Preparation of Ta₂O₅ nanoparticles by using cathode glow discharge electrolysis

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Abstract
Tantalum pentoxide nanoparticles (Ta₂O₅ NPs) were fabricated by cathode glow discharge electrolysis (CGDE) generated between a needle-like platinum wire cathode and a tantalum foil anode in 6 g L⁻¹ Na₂SO₄ electrolyte solution containing 5 ml hydrofluoric acid (HF) and 0.075 g cetyltrimethyl ammonium bromide (CTAB). The chemical structure, composition and morphology of the obtained powder were analyzed by using XRD, FT-IR, SEM/EDS, XPS and UV–vis DRS. The results found that Ta₂O₅ NPs with orthorhombic structure and wide band gap (3.6 eV) are successfully fabricated at 500 V discharge voltage in about 3 h. CTAB as a stabilizing agent can reduce the agglomeration due to electrolysissolution and attaching the surface of the synthetic products. A possible preparation mechanism of Ta₂O₅ NPs is proposed. Firstly, the tantalum foil anode is oxidized to form a compact Ta₂O₅ layer. Then, Ta₂O₅ surface is etched to form soluble [TaF₇]⁻ complexes in the presence of HF. After that, soluble [TaF₇]⁻ complexes can react with H₂O to form Ta(OH)₃. Finally, Ta(OH)₃ is further converted to Ta₂O₅ from plasma-liquid interface into solution.

1. Introduction

Nanoparticles (NPs) of transition metal oxides have drawn extensive concerns because of their potential in purification, bioapplications, nanoelectronics and optoelectronics [1–3]. Among them, tantalum pentoxide (Ta₂O₅) is one of the most significant transition metal oxides and has aroused considerable interest over the past ten years because of its wide band gap (E_g ~ 4.0 eV), high refractive index (n ~ 2.2), high dielectric constant (ε ~ 24), excellent chemical and thermal stability, and good biocompatibility [4–6]. Based on above properties, Ta₂O₅ NPs has been widely utilized in different fields such as orthopedics, photovoltaic devices, electronic industries, gas sensors and antireflective layer material. In addition, Ta₂O₅ NPs is also an important photocatalyst and has been used successfully for the hydrogen production from splitting of water and photodegradation of organic pollutants [6, 7] owing to its more negative conduction band potential compared to TiO₂.

Up to date, many methods such as anodization methods [8–10], sol-gel technique [11], hydrothermal oxidation [12], magnetron sputtering method [13], pulsed laser deposition [14], electrospinning [15], solvothermal [16] and microemulsion [17] have been used for the fabrication of Ta₂O₅ NPs with various morphologies including mesoporous, spheres, nanowires, nanotubes, nanorods, nanoparticles [8–17]. However, most of them have some significant drawbacks, for example, complex reaction and treatment process, poor crystallinity, or very narrow spectrum response range [17].

The glow-discharge electrolysis (GDE) is a novel unusual electrochemical process, in which a light emitting plasma is produced around one of the electrodes immersed inside a liquid electrolyte [18]. In the GDE process, it involves plasma–liquid interfaces where many active species, i.e., H, O, HO, and H₂O₂ are generated according to the non–faradaic effects and can induce some extraordinary chemical reactions, such as, oxidation, reduction, addition and elimination reactions [18–20]. Therefore, over the past two decades, GDE has been widely used for water purification [21], surface modification [22], synthetic chemistry [23, 24], and spectral analysis [25].

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However, there are few reports for the preparation of NPs from bulk material using GDE [26, 27]. Recently, spindle-like CuO NPs were prepared in a single-step approach by using cathode glow discharge electrolysis (CGDE) [28]. Compared with other methods, CGDE technique can be an environment-friendly technique since the NPs are produced with no need any additional chemicals. In addition, CGDE has mild reaction conditions and simple steps [26–28].

Herein, Ta₂O₅ NPs was successfully fabricated in 6 g L⁻¹ Na₂SO₄ electrolyte containing 5 ml hydrofluoric acid (HF) and 0.075 g cetyltrimethyl ammonium bromide (CTAB) by CGDE technique, in which Pt needle point and tantalum foil were served as cathode and anode, respectively. The active species in the CGDE were determined by optical fiber spectrometer for clarifying the formation processes of Ta₂O₅. The structure, composition and morphology of Ta₂O₅ NPs were analyzed by FT-IR, XRD, SEM/EDS and XPS. The band gap energy of Ta₂O₅ NPs was obtained based on UV–vis DRS. In addition, a possible synthesis mechanism of Ta₂O₅ NPs was proposed.

2. Experimental

2.1. Materials

Sodium sulphate (Na₂SO₄), hydrofluoric acid (HF), and cetyltrimethyl ammonium bromide (CTAB) were analytical purity and supplied by Sinopharm Reagent Co. Ltd. Tantalum foil (>99.99%) was purchased from Qinghe County Jinjia Metal Material Co. Ltd.

2.2. Experimental device and process

The device was composed of a 200 ml polytetrafluoroethylene (PTFE) reactor and a LW 100J1 DC high voltage power supply as presented in figure 1. The reactor was furnished with a Pt needle point (0.5 mm diameter) cathode and tantalum foil (100 mm × 5 mm × 0.2 mm) anode. The gap between the cathode and anode was 5 mm. The reactor vessel was charged with 150 ml of electrolyte solution containing 6 g L⁻¹ Na₂SO₄, 5 ml HF and 0.075 g CTAB. The reactor vessel was placed in a circulation water bath with 25 °C. During the process of preparation, the solution was constantly stirred at 120 r min⁻¹. When the applied voltage was over 450 V, the light emitting plasma was generated in the solution around the Pt point. The discharge was continued for 3.0 h at 500 V and tantalum foil was gradually consumed forming a large quantity of white Ta₂O₅ NPs. The products were collected by centrifugation at 12,000 r min⁻¹, washed with distilled water and ethanol several times, dried at 60 °C, and then grinded for further analysis.
2.3. Characterization

The active species produced by CGDE under the preparation of Ta$_2$O$_5$ NPs process were detected using optical emission spectroscopy (OES, Avaspec-2048–8 optical fiber spectrometer, Avantes). The integration time was 200 ms and the spectral resolution was 0.1 nm. The DIGILAB FTS 3000 Fourier transform infrared spectroscopy (FT-IR) with a KBr pellet and the Rigaku D/max-2400 X-ray diffractometer (XRD) with CuK$_\alpha$ radiation ($\lambda = 0.154 \text{ nm}$) were utilized to analyze the structural of Ta$_2$O$_5$ NPs. Surface morphology and elemental composition of Ta$_2$O$_5$ NPs were performed by using a JSM-6701F scanning electron microscopy/energy dispersive x-ray spectroscopy (SEM/EDS). The chemical state of the sample is analyzed by PHI-5702 X-ray photoelectron spectroscopy (XPS). The band gap of Ta$_2$O$_5$ NPs was obtained using PuXin TU-1901 UV–vis diffuse reflectance spectroscopy (UV–vis DRS).

3. Results and discussion

3.1. Voltage-current curve of CGDE

Figure 2 illustrated the typical current-voltage ($I$–$V$) curve of CGDE in 6 g L$^{-1}$ Na$_2$SO$_4$ solution with 5 ml HF and 0.075 g CTAB. In the regions AB, the current increases with applied voltage, where Ohm’s Law is approximatively obeyed and normal electrolysis occurs with small gas bubbles liberating two electrodes [18, 29]. With further increasing the voltage (in the regions BC), the current becomes unstable and decreases severely, vapor sheath appears, and faint flashes are observed due to gas breakdown. Meanwhile, the $I$–$V$ curve in the regions BC presents a negative slope. This is because in BC region the surface of the Pt electrode is covered by a vapor sheath which undergoes the process of formation and collapse leading to the oscillation of the current [19, 20]. In addition, as the equivalent resistance of the formed gas film covering the Pt electrode increases dramatically resulting in a sharp decrease of the current [30]. Once the voltage reaches in CD region, the current remains nearly a constant because of forming a relatively stable vapor sheath on the Pt electrode surface [30]. This stable region has been termed as the Kellogg region. Moreover, in CD region, yellow glow is generated due to the presence of Na$^+$ ion in anolyte. Beyond point D, the current and the glow increase with applied voltage. In the process of glow discharge, superheating of the liquid electrolyte above its vaporization temperature and streamer discharge in the formed vapor bubble have been observed. This confirms that the discharge comes from Joule heating of the solvent and not gas evolution resulting from electrolysis [30–32]. During the preparation of Ta$_2$O$_5$ NPs, 500 V was selected as the optimal discharge voltage because of stable glow discharge, low energy consumption, and long operation life of Pt electrode.

3.2. The structural characterization of Ta$_2$O$_5$NPs

3.2.1. FT-IR analysis

Figure 3 illustrates the FT-IR spectra of products without (a) and with CTAB (b). The bands at 1640 and 3431 cm$^{-1}$ are ascribed to the bending and stretching vibration of O–H group in adsorbed water molecules. The peaks at 528 and 769 cm$^{-1}$ are attributed to the stretching vibrations of Ta–O bonds and Ta–O–Ta bridge, respectively [33, 34]. The results of FT-IR preliminarily suggested that the fabricated products can be Ta$_2$O$_5$ NPs using CGDE.
3.2.2. XRD analysis

Figure 4 presents the XRD patterns of as-prepared Ta$_2$O$_5$ NPs. The obvious diffraction peaks at $2\theta = 22.78^\circ$, $28.90^\circ$, $37.21^\circ$, $46.64^\circ$, $51.05^\circ$, $55.66^\circ$, $58.35^\circ$, $63.93^\circ$, and $71.44^\circ$ corresponded to (001), (210), (201), (002), (092), (0221), (2220), (2221), and (4160) lattice planes respectively are in good agreement with the standard JCPDS data (Card No. 25-0922) [11], demonstrating that the Ta$_2$O$_5$ with orthorhombic structure are successfully prepared. The sharp peak and high intensity indicated that prepared samples are NPs with good crystallinity. No peaks for other impurities are observed in the XRD patterns, indicating that the products have high purity. The XRD data further confirms that the pure orthorhombic phase Ta$_2$O$_5$ NPs are successfully fabricated.

The crystallite sizes of the Ta$_2$O$_5$ NPs are estimated by using the Debye–Sherrer equation at (001) diffraction peak [34]:

$$D = \frac{0.9 \lambda}{\beta \cos \theta}$$

The average crystallite sizes of the Ta$_2$O$_5$ NPs at (001) lattice planes are calculated to be about 23.5 and 21.4 nm for the samples a and b, respectively, which are almost equal. The approximate crystallite size and structure (orthorhombic structure) suggested that adding CTAB can only change the growth pattern of crystallite, not influence the composition of Ta$_2$O$_5$ NPs.
3.2.3. SEM analysis

Figure 5 shows the SEM images of sample without and with CTAB (b) at 500 V discharge voltage. As shown in figure 5(a), the particles are agglomerated severely, and almost appear to be large bulk TazO5. However, after adding surfactant CTAB (figure 5(b)), the agglomeration of particles is reduced significantly, and the particle size is decreased markedly. This is because CTAB is a cationic surfactant which ionizes completely in solution to form CTA+ . CTA+ possibly attached to the surface of the synthetic product by electrostatic interactions to reduce the surface Gibb’s free energy of TazO5 NPs, stabilize the particles and reduce agglomeration [28, 35]. In order to further examine the size distribution of TazO5 NPs, the particle sizes after adding CTAB (figure 5(b)) were also statistically analyzed via ImageJ software. It is found from the histogram (figure 5(c)), the average size of TazO5 NPs is about 325 nm, which is inconsistent with the size estimated by the Scherrer equation. The result illustrated that TazO5 NPs is formed by certain growth pattern of TazO5 nanocrystals under CTAB.

The EDS was used to determine the stoichiometry of the synthetic product. According to the figure 5(d), the product only consists of Ta and O elements. The EDS analysis of sample obtained the weight percent of tantalum and oxygen are found to be 82.39% and 17.61%, respectively. Therefore, the atomic ratio of O:Ta is calculated to be about 2.4, and near to the stoichiometry Ta2O5.

3.2.4. XPS analysis

Figure 6(a) shows the XPS full spectrum of as-synthesized TazO5 NPs. The characteristic peaks of Ta 4f, Ta 5p, Ta 5s, Ta 3d, Ta 4p, O 1s and O KLL further proved that the products are composed of Ta and O elements [36]. Figures 6(b) and (c) illuminated the O 1s and Ta 4f core level XPS spectra by the curve fittings using the XPS PEAK 4.1 program. As shown in figure 6(b), O 1s core spectrum consists of two peaks originated because of the presence of Ta–O and OH at the binding energy 530.58 and 532.20 eV [37], respectively. In figure 6(c), the chemical states of Ta 4f consist of a Ta 4f doublet at 26.25 and 28.09 eV [38], which are attributed to the 4f7/2 and 4f5/2 core levels of Ta5+ in TazO5 [38, 39]. No evidence is found for the presence of other Ta oxides, demonstrating the synthesized products are indeed TazO5. The result of XPS is well consistent with the analysis of XRD pattern and the EDS data regarding the stoichiometry of the TazO5 NPs.

3.2.5. UV–Vis DRS analysis

Figure 7 shows the UV–Vis DRS of samples. The UV absorption edge of sample is around 335 nm. The wavelength was consistent with that prepared by sol-gel [40]. According to the Tauc’s formula [34], the band gap value is about 3.6 eV, suggesting that prepared TazO5 NPs can be used as a photocatalyst.

3.3. Discussion of preparation mechanism

3.3.1. Spectral characteristics of CGDE

To explain the mechanism for the preparation of TazO5 NPs, optical emission spectra of CGDE were determined by using optical fiber spectrometer. Figure 8 shows a typical emission spectrum of CGDE. As can be seen from figure 8, the lines at 283.0, 289.0 and 309.0 nm are assigned to OH bands (A2Σ+ – X2ΠII) [25]. This is because electrons collide with H2O to generate a large number of OH radicals [41]. Ionic lines attributed to O II (458.5 and 463.1 nm) and atomic lines attributed to H (Hα, at 656.3 nm and Hγ at 486.1 nm) and O I lines (715.6, 777.4, 795.2 and 844.6 nm) are observed, which are generated when the water vapor around the Pt electrode is bombarded by high energy electrons [25, 41]. In addition, as the CGDE is achieved in Na2SO4 solution, the Na I atomic lines are also observed at 568.3, 588.9, 589.6 and 819.4 nm [25]. Based on the emission spectrum, we found that the radicals such as H·, O·, O2·−, ·OH are generated during the process of CGDE.

3.3.2. Calculation of plasma parameters

Plasma parameters such as electron temperature (Te), rotation temperature (Trot) and electron density (Ne) are important factors in studying the preparation mechanism of TazO5 NPs. The Te can be estimated by using the two-line intensity ratio method of Hα (656.3 nm) and Hγ (486.1 nm) according to the following equation (1) [42, 43]:

\[
T_e = \frac{(E_2 - E_1)}{k} \left( \ln \frac{A_2g_2\lambda_2}{A_1g_1\lambda_1} - \ln \frac{\lambda_1}{\lambda_2} \right)
\]  (1)

The values of E, g and A for Hα and Hγ are referred from the NIST databases. Based on the formula (1), the Te was approximately 2384 K at 500 V discharge voltage. The rotation temperature (Trot) is obtained through a simulated spectrum of OH at 305–320 nm wavelength range using LIFBASE software [44]. As shown in figure 9(a), at 2000 K, the simulated spectrum is coincided with the experimental data, suggesting that Trot is about 2000 K.
Figure 5. SEM image of the prepared Ta$_2$O$_5$ NPs (a) without and (b) with CTAB, (c) particle size histogram of (b), and (d) EDS of (b).
The electron density \( N_e \) can be estimated by using Stark-broadening of \( \text{H}_\alpha \) \[41]\:

\[
N_e = 2.42 \times 10^{17} (\Delta \lambda_{1/2})^{4.4713}
\]  

where, \( \Delta \lambda_{1/2} \) (nm) is the full width at half maximum (FWHM) of \( \text{H}_\alpha \) line. Figure 9(b) shows the \( \text{H}_\alpha \), 656.3 nm line and its Lorentz fitting curve. As shown in figure 9(b), the FWHM was 0.255 nm. So the electron density \( N_e \) was \( 3.24 \times 10^{16} \text{ cm}^{-3} \).
3.3.3. Proposed preparation mechanism

According to the experimental results, the possible mechanism of preparing Ta2O5 NPs was summarized in figure 10. First, the tantalum foil anode is oxidized to Ta2O5 on its surface (see Reaction (1) and (2)) [8].

\[
\text{Anode: Ta} \rightarrow \text{Ta}^{5+} + 5e^- \\
2\text{Ta}^{5+} + 5\text{H}_2\text{O} \rightarrow \text{Ta}_2\text{O}_5 + 10\text{H}^+ \tag{2}
\]

The overall reactions that occur during anodization can be described as Reaction (3) [8]:

\[
2\text{Ta} + 5\text{H}_2\text{O} \rightarrow \text{Ta}_2\text{O}_5 + 10\text{H}^+ + 10e^- \tag{3}
\]

Without the addition of HF, the surface of tantalum foil is oxidized to form a compact Ta2O5 layer which is self-limiting because of declining the current with increasing the film thickness. Adding HF, Ta2O5 surface was etched to form soluble [TaF7]2− complexes (Reaction (4)) [8, 45].

\[
\text{Ta}_2\text{O}_5 + 14\text{F}^- + 10\text{H}^+ \rightarrow 2[\text{TaF}_7]^{2-} + 5\text{H}_2\text{O} \tag{4}
\]

In the cathode, all kinds of active species including e\text{aq}, \text{H}, \text{OH}, \cdot \text{O}, \cdot \text{O}^{2-}, \text{OH}^-, \text{H}_2\text{O}_2, \text{H}_2 and \text{O}_2 are generated at the plasma–liquid interface due to high electron temperature by the reaction between electrons and \text{H}_2\text{O} (see Reaction (5)) [18–21].

\[
\text{Cathode: } \text{H}_2\text{O} + e^- \sim \rightarrow e_{\text{aq}}, \text{H} \times , \text{OH} \times , \cdot \text{O}, \cdot \text{O}^{2-}, \text{H}_2, \text{H}_2\text{O}_2, \text{OH}^- \tag{5}
\]
Meanwhile, soluble $[\text{TaF}_7]^{2-}$ complexes can react with $\text{H}_2\text{O}$ to form $\text{Ta(OH)}_5$ (see Reaction (6))

$$[\text{TaF}_7]^{2-} + 5\text{H}_2\text{O} \rightarrow \text{Ta(OH)}_5 + 7\text{F}^- + 5\text{H}^+ \quad (6)$$

$\text{Ta(OH)}_5$ is further converted to $\text{Ta}_2\text{O}_5$ (see Reaction (7)).

$$2\text{Ta(OH)}_5 \rightarrow \text{Ta}_2\text{O}_5 + 5\text{H}_2\text{O} \quad (7)$$

The overall reactions produced of $\text{Ta}_2\text{O}_5$ can be described as Reaction (8):

$$2\text{Ta} + 5\text{H}_2\text{O} \rightarrow \text{Ta}_2\text{O}_5 + \text{H}_2 \quad (8)$$

After adding CTAB, it can form $\text{CTA}^+$ and attach to the surface of the synthetic product to reduce agglomeration [35].
4. Conclusions

Ta$_2$O$_5$ NPs was successfully prepared in 100 ml 6 g L$^{-1}$ Na$_2$SO$_4$ solution containing 5 ml HF and 0.075 g CTAB by using CGDE technique, in which Pt needle point and tantalum foil was acted as cathode and anode, respectively. The surfactant CTAB effected the morphology and particle size of Ta$_2$O$_5$ NPs. After adding CTAB, the particle size of the formed Ta$_2$O$_5$ is decreased significantly. Prepared Ta$_2$O$_5$ NPs can be used as a photocatalyst. Compared with other technique, CGDE will offer many benefits for fabricating various nanoparticles due to simple experimental setup, no gas supply, no secondary pollution, and mild reaction conditions.

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Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Declaration of interests

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.

Conflicts of interest

The authors declared that they have no conflicts of interest to this work.

Data availability statement

The data that support the findings of this study are openly available at the following URL/DOI:

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