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Wet-Chemical Synthesis of InTaO_4 for Photocatalytic Decomposition of Organic Contaminants in Air and Water with UV–vis Light

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ABSTRACT: A wet-chemical technique was used to synthesize InTaO_4 , AgNbO_3 , InNbO_4 , and various metal doped InTaO_4 catalysts. Their physicochemical properties were characterized by UV–vis diffusion reflectance spectroscopy, X-ray diffraction (XRD), scanning electron microscopy (SEM), and energy-dispersive X-ray spectroscopy (EDS). Photocatalytic activities of these materials were evaluated in liquid-phase decomposition of methylene blue and phenol as well as gas-phase degradation of toluene under UV–vis light. InTaO_4 exhibited higher activity than AgNbO_3 and InNbO_4 in photodecomposition of organic compounds in gas and liquid phases. Ni-doping could enhance the photocatalytic activities. Commercial TiO_2 –P25 had better activity, but Ni– InTaO_4 showed more stable performance in catalytic oxidation of toluene.

1. INTRODUCTION

Recent investigations show that InTaO_4 is a good photocatalyst for splitting water to H_2 under UV light irradiation, and has better performance than commercial TiO_2 –P25.^{1,2} Visible light photocatalysis for water splitting and/or organic contaminant decomposition using various tantalates and niobates have also been reported.^{3–5} However, InTaO_4 and InNbO_4 only showed good performance in H_2 production via visible light irradiation but less activity in O_2 production. Three important factors, (i) position of conduction/valence bands, (ii) moment of photo-generated carriers, and (iii) M–O–M angle, influence the photocatalytic activities of the materials. The more negative or positive the conduction/valence band is, the higher the redox potential is and consequently the higher photocatalytic activity presents. The more closure M–O–M is to 180° , the higher the carrier mobility is and consequently the better the activity of the material is. Zou et al.¹ demonstrated that InTaO_4 and InNbO_4 are consisted of two octahedrons, InO_6 and $\text{TaO}_6/\text{NbO}_6$, where conduction bands are made of empty 5d and 4d orbitals of Ta and Nb, respectively, and are positioned at different energy levels. However, valence bands which are made of O_{2p} orbitals of InO_6 in both the materials have the same potential level. Thus due to the lower conduction band position, InNbO_4 band gap (2.5 eV) was smaller than that of InTaO_4 (2.6 eV), but hydrogen was evolved at lower rate on InNbO_4 than that evolved on InTaO_4 when irradiated with visible light in the presence of cocatalyst NiO. Higher photocatalytic activity of InTaO_4 than InNbO_4 was mainly attributed to the position of the conduction band, M–O–M angle value, and its subsequent effect on electron moment. It was found that Ta–O–Ta is more closure to 180° , which alleviates moment of electron in conduction band and thus resulting in increased photocatalytic activity.

Many attempts such as doping with cations (Mn, Fe, Co, Cu, and Ni,⁶ Sc, V, Zn, Ti, and Cr,⁷ and anions (N)^{8,9}) have been made to enhance the photocatalytic efficiency of InTaO_4 . A significant improvement in photocatalytic production of H_2/O_2 was achieved on 1% nickel-coated InTaO_4 , which was attributed to the alleviated electron moment caused by the change in M–O–M angle,¹⁰ a

narrowed band gap by doping,² and electron trapping of the NiO layer.⁷

Previously, all the above materials were prepared by solid state reaction. Some other techniques were also employed for synthesis of InTaO_4 , such as a reactive pulsed LASER ablation method⁸ and sol–gel techniques.¹¹ However, few investigations have been reported in wet-chemical synthesis of InTaO_4 or AgNbO_3 , which is usually facile and environmentally friendly, and their application for decomposition of organic contaminants.⁹ Hiroshi et al.⁹ found that InTaO_4 doped with vanadium could decompose gaseous isopropyl alcohol (IPA) into acetone and CO_2 at ultraviolet radiation but showed little activity under visible light irradiation. Chiou et al.¹¹ reported that InTaO_4 prepared by sol–gel techniques demonstrated higher photocatalytic activities for H_2 production because of a smaller band gap (2.62 eV), particle size (0.05–0.1 μm), and more uniform crystalline phase compared to the one prepared by solid state reactions.

Photocatalytic decomposition of contaminants in gas and water is important in environmental remediation.^{12–14} Many types of photocatalyst have been tested; however, few investigations have been reported using InTaO_4 systems. In this paper, we report synthesis of pristine InTaO_4 , AgNbO_3 , InNbO_4 , and doped InTaO_4 by a simple wet-chemical technique. The synthesized materials were tested for photocatalytic decomposition of methylene blue in aqueous solution and toluene in gaseous phase under UV–vis light.

2. EXPERIMENTAL SECTION

2.1. Synthesis of Various Catalysts. Polycrystalline pristine InTaO_4 , AgNbO_3 , InNbO_4 , and doped InTaO_4 photocatalysts

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were prepared by a solution method. All of chemicals were obtained from Sigma-Aldrich at 99.99% purity and were used as received. In a typical synthesis, 10 mmol of indium(III) nitrate hydrate ($\text{In}(\text{NO}_3)_3 \cdot x\text{H}_2\text{O}$) and 8 mmol of tantalum(V) chloride (TaCl_5) were dissolved separately in a 50-mL ethanol solution with continuously stirring and heating at 70 °C for 30 min to make solutions A and B, respectively. Dopant solution C was also prepared similarly by dissolving 0.01 mmol of dopant salt (cobalt(II) nitrate hexahydrate, copper(II) nitrate trihydrate, and nickel(II) nitrate hexahydrate, etc.) in 30 mL of ethanol. Both solutions A and B were then mixed and 0.5–1.0 mL of concentrated nitric acid was added to the mixture. The mixture was kept heating at 80 °C for refluxing of 6–8 h. A clear solution without any precipitation was then aged for 12–24 h at room temperature and then kept in an oven at 50–60 °C until dried completely. After being ground, the samples were calcined at 1150 °C for 24–30 h. AgNbO_3 and InNbO_4 were also prepared by the similar technique where niobium(V) chloride (NbCl_5) and silver nitrate (AgNO_3) were used as Nb and Ag precursors, respectively.

2.2. Characterization of Catalysts. The crystal structures of samples were analyzed by X-ray diffractometer (Bruker D8 Advance equipped with a Lynx eye detector, Bruker-AXS, Karlsruhe, Germany) operated at 40 kV and 30 mA. The scanning rate was 0.2 s/step with 2θ (10°–90°) and step size of 0.02°. Cu K α ($\lambda = 1.54178$ Å) was used as a X-ray source with divergent slit of 0.300 and 2.5° primary and secondary soler slits. The optical absorption of samples was determined by UV–vis absorbance spectroscopy using the diffuse reflectance method (JASCO V-670 Spectrometer). Morphology and chemical compositions of the materials were examined by scanning electron microscopy (ZEISS NEON 40EsB) equipped with an energy dispersive spectrometer (SEM-EDS). Surface chemistry of catalyst samples was examined by X-ray photoelectron spectroscopy (XPS) and FTIR analyses. FTIR analysis was performed on a Perkin-Elmer model FTIR-100 with a MIR detector. XPS data were acquired using a Kratos Axis ULTRA X-ray photoelectron spectrometer incorporating a 165-mm hemispherical electron energy analyzer. The incident radiation was monochromatic Al K α X-rays (1486.6 eV) at 225 W (15 kV, 15 mA). Survey (wide) scans were taken at analyzer pass energy of 160 eV and multiplex (narrow) high-resolution scans at 40 eV. Survey scans were carried out over 1200.0 eV binding energy range with 1.0 eV steps and a dwell time of 100 ms. Narrow high-resolution scans were run with 0.05 eV steps and 250 ms dwell time.

2.3. Photocatalytic Evaluation. Photocatalytic activities of prepared samples were evaluated in decomposition of methylene blue in water and toluene in air with UV–vis radiations. The light source was a 500 W mercury–xenon lamp (UXM-502MD, Ushio) with average intensities of 5 mW/cm² at 220–280 nm, 38.5 mW/cm² at 280–400 nm, and 200 mW/cm² at >400 nm. In a typical run of methylene blue decomposition in water, 10 ppm methylene blue solution with 100 mg of catalyst particles were continuously stirred in a Pyrex glass reactor which was irradiated with the UV–vis light without any filter. Temperature of the reactor was maintained by passing cooling water through the reactor continuously. Concentration of methylene blue was determined by taking 1-mL aliquots from the reactor at regular intervals. The aliquots were evaluated by measuring the absorbance at $\lambda = 664$ nm on a visible spectrophotometer (Spectronic Instruments model 4001/4).

Toluene decomposition in gas was evaluated in a flow reactor connected with a gas chromatograph (Shimadzu GC-17A) fitted

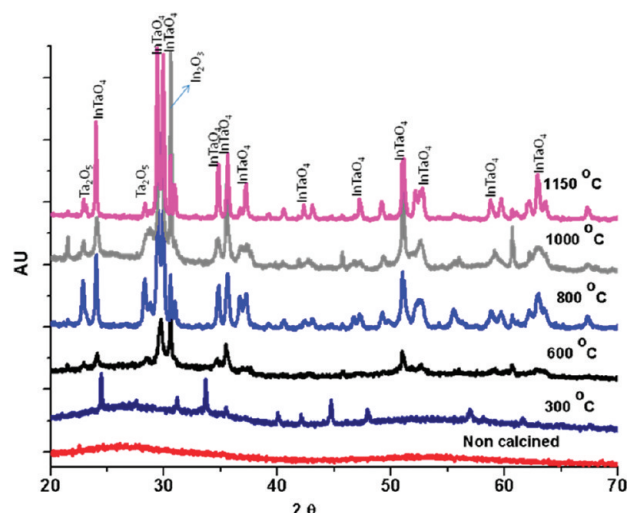


Figure 1. XRD analysis of InTaO_4 synthesized at various temperatures.

with a GS-GSPRO column of 60 m long and 0.32 mm inner diameter. Oven temperature of the GC was 220 °C and FID detector temperature was kept at 250 °C. In a typical experiment, 200–300 mg of a photocatalyst was dispersed in 10 mL of deionized water in a Petri dish of 450-mm diameter and was dried in an oven overnight. The Petri dish was kept inside an airtight reactor fitted with a removable quartz sheet in order to allow UV–vis radiation in. A flow of toluene at 100 ppm was continuously passed into the reactor (1.08 L) at a flow rate of 60 mL/min. The temperature of the reactor was controlled at room temperature by continuously passing cooling water through the reactor and also by blowing the reactor with fresh air from outside.

3. RESULTS AND DISCUSSION

3.1. Characterization of Catalysts. XRD analysis of the prepared InTaO_4 calcined at varying temperatures (Figure 1) indicates that the sample without calcination presented an amorphous phase. However, crystallization could be observed at calcination of 300 °C and complete crystalline structure of InTaO_4 was obtained at elevated temperature of 1150 °C. At lower temperatures (300–1000 °C), In_2O_3 and Ta_2O_5 at $2\theta = 30.58^\circ$, 21.47° and $2\theta = 22.820^\circ$, 28.267° , respectively, were observed. However, indium tantalum oxide (InTaO_4) was found to be the major product, which has a monoclinic structure with $a = 5.150$, $b = 5.770$, $c = 4.821$, $\alpha = 90.0^\circ$, $\beta = 91.35^\circ$, $\gamma = 90.0^\circ$, primitive $P2_1/a$ (13). This InTaO_4 has a crystal structure similar to those prepared by solid state reaction, but in different space group of $P2_1/c$.¹

To obtain a pure product of InTaO_4 , molar ratios of the precursor salts (indium nitrate and tantalum chloride) were varied in preparation. It was found that the sample synthesized at the molar ratio of 1:0.5 (In:Ta) had much lower In_2O_3 and Ta_2O_5 impurities (Figure 2). Therefore, the molar ratio of In:Ta was kept at 1:0.5 for doped InTaO_4 synthesis. The metals of doping were Bi, Cu, Ag, V, and Ni at 1–2%. XRD patterns (Figure 3) showed no significant structure change occurring on those doped samples. This revealed that 1–2% metal doping into pristine InTaO_4 may only bring about some physical changes but no changes in crystalline structure or chemical phase.² Zou et al.¹⁰ also reported no modification in crystalline parameters

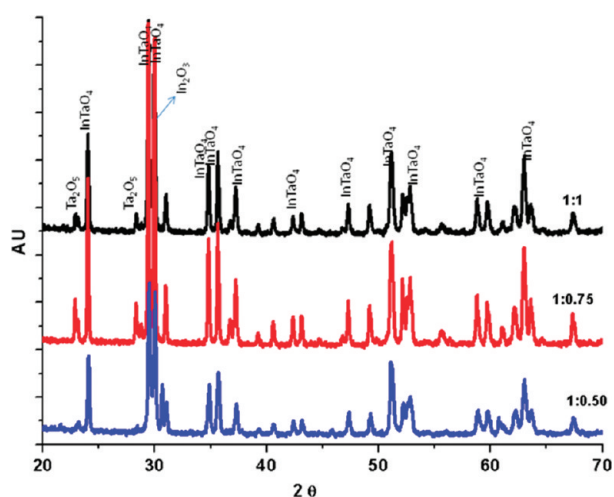


Figure 2. XRD analysis of InTaO_4 synthesized at different ratios of In and Ta.

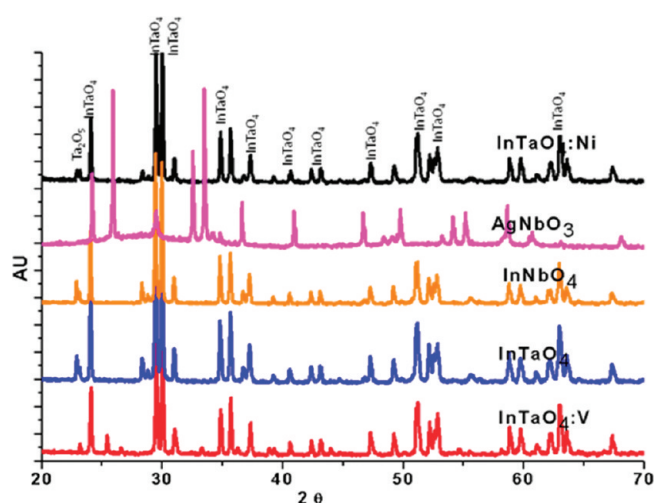


Figure 3. XRD analysis of AgNbO_3 , InNbO_4 , and Ni-, V-doped InTaO_4 catalysts.

by doping InTaO_4 with Ag, Ni, Cu, and Co. However, a slight decrease in crystal parameters ($a = 4.833$, $b = 5.778$, $c = 5.157$, $\alpha = 90.000^\circ$, $\beta = 91.380^\circ$, $\gamma = 90.000^\circ$) was found in bismuth doped sample ($\text{InTaO}_4:\text{Bi}$). The steadiness of crystal structure and crystal parameters of InTaO_4 by doping with Ag^{2+} , Ni^{2+} , Cu^{2+} , and Co^{2+} may be due to either their interstitial position or surface cover of the oxides on InTaO_4 . Bismuth with different ionic radius may cause the structural variation. Thus it is concluded that substitutional dopant could cause variations in crystal parameters if the dopant ionic size is different from that of the substituted ion while the interstitial doping of foreign elements may only change the physical properties such as an increase in defect sites and band gap modification.

XRD analyses of AgNbO_3 and InNbO_4 (Figure 3) show that silver niobium oxide (AgNbO_3) has an orthorhombic structure with $a = 5.602$, $b = 7.824$, $c = 5.540$, $\alpha = 90.000$, $\beta = 90.000$, $\gamma = 90.0$, primitive $Pnma$ (62). Indium niobium oxide has a monoclinic structure with $a = 4.843$, $b = 5.773$, $c = 5.147$, $\alpha = 90.0^\circ$, $\beta = 91.230^\circ$, and $\gamma = 90.0^\circ$, Primitive $P2/c$. Chemical compositions of doped and nondoped samples determined by SEM-EDS

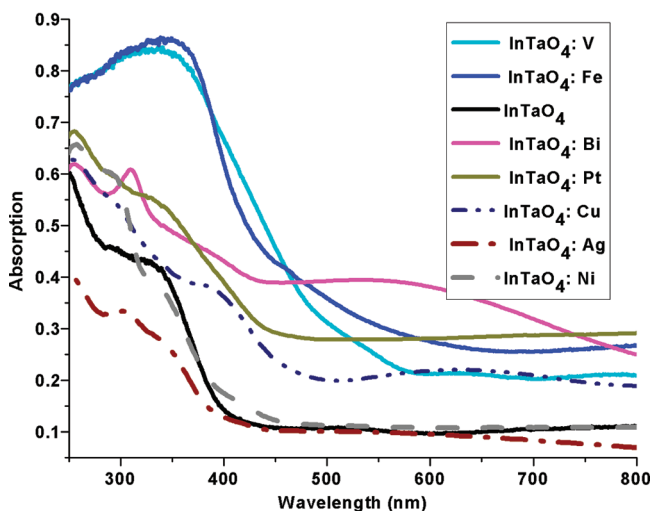
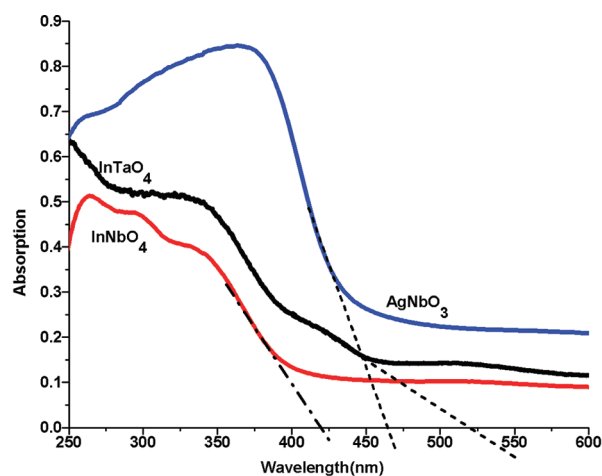
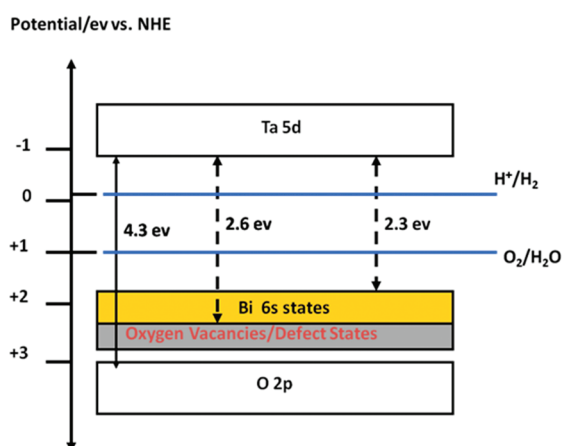


Figure 4. UV-vis diffusive reflectance spectra of AgNbO_3 , InNbO_4 , InTaO_4 , and metal-doped InTaO_4 .

show no dopant atoms, which is probably due to the low contents of dopants.

UV-vis diffuse reflectance spectra of nondoped and doped InTaO_4 (Figure 4) indicate that doping of metal ions to InTaO_4 brought out a significant change to the absorption profile by extending strong absorption to visible range. Figure 4 shows that there is a shoulder peak between 284 and 340 nm in the optical absorption of pristine InTaO_4 , which reveals that this material may have two band gaps. The absorption edge at around 284 nm is due to the electronic transition between conduction band and valence band, and another absorption edge at around 450 nm is because of the defect sites mostly being composed of oxygen vacancies. Hiroshi et al.⁹ have also observed the similar effect of two onsets in the absorption spectrum of nondoped InTaO_4 . The band gap of the nondoped InTaO_4 estimated with a formula¹⁵ ($E_g = 1239/\lambda$ (wavelength nm)) was found to be 2.4 eV,¹⁶ however, Zou et al.¹⁰ reported a band gap of 2.6 eV for a nondoped InTaO_4 . Although there are different views¹⁷ regarding the band gap due to the two onsets in the absorption spectrum of InTaO_4 , however, based on its first absorption edge at lower wavelength, this material has a larger and indirect band gap of 4.3 eV.^{8,9}

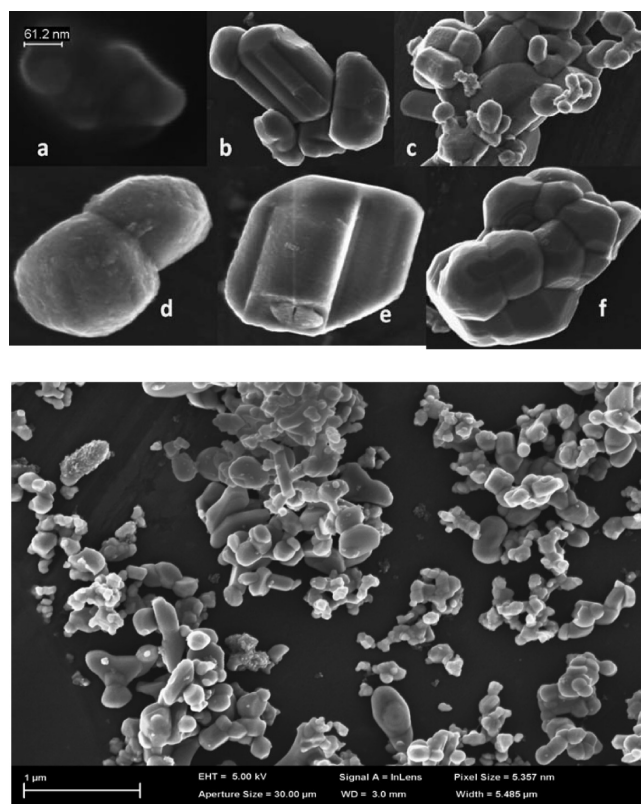
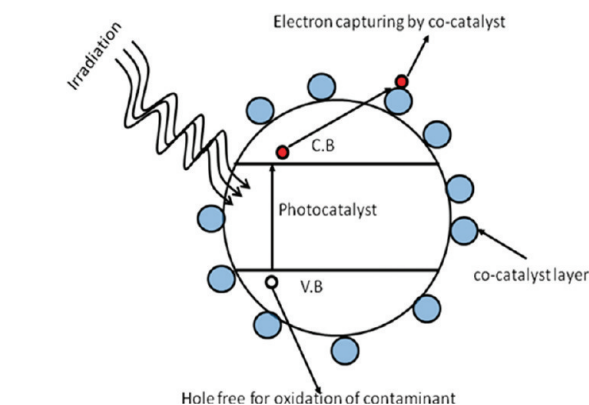
Figure 4 also shows the optical absorption of AgNbO_3 and InNbO_4 . InTaO_4 and AgNbO_3 have an onset at 400 and 450 nm,

Scheme 1. Band Gap Modification of InTaO₄ Doped with Various Ions (Bi, Cu, Ni)

respectively, while InNbO₄ has an absorption onset at lower wavelength of 380 nm. Unlike InTaO₄, AgNbO₃ has very sharp absorption, whereas InNbO₄ has a flat shoulder absorption between 280 and 380 nm. Band gaps of InNbO₄ and AgNbO₃ were found to be 2.9 eV and 2.8 eV, respectively. Ye et al.¹⁸ reported a band gap of 2.5 eV for InNbO₄ and Li et al.¹⁹ reported a similar band gap of 2.8 eV for AgNbO₃.

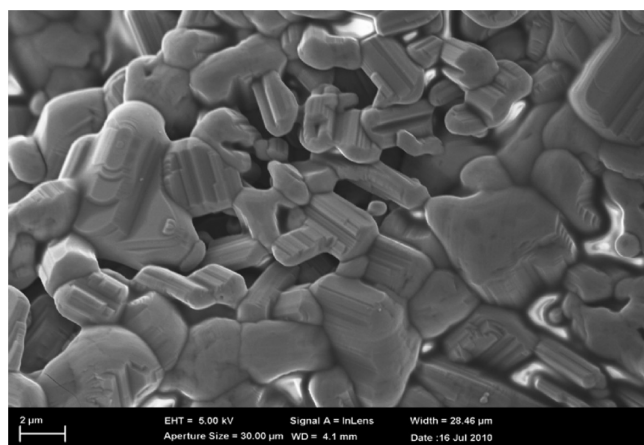
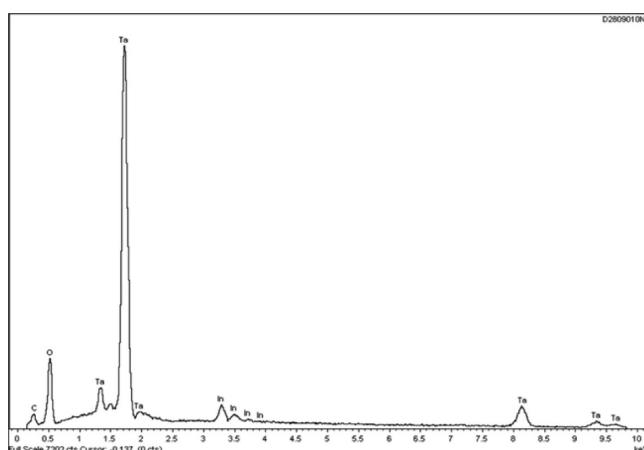
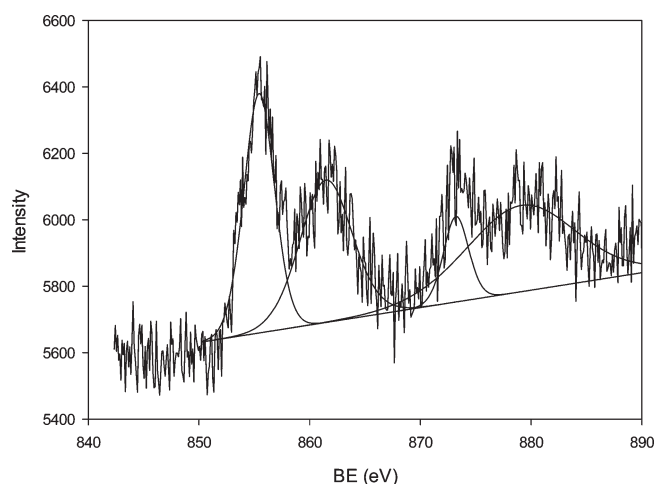
Doping Pt, Cu, Ag, Ni, Co, Fe, and Bi ions in InTaO₄ modified the optical absorption of InTaO₄ and shifted the absorption to the longer wavelength (Figure 4). This red-shift in doped InTaO₄ was also reported by other investigators.^{7,9} Dopants like Fe, V, Bi, and Cu enhanced the optical absorption significantly and produced a shift of absorption edge to the longer wavelength, whereas Ag doping had a worse effect on the optical absorption by reducing the overall optical absorption. Dopant ions like Bi, Fe, V, and Cu would substitute In ions in InO₆ octahedron and brought out changes to the crystal structure in lattice parameters reducing the band gap. The changes in lattice parameters also affected the volume of InO₆ octahedron producing defect sites and enhancing optical absorption.

As illustrated in Scheme 1, dopants like Bi, Fe, V, and Cu introduced a narrow band just above the valence band of InTaO₄, and pushed the valence band toward more negative position, thus reducing the effective band gap of InTaO₄. Therefore, owing to the above two reasons, i.e., increase in defect sites, and formation of dopant band above the valence band, doped InTaO₄ absorbed more light in the visible range. Contrary to Bi, Fe, V, and Cu ions doping, Ni doping into InTaO₄ brought about very limited modification to the optical absorption while it induced a moderate increase in UV light absorption (Figure 4). This effect of different absorption characteristics at longer or shorter wavelengths suggests that different dopant ions may have different positions (interstitial, substitution) in the crystal thus producing defect states at various energy levels. We suggest that Ni ion doping induces both deep and shallow defect states, which promote the absorption mostly in shorter wavelength and minor changes at longer wavelengths. It must be noted that various reports^{1,2} on Ni-doped InTaO₄ have suggested internal transitions in partially filled Ni 3d levels, which boosted absorption both in the longer and shorter wavelengths. However, XPS analysis (discussion later) confirmed the presence of Ni²⁺ 2p state, which meant that NiO is formed on the surface of InTaO₄.

Scheme 2. Diagram of Ni-doped InTaO₄ in PhotocatalysisFigure 5. SEM images of InTaO₄ (a–d) and Ni-doped InTaO₄ (e–g).

The presence of NiO on the surface acts as cocatalyst and traps the photogenerated electrons, thus reducing the electron–hole pair recombination. This electron trapping (Scheme 2) of the cocatalyst (NiO) can not produce a significant modification to the band gap but assist in suppression of electron–hole recombination process.

SEM images (Figure 5a–d) of nondoped InTaO₄ show the presence of varying geometrical shapes and particle sizes, i.e., elongated, spherical, and plate-like structures. These images suggested the growth and agglomeration of InTaO₄. SEM images of Ni doped InTaO₄ (Figure 5e–g) show similar particles morphology and particle size as that of nondoped InTaO₄, suggesting that doping did not change the morphology of InTaO₄. SEM image (Figure 6) of InNbO₄ shows that this

Figure 6. SEM image of InNbO₄.Figure 7. EDS analysis of InTaO₄:Ni.Figure 8. XPS Ni 2p spectrum of InTaO₄:Ni.

material crystallizes in plate-like structure and has a comparatively larger microsize to that of InTaO₄.

Although EDS (Figure 7) and XRD investigations could not detect Ni and/or NiO, however, XPS of Ni doped InTaO₄ confirmed the existence of NiO (Figure 8). Ni 2p_{3/2} state of NiO

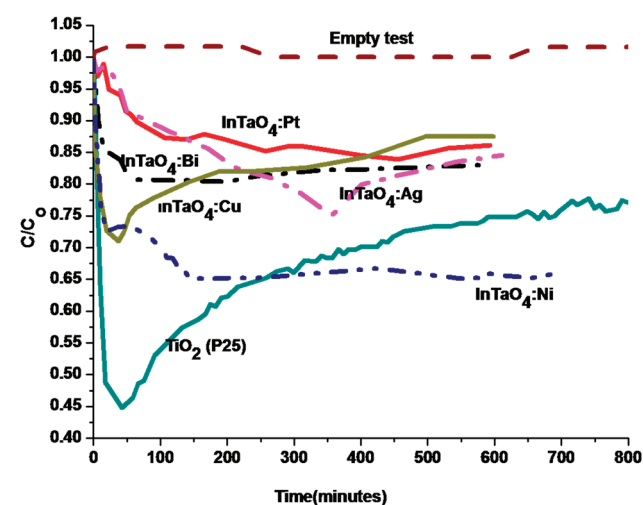
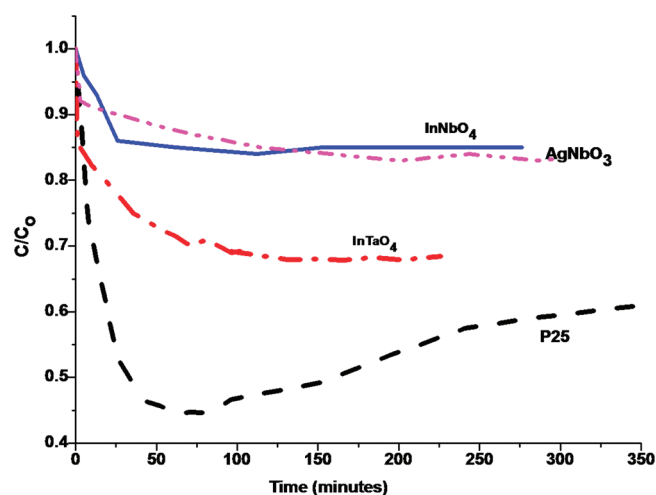


Figure 9. Photocatalytic decomposition of toluene on various photocatalysts with UV–vis irradiation.

was observed at binding energy (BE) of 855.5 eV¹¹ where its corresponding satellite peak is close to 861.0 eV. The satellite is about 6.0 eV higher than the main BE peak. Another peak observed at 873.6 eV can be assigned to Ni 2p_{1/2} states of NiO.²⁰ Chiou et al.¹¹ also observed the presence of metallic Ni at binding energy of 852.9 eV, of the sample reduced in H₂ environment. However, we could not observe existence of metallic Ni²¹ in the Ni-doped sample of InTaO₄.

Therefore, XRD and SEM-EDS analyses show that doping at 1–2% in InTaO₄ would not change its bulk and structural composition, but doping could modify the photoresponsive properties of InTaO₄ inducing a change in photocatalytic behavior.

3.2. Photocatalytic Activities. Photocatalytic activities of InTaO₄, InNbO₄, AgNbO₃, and doped InTaO₄ catalysts in decomposition of toluene gas were determined in a closed continuous-flow reactor irradiated with UV–vis light (Figure 9). TiO₂–P25 presented a fast decomposition of toluene at 50% in the first hour under UV–vis irradiation but strong deactivation afterward. After 800 min, toluene decomposition efficiency was reduced to 20%. InTaO₄ exhibited a gradually increased efficiency of toluene degradation and reached 30% at the first hour and maintained the stable activity after. InNbO₄ and AgNbO₃²²

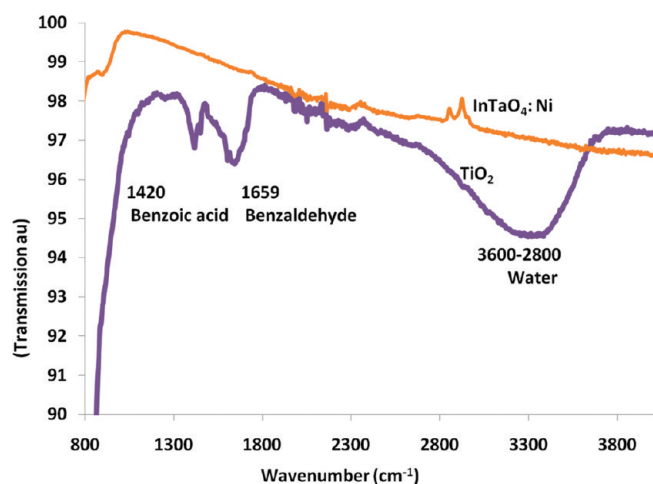


Figure 10. FTIR spectra of used TiO_2 and $\text{InTaO}_4\text{:Ni}$.

showed similar activities with decomposition of toluene at 10–15% within the first 2 h. The deactivation of $\text{TiO}_2\text{--P25}$ was due to a strong affinity to water and reaction intermediates which covered the surface reactive sites for photocatalytic decomposition of toluene. It was also found that $\text{TiO}_2\text{--P25}$ changed the color from white to black after reaction; however InTaO_4 showed a pale yellow indicating much less intermediate adsorption on the surface.

Figure 10 shows FTIR analysis of $\text{TiO}_2\text{--P25}$ and InTaO_4 after use for photocatalytic oxidation of toluene under UV–vis light irradiation. Similar to other research work,^{23–25} strong water and intermediate adsorption was observed on TiO_2 surface. A broad and intense peak at 2800–3600 cm^{-1} centered at 3350 cm^{-1} corresponds to the stretching mode of water molecules²³ whereas the band at 1590–1700 cm^{-1} centered at 1659 cm^{-1} corresponds to aromatic molecules of benzaldehyde. Another aromatic intermediate observed with a single band at 1420 cm^{-1} was found to be benzoic acid.^{23,26} It can be deduced from the FTIR analysis that strong adsorption of water molecules and aromatic intermediates covered the active sites of TiO_2 and caused the irreversible deactivation.^{26,27} However, such strong adsorption of water molecules and intermediates did not occur on doped and nondoped InTaO_4 surface and therefore they would work longer than the commercial TiO_2 .

Photocatalytic activity of Ni, Ag, Cu, Pt, and Bi doped InTaO_4 samples for toluene decomposition were also investigated (Figure 9). The order of photocatalytic activity in the first 2 h was $\text{TiO}_2\text{--P25} > \text{InTaO}_4\text{:Ni} > \text{InTaO}_4\text{:Cu} > \text{InTaO}_4\text{:Bi} > \text{InTaO}_4\text{:Ag} > \text{InTaO}_4\text{:Pt}$. As shown in Figure 9, the tests in a longer period indicated that Ni-doped InTaO_4 was far better than the commercial $\text{TiO}_2\text{--P25}$. The activity of $\text{InTaO}_4\text{:Ni}$ remained unchanged in 14 h. In the second run after regeneration, $\text{TiO}_2\text{--P25}$ lost 90% of its efficiency, while $\text{InTaO}_4\text{:Ni}$ maintained its photocatalytic activity. XRD analysis of $\text{InTaO}_4\text{:Ni}$ after reaction for 14 h (Figure 11) clearly indicates that the material was quite stable and did not change its structure. This strong stability of the material suggests that InTaO_4 doped with Ni can be a better alternative to the commercial photocatalytic material $\text{TiO}_2\text{--P25}$. Zou et al.²² found that $\text{TiO}_2\text{--P25}$ had better performance than Ni-doped InTaO_4 in H_2 production from water splitting under UV light irradiation. However, InTaO_4 doped with Mn, Co, and Ni showed better performance than $\text{TiO}_2\text{--P25}$ for H_2 production under visible light irradiation.^{2,10} Chiou et al.¹¹ have demonstrated

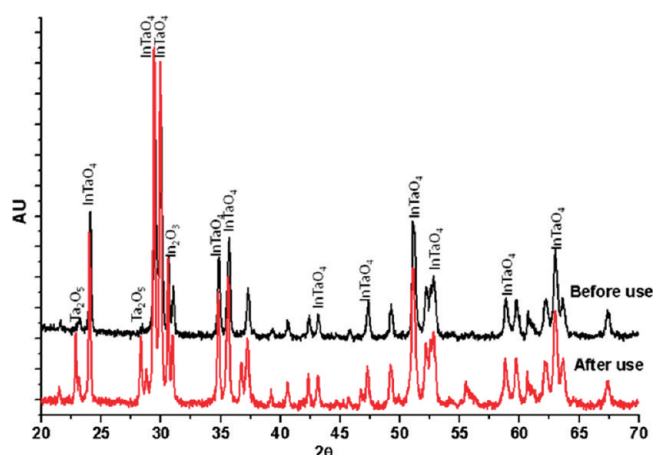


Figure 11. XRD analysis of $\text{InTaO}_4\text{:Ni}$ before and after use for photocatalytic reaction.

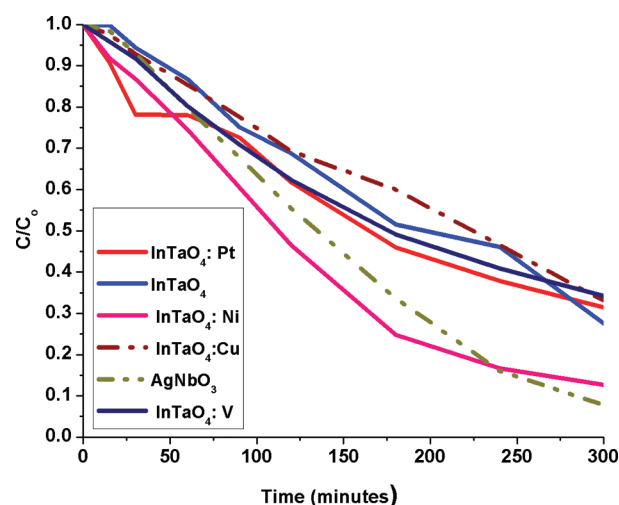


Figure 12. Decomposition of methylene blue on various photocatalysts with UV–vis irradiation.

that 3 wt % Ni loading produced both NiO and NiOH on InTaO_4 surface. Upon reduction in H_2 environment metallic Ni shell on surface was created and acted as electron trapping centers, enhancing photocatalytic water splitting. Thus it is deduced that the better performance of $\text{InTaO}_4\text{:Ni}$ is the combined effect of both the band gap contraction and electron capturing by NiO as explained in Scheme 2. Although doping of InTaO_4 with Ag, Cu, Pt, V, and Bi produced red-shifts of the optical absorption but they did not make a cover on the InTaO_4 surface like NiO. These metal dopants reduced the optical band gap, but they may also enhance the electron–hole pair recombination, by creating midgap defect states. The generation of the defect states plays a detrimental role in recombination centers which may reduce the photocatalytic decomposition of toluene. Ni doping modified the band gap but suppressed the recombination via electron trapping on NiO, making it exhibit better performance. Recently, Shu et al.²² have demonstrated a similar effect of electron trapping by NiO loaded on AgNbO_3 for methylene blue decomposition under UV irradiation.

Photocatalytic performance of nondoped and doped InTaO_4 materials in methylene blue decomposition in water with UV–vis light (Figure 12) showed that Ni-doped InTaO_4 degraded methylene blue at a much faster rate than the others.

AgNbO₃ had performance almost comparable to InTaO₄:Ni. InTaO₄:Ni and AgNbO₃ were capable of decomposing about 90% methylene blue within 6 h, while Cu, Pt, V, and nondoped InTaO₄ could degrade about 60% methylene blue. It is noted that doped and nondoped InTaO₄ did not show the same trend in methylene blue decomposition as that in toluene decomposition; however, Ni-doped InTaO₄ has demonstrated the best performance. Another test revealed that Ni-doped InTaO₄ could also decompose about 75% of phenol within 3 h (figure not shown). As explained earlier, the better photocatalytic performance of Ni-doped InTaO₄ among the others are mainly attributed to the dual effects of band gap reduction and electron trapping by NiO.

4. CONCLUSIONS

Tantalum- and niobium-based compounds such as InTaO₄, InNbO₄, and AgNbO₃ can be prepared by a simple wet-chemical technique. Most of these materials possess poly crystalline structures and large size agglomerates. InTaO₄ exhibits better activity than InNbO₄ and AgNbO₃. The physical properties and photocatalytic activities of InTaO₄ are also influenced by doping with metals like Pt, Cu, Ag, V, Bi, and Ni. Bi-doped InTaO₄ will result in more visible light absorption, but Ni-doped InTaO₄ shows stronger UV absorption. InTaO₄:Ni can efficiently decompose all the contaminants of toluene, methylene blue, and phenol in air or water under UV–vis light. The higher photocatalytic activity of InTaO₄:Ni is attributed to the band gap modification and electron trapping center of NiO. InTaO₄:Ni also has better performance in multiple uses and longer lifetime than the commercial TiO₂–P25.

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