$ ; Green synthesis; Photocatalytic; Sol-gel; Visible light

## Introduction

Nowadays, green synthesis as an alternative method for synthesizing nanoparticles still have attractive attention because of environmentally friendly, cost-effective, and produce high purity product. Green synthesis method uses biological systems such as plants, algae, fungi, bacteria, and biological molecules. Among them, plants could be the best alternative in nanomaterials synthesis due to their availability, simple and safe preparation (Ariyanta et al., 2020; Salem & Fouada, 2020).

*Senna occidentalis* (L.) Link is shrubs that contains secondary metabolites (alkaloids, tannins, flavonoids, saponin, and phenols (Daniel Azubuike, 2015). The

secondary metabolites in *Senna occidentalis* (L.) Link extracts (SOE) has a role as sources of a weak base and capping agent in synthesis of nanoparticles (Akinkunle et al., 2019; Yulizar et al., 2021).

Cerium vanadate ( $\text{CeVO}_4$ ) is a semiconductor metal based orthovanadates and has been widely studied in many area including in catalysis, lithium ion batteries, gas sensors and photocatalytic fields (Rahimi-Nasrabadi et al., 2017; Kumar et al., 2017). The synthesis of  $\text{CeVO}_4$  has been previously reported using different methods: solid-state reaction, hydrothermal, precipitation, microwave method, ultrasound, sono-chemical, and sol-gel (Phuruangrat et al., 2021; Zheng et al., 2015; Mosleh & Mahinpour, 2016).

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In this work,  $\text{CeVO}_4$  was synthesized via sol-gel using *Senna occidentalis* (L.) Link for the first time. The advantages of sol-gel method are simple, controllable of microstructural property, high purity crystal and low cost (Phuruangrat et al., 2021). The structure of the synthesized  $\text{CeVO}_4$  nanoparticles were structurally characterized and their activity was assessed toward photocatalytic degradation of Malachite green (MG) under visible light irradiation.

## Method

### Material and methods

The commercial Malachite green (MG), Cerium (III) nitrate hexahydrate  $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  (98 %), and ammonium metavanadate ( $\text{NH}_4\text{VO}_3$ ) were purchased from Merck Chemical Co, Germany. All the materials used were of analytical grade and used without special treatment. Deionized water ( $0.055 \mu\text{S}/\text{cm}$ ) produced by a water purification system (Millipore Milli-Q) was used in all experiment runs. Fresh leaves of (*Senna occidentalis* (L.) Link) were collected from Kp. Blentuk, Cimahpar, Bogor, West Java, Indonesia. The plant was identified in the Botanical Laboratory, Directorate of Scientific Collection Management, National Research and Innovation Agency (BRIN).

### Preparation of *Senna occidentalis* (L.) Link extracts (SOE)

*Senna occidentalis* (L.) Link leaves powder was macerated using methanol in ratio 1:5 (w/v) for a week under keep stirring at least 20 min in every single day. After that, the methanol filtrate was partitioned using n-hexane in ratio 1:1 (v/v) to get fractions of methanol and n-hexane. Methanol fraction was concentrated, and the residue was diluted by deionized water, which was the aqueous fraction of SOE was used for  $\text{CeVO}_4$  synthesis. This method was adopted from Indriyani et al., (2021) with slight modification.

### Synthesis and characterization of $\text{CeVO}_4$ nanoparticles

In a typical procedure,  $\text{CeVO}_4$  nanoparticles were synthesized by sol-gel method as follows. First, 100 mL 0,1 M ( $\text{Ce}(\text{NO}_3)_3 \cdot 6\text{H}_2\text{O}$  and 100 mL  $\text{NH}_4\text{VO}_3$  (molar ratio 1:1) were mixed and dissolved with deionized water under stirring. Then the mixture was gradually added with 20 mL of 1% of SOE under a constant stirring at 80 °C for one hour. Subsequently, the gel precursors were dried by heating at 100°C for 12 h and followed by consecutively washing with ethanol and water. The dried powder was then calcined at 400 °C for two hours.

The X-Ray diffraction patterns of the material was recorded in a XRD, (Rigaku SmartLab) at  $2\theta$  from 20 to 80°. Fourier Transform Infrared Spectrophotometer (FTIR) (Bruker-Tensor II) was recorded in the wavenumber range of  $500\text{-}4000 \text{ cm}^{-1}$ . Field Emission Scanning Electron Microscope – Energy Dispersive X-

Ray Spectrometer (FESEM-EDS) (JEOL JIB 4610F) was applied for the surface evaluation colour mapping. The UV-Vis Diffuse Reflectance Spectrophotometer (UV-Vis DRS) (Shimadzu 2450) was used to determine the optical band gap energy of the synthesized  $\text{CeVO}_4$  nanoparticles.

### The photocatalytic activity

Photocatalysis was conducted in a photoreactor fitted with Sodium lamps (Phillips SON 70 W, 220 V) as visible light source. The photocatalytic activities were evaluated by measuring the photocatalytic degradation of MG in the aqueous solutions. A 30 mL of  $5 \times 10^{-6}$  M MG aqueous solution and 10 mg of catalyst were mixed. The lamp irradiation was applied under continuous stirring for 120 min. The irradiated MG solution (6 mL) was taken in every 10 minutes, followed by UV-Vis measurement in the wavelength ranging from 200 nm to 800 nm.

## Result and Discussion

$\text{CeVO}_4$  was prepared by green synthesis method using *Senna occidentalis* (L.) Link leave extract SOE). SOE were extracted using methanol to acquire secondary metabolites in SOE. The advantages macerated using methanol are of low energy required because the process is occurred at low temperature (room temperature). Low temperature may prevent the destruction of secondary metabolites (Elviera et al., 2022). The synthesis of  $\text{CeVO}_4$  using water fraction of SOE obtained from evaporated methanol. Phytochemical qualitative analysis of the water fraction of SOE indicated that the component of the secondary metabolite compounds consisted of alkaloids, flavonoids and polyphenols. This findings are similar to the study reported by Daniel Azubuike, 2015; Tamasi et al., 2021). Alkaloids as sources of weak bases and flavonoids and polyphenols may act as capping agent and play an important role in avoiding the agglomeration of nanoparticle (Yulizar et al., 2020).

Crystalline structure of  $\text{CeVO}_4$  with 400 °C calcination was analyzed and the results is shown in Fig. 1. All the diffraction peaks indexed to the tetragonal  $\text{CeVO}_4$  a, which are in a good agreement with the JCPDS card No. 12-0757. All diffraction peaks at  $2\theta$  values of  $18.34^\circ$ ,  $23.89^\circ$ ,  $30.51^\circ$ ,  $32.52^\circ$ ,  $34.29^\circ$ ,  $36.89^\circ$ ,  $39.02^\circ$ ,  $43.51^\circ$ ,  $46.58^\circ$ ,  $49.54^\circ$ ,  $55.69^\circ$ ,  $60.29^\circ$ ,  $62.54^\circ$ ,  $68.09^\circ$  can be indexed to the (101), (200), (211), (112), (220), (202), (301), (103), (321), (312), (400), (213), (420), (004) (Ghotekar et al., 2018).

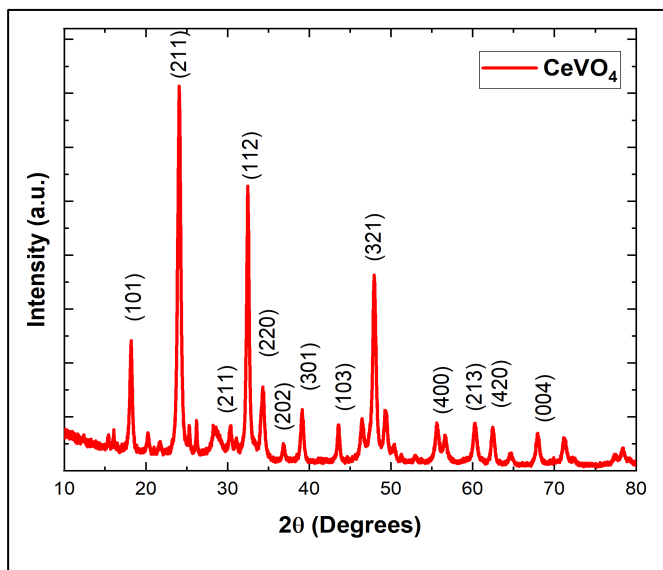


Figure 1. XRD patterns of CeVO<sub>4</sub>

The functional groups of the aqueous fraction of SOE are shown in Fig. 2 (red line). The O-H stretching vibrations at 3363 cm<sup>-1</sup>, the C-H stretching at 2925 cm<sup>-1</sup>, the bending of N-H at 1604 cm<sup>-1</sup>, C-C stretching at 1355 cm<sup>-1</sup>, C-O 1043 cm<sup>-1</sup>, and C-H aromatic 797 cm<sup>-1</sup>. (Kabala et al., 2020). For the FT-IR spectrum of CeVO<sub>4</sub> nanoparticles some vibrations appears at wavenumber 500, 714 and 750 cm<sup>-1</sup> as shown in Fig. 2 (black line), which are attributed to the Ce-O bonding and V-O bonding of VO<sub>4</sub><sup>3-</sup> (Muthuvel et al., 2020; Yulizar et al., 2020).

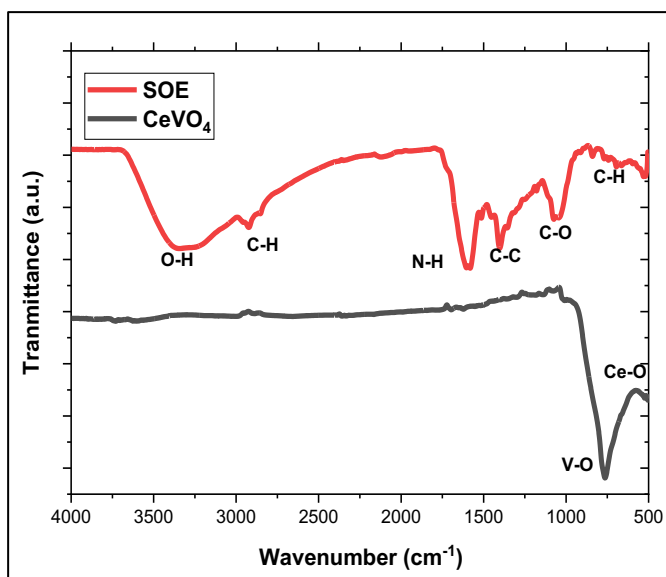


Figure 2. FT-IR spectra of *Senna occidentalis* (L.) Link leaves extract and CeVO<sub>4</sub>

The optical properties of CeVO<sub>4</sub> were investigated by UV-Vis DRS and calculated using the following Kubelka Munk formula, as equation 1.

$$\alpha hv = A (hv - E_g) A (hv - E_g)^2 \tag{1}$$

where  $\alpha$  is the absorption coefficient,  $h$  is plank's constant,  $\nu$  is light frequency,  $A$  is constant, and  $E_g$  is the band gap energy (Indriyani et al., 2021). Increase in electron-hole pairs generation can be indicated by the strong absorption of photocatalyst in the visible light area, which possibly enhances the photocatalytic activity of CeVO<sub>4</sub>. The optical band gap energy of the prepared CeVO<sub>4</sub> (Fig. 3) determined by the Kubelka-Munk plots is found to be 2.76 eV.

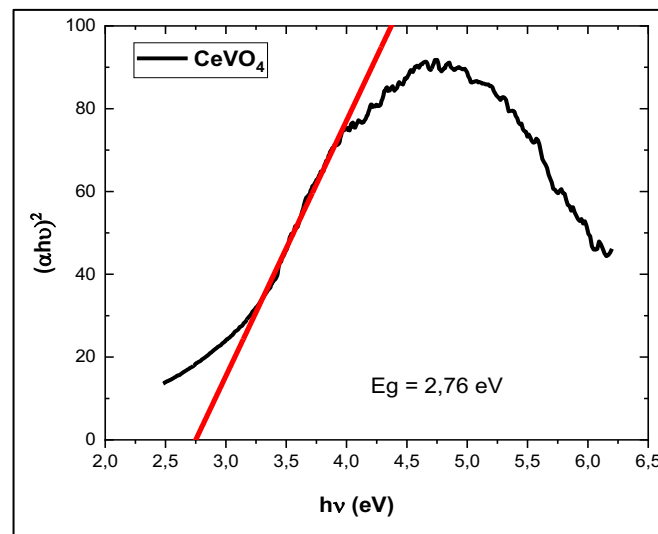
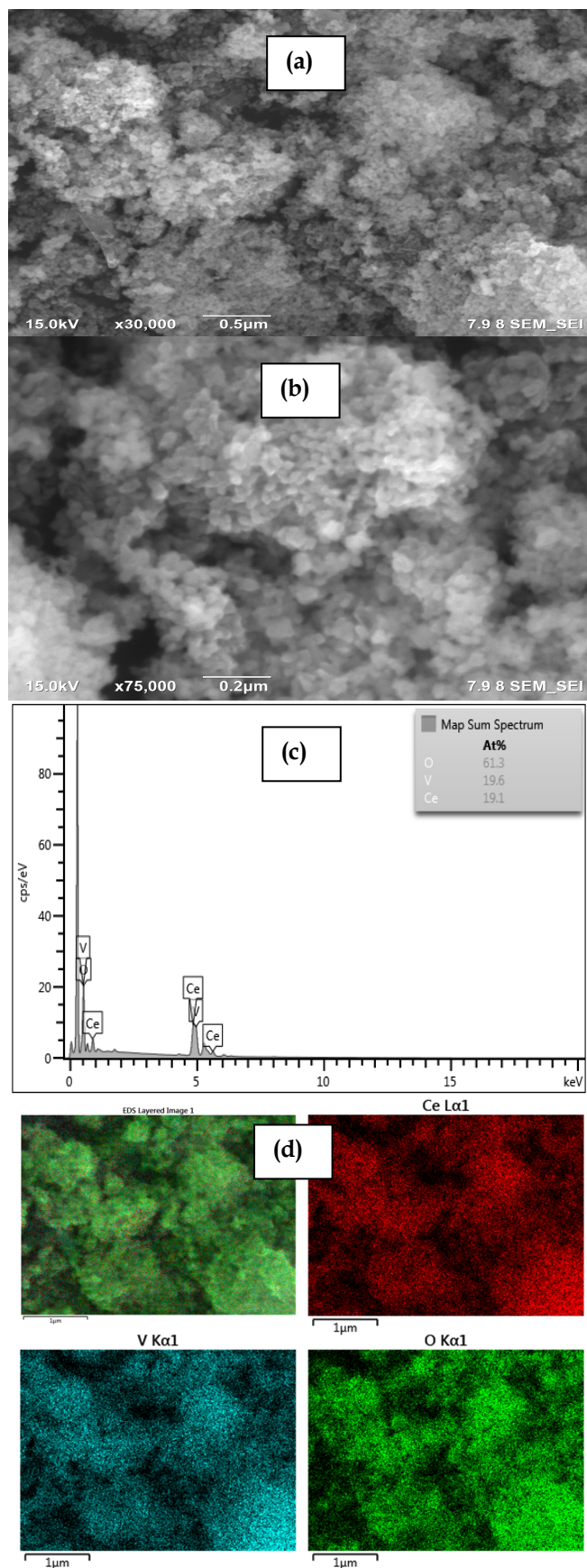


Figure 3. UV-Vis DRS spectra of CeVO<sub>4</sub>

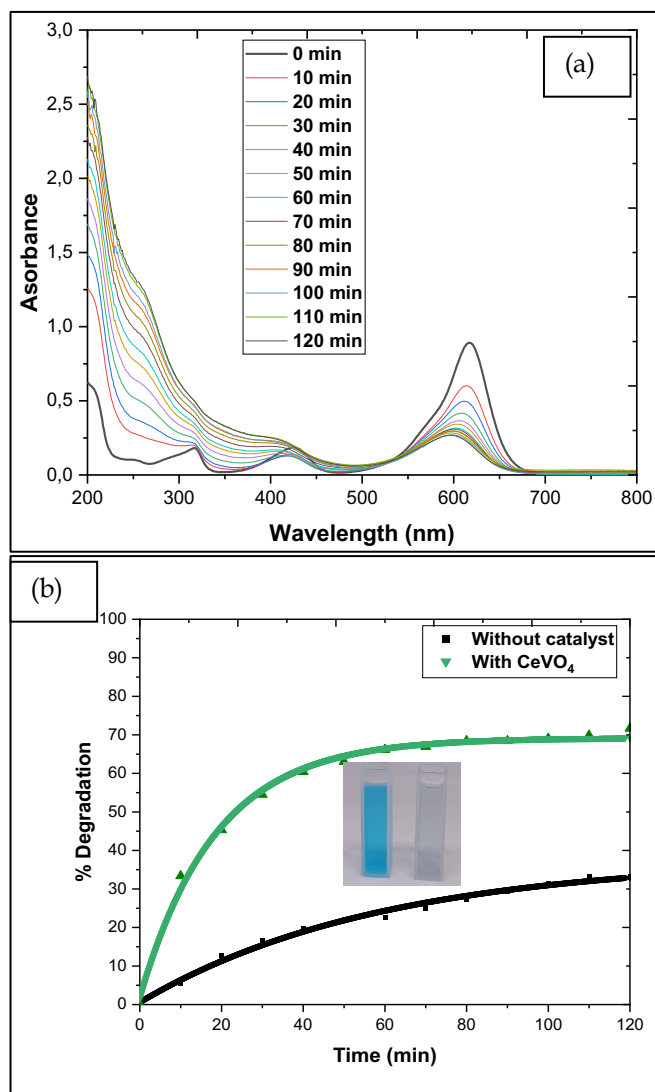
Morphology of the synthesized CeVO<sub>4</sub> was examined by FESEM technique at several magnifications. From Fig. 4, agglomerated spherical particles with uniform shape are obviously seen. The presence of the elements Ce, V, O in the catalyst were confirmed by EDX analysis from the particular selected area with an atomic percentage of 19.4% (Ce), 19.6% (V), and 61.3% (O). To confirm the presence of these elements, the evaluation was analyzed by using elemental mapping (Fig. 4c).

The photocatalytic activity of CeVO<sub>4</sub> nanoparticles was evaluated for MG solution as a model dye pollutant under visible light and the results are shown in Fig. 5 As shown in Fig 5a, the CeVO<sub>4</sub> rapidly degraded the MG, making the concentration of MG decrease to 77%. Interestingly, the concentration of MG was only degraded at about 33% without CeVO<sub>4</sub> in the system. This finding indicated that CeVO<sub>4</sub> could improve light harvesting, increase the number of photogeneration electron and holes that participate in photocatalysis reactions (Ekthammathat et al., 2013).





**Figure 4.** SEM images of CeVO<sub>4</sub> nanoparticles with a magnification at (a) 0,5 μm, (b) 0,2 μm, (c) EDX spectrum of CeVO<sub>4</sub> nanoparticles (d) Mapping image of CeVO<sub>4</sub>.



**Figure 5.** (a) UV-Vis absorption spectra of MG degradation using CeVO<sub>4</sub> nanoparticles (b) MG degradation percentage (inset: colour change of MG after 0 min (blue sol.) and 120 min (clear sol.) of reaction time)

## Conclusion

Tetragonal zircon type structure of CeVO<sub>4</sub> has been successfully synthesized via water fraction of *Senna occidentalis* (L.) Link leaves extract (SOE). Phytochemical screening and FTIR analysis of SOE shows that SOE contains some secondary metabolites such as alkaloids, flavonoids and polyphenols, which may act as weak base source and capping agent. The band gap energy of CeVO<sub>4</sub> was found to be 2.76 eV, shows that this catalyst could effectively work in visible region. The CeVO<sub>4</sub> was able to degrade MG up to 77%.

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