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# Deactivation and Regeneration of NaTaO<sub>3</sub> Photocatalyst in Co-operating Dehydrogenation Coupling of Isopropanol and Hydrogenation Coupling of Acetone Reaction System

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# ABSTRACT

Photocatalyst activity is influenced by many factors, such as adsorption of by-products, runoff of surface hydroxyl groups, and carriers. In this study, a simple and efficient photocatalyst regeneration method was developed. Results indicated that NaTaO<sub>3</sub> photocatalyst lost its photo-activity after three cycles of reaction that involves coupling of isopropanol and hydrogenation coupling of acetone reaction system. Runoff of Na on the

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surface was the main reason for the deactivation of NaTaO<sub>3</sub> photocatalyst. After hydrothermal treatment of the deactivated NaTaO<sub>3</sub> with 10 M NaOH at 180 °C for 12 h, its photocatalytic activity was restored to the original level. The photocatalytic activity remained stable even after 10 cycles **INTRODUCTION** 

# Given that photocatalysis is environmentally friendly and can effectively solve energy and environmental problems brought by traditional chemical engineering (1-2), it has been extensively adopted in various applications, such as photocatalytic hydrogen production (3), photocatalytic organic synthesis (4), and pollutant disposal (5). Numerous research and reports on the development of new photocatalytic materials, modification of photocatalysts, and design of high-efficiency photocatalytic reaction have been conducted.

Catalytic hydrogenation and dehydrogenation reactions play a key role in the chemical industry, particularly in the synthesis of various pharmaceuticals and fine chemicals, in addition to bulk chemicals (6). This knowledge compelled us to combine hydrogenation reactions and dehydrogenation reactions in one catalytic process to achieve excellent thermal balances, efficient hydrogen use, and consequently high overall efficiency (7). Our group previously reported co-operating the dehydrogenation coupling of isopropanol and hydrogenation coupling of acetone reaction system. The photogenerated holes and electrons were efficiently used, which dramatically promoted the total efficiency and cleanliness of photocatalytic systems and photocatalytic organic syntheses (8). However, the instability of photocatalysts has been the key restriction in its industrialization, the same with other photocatalytic reaction systems (9). Therefore, developing photocatalysts with favorable stability is essential for the development of photocatalysis technology. Photo-activity and the stability of photocatalysts are related to multiple factors, mainly including its preparation, pretreatment technology, reaction conditions, and operation (10-11). For different structures and reaction systems, their deactivation mechanisms are different. In a photocatalytic reaction system, studying the deactivation mechanism and maintaining stability of photocatalyst constitute a research topic of great practical significance (12,13). Currently, methods such as photo-irradiation (14), heat treatment (15), and ultrasonic wave regeneration (16) are generally used. However, different reaction systems have different photocatalyst deactivation

mechanisms. Thus, the deactivation mechanism should be considered to obtain a photocatalyst with high catalytic activity and good stability. Based on the above research, a simple and effective photocatalytic regeneration method was developed by studying the photocatalytic deactivation mechanism of coupling the reaction systems of isopropanol dehydrogenation and acetone hydrogenation through NaTaO<sub>3</sub> photocatalysis.

# MATERIALS AND METHODS

*Preparation of NaTaO<sub>3</sub> photocatalysts.* The synthesis of NaTaO<sub>3</sub> photocatalysts was similar to the method reported before (17). Firstly, 4.42 g of  $Ta_2O_5$  was mixed with 30 ml of NaOH with desired concentration and stirred at room temperature for 2 h. Then the mixture was filled into a Teflon reactor (50 mL) and was heated up to  $180^{\circ}C$  for a 24 h reaction. The NaTaO<sub>3</sub> photocatalyst was soaked in 100 mL of 1 M HCl solution for 12 h to replace the Na<sup>+</sup> on the surface of NaTaO<sub>3</sub>.

*Regeneration of deactivated NaTaO*<sub>3</sub> *photocatalysts.* Ultrasonic cleaning to remove the byproducts on the surface of the NaTaO<sub>3</sub>, then hydro-thermal treated with 10 M NaOH at 180°C for 12 h. After the reaction, the NaTaO<sub>3</sub> products were collected by centrifugation and washed with deionized water and ethanol to remove the redundant NaOH, and finally dried at 80°C for 12 h.

*Characterization of NaTaO*<sub>3</sub> *photocatalysts.* The surface functional groups of the NaTaO<sub>3</sub> photocatalysts were observed by Fourier Transform infrared spectroscopy (FTIR, Nicolet 380). The crystal structures were characterized by an X-ray diffraction (XRD) (Bruker D8 Advance) with a Cu K $\alpha$  ( $\lambda$ =0.15406 nm), operated at 40 kV. The element contents and their binding state on NaTaO<sub>3</sub> surface were measured by X-ray photoelectron spectroscopy (XPS, Kratos Axis Ultra, Al KR source), the C 1s reference of 284.5 eV as the calibration of binding energy.

*Photocatalysis tests.* All photocatalytic tests were operated in a 250 mL inner-irradiation-type Pyrex reactor under 300 W UV light and atmosphere pressure (Figure S1). The photocatalytic experiments were performed in 200 mL of aqueous solution, using 1.0 g of NaTaO<sub>3</sub> photocatalyst. Liquid samples were analyzed by a gas chromatograph (Haixin; GC-950) equipped with a flame ionization detector (FID).

#### **RESULTS AND DISCUSSION**

#### Stability evaluation of NaTaO<sub>3</sub> photocatalyst in coupling reaction system

Our previous results indicated that NaTaO<sub>3</sub> photocatalyst presented excellent photocatalytic activity in co-operating isopropanol dehydrogenation coupling and acetone hydrogenation coupling reaction systems (acetone and isopropyl alcohol conversion rates were 61.25% and 53.12%, respectively, and pinacol selectivity was greater than 90%) (8). After one cycle of the reaction, NaTaO<sub>3</sub> was directly used for photocatalytic coupling reaction without any treatment. Its photocatalytic activity evidently dropped, isopropyl alcohol conversion rate was only 19.18%, and pinacol selectivity was about 82.67%. When the reaction entered its third cycle, isopropyl alcohol conversion rate was only 7.60% and pinacol selectivity was reduced to 77.31%. Results are presented in Table 1.

#### Analysis of deactivated NaTaO<sub>3</sub> photocatalyst morphology and structure

Figure 1 shows the XRD patterns of NaTaO<sub>3</sub> photocatalyst before and after reaction. It showed that deactivated photocatalyst still had characteristic peak of NaTaO<sub>3</sub> with complete crystal structure (17). This observation indicated that the crystal structure was not damaged.

FTIR can effectively analyze surface conditions of the photocatalyst and species adsorbed on its surface. As shown in Figure 2, after the reaction of NaTaO<sub>3</sub> photocatalyst for 12 h, its infrared spectrogram had apparent characteristic absorption peaks of C=C, C=O, - CH<sub>3</sub>, and -CH and C-OH when compared with NaTaO<sub>3</sub> photocatalyst before the reaction. This result indicated that after the photocatalytic reaction, the by-products or intermediate products might have been adsorbed or accumulated on the NaTaO<sub>3</sub> surface.

UV-vis spectra were obtained to effectively analyze the light absorption capacity of the photocatalyst. As shown in Figure 3, the absorption capacity of NaTaO<sub>3</sub> photocatalyst was basically unchanged even after 12 h of reaction.

Element contents and their binding state on the NaTaO<sub>3</sub> surface were analyzed by X-ray photoelectron spectroscopy (XPS). XPS data showed that the binding energies of Ta 4f (Figure 4b) and Na 1s (Figure 4d) on NaTaO<sub>3</sub> surface had obvious displacement after the reaction (Figure 4). Through the hydrothermal treatment of 10 M NaOH with deactivated NaTaO<sub>3</sub> photocatalyst, binding energies of the photocatalyst's Ta 4f (Figure 4c) and Na 1s This article is protected by copyright. All rights reserved.

(Figure 4f) returned to their original positions after the treatment, and the photocatalytic activity of NaTaO<sub>3</sub> was recovered. Na, O, and C atom contents on the catalyst's surface were analyzed, and results indicated that the Na content on NaTaO<sub>3</sub> photocatalyst's surface decreased from 21.41% to 17.58% after coupling reaction. On the other hand, through hydrothermal treatment with 10 M NaOH at 180°C for 12 h, the Na content approximately returned to its original value (Table 2).

#### NaTaO<sub>3</sub> photocatalyst deactivation mechanism in coupling reaction system

The intermediate products, by-products, and surface-active species were detected to analysis the relationship between the photocatalytic activity and intrinsic of NaTaO<sub>3</sub>. Then, the main reason for the deactivation of the NaTaO<sub>3</sub> was analyzed. Figure 1 shows that the crystal structure of the catalyst before and after the reaction did not change, which meant that NaTaO<sub>3</sub> was not damaged. Moreover, the UV-vis spectra were basically unchanged. These results showed that NaTaO<sub>3</sub> deactivation might be caused by the adsorbed impurities occupation of active sites or the loss of surface active constituents.

Studies showed that one of the main reasons for a photocatalyst's deactivation was that adsorption and accumulation of intermediate products or by-products on the photocatalyst's surface (18). The infrared data in Figure 2 shows that after NaTaO<sub>3</sub> photocatalyst was subjected to illumination in a coupling reaction system for 12 h, its infrared spectrogram had obvious C=C, C=O, -CH<sub>3</sub>, and -CH<sub>2</sub> characteristic absorption peaks when compared with NaTaO<sub>3</sub> before the photocatalytic reaction. This result indicated that after the photocatalytic reaction lasted for a period, the reactants (isopropanol and acetone) and products (pinacol) or intermediate products (2,5-hexanedione, and others by GC/Mass analysis, in Figure S3) might been adsorbed or accumulated on the NaTaO<sub>3</sub> surface.

Hence, deactivated NaTaO<sub>3</sub> was treated by ultrasonic washing with acetone, and then it was roasted in a muffle furnace under 500°C (Table 3). Then, NaTaO<sub>3</sub> photocatalytic activity was evaluated after the adsorbed species on the surface were removed (Table 3). Experimental results indicated that ultrasonic washing and high-temperature roasting treatment could remove adsorbed species occupying the catalyst's surface. However, photocatalytic activity efficiency remained low. This result indicated that NaTaO<sub>3</sub> photocatalyst deactivation was not caused by the adsorption of by-products or intermediate products on NaTaO<sub>3</sub> surface. Also, studies have reported that over-consumption of hydroxyl on the photocatalyst's surface would result in photocatalyst deactivation. Photogenerated OH

with strong oxidizability is a key factor for promoting a series of oxidative reactions (19). Hydrothermal treatment of deactivated photocatalyst could remove some intermediate products adsorbed on its surface, could supplement hydroxyl consumed in the reaction, promote OH generation, and regenerate deactivated catalyst. However, results showed that the hydrothermal treatment of NaTaO<sub>3</sub> photocatalyst at 180°C did not improve the conversion rates of reactants. On the contrary, the treatment inhibited the conversion rate of acetone (Table 3). This result also indicated that the over-consumption of OH on the catalyst's surface was not the main reason for the deactivation of NaTaO<sub>3</sub> photocatalyst. The XPS data showed that the displacement of binding energies of Ta and Na atoms on NaTaO<sub>3</sub> photocatalyst's surface after reacting for 12 h changed (Figure 4), which meant the microstructure of NaTaO<sub>3</sub> changed after illumination. As shown in Figure 4, the partial surface NaTaO<sub>3</sub> (E Ta4 $f_{7/2}$  = 27.2 eV and E Ta4 $f_{5/2}$  = 25.7 eV) changed into (E Ta4 $f_{7/2}$  =29.0 eV and E Ta4 $f_{5/2}$  =27.0 eV), which were consistent with previous reference data. Ta has an energy difference in the range 4.1-5.4 eV, depending on the surface preparation procedure (20). And also, the partial surface  $NaTaO_3$  (E Na1s = 1069.5 eV) changed into (E Na1s =1074.1 eV). The results showed that Na content on NaTaO<sub>3</sub> surface decreased after reacting for 12 h (decreased from 21.41% to 17.58%, Table 2). Literature has reported that in NaTaO<sub>3</sub> photocatalytic hydrogen production experiment, Na loss on the catalyst's surface is one of the key factors of photocatalyst deactivation (21). Excess Na enhanced the population of excited electrons and suppressed recombination. Moreover, surface Na<sup>+</sup> acted as H<sup>+</sup> transport channel in the coupling reaction system (8). However, the sodium ions dissolved in aqueous solution, in a phenomenon called photocorrosion, as the photocatalytic reaction occurred, which led to the loss of  $Na^+$  on the NaTaO<sub>3</sub> surface. Thus, in this coupling reaction system, surface Na can be assumed critical for photocatalytic coupling reaction. Hence, photocatalytic activity evaluation of deactivated NaTaO<sub>3</sub> after photocatalysis was implemented after its hydrothermal treatment with 10 M NaOH at 180°C for 12 h. After the treatment, the photoactivity of NaTaO<sub>3</sub> returned to its original level before the reaction. The XRD data indicated that the crystal structure of NaTaO<sub>3</sub> photocatalyst after NaOH hydrothermal treatment basically remained unchanged (Figure 1). XPS data indicated that coordination and contents of Na and Ta on NaTaO<sub>3</sub> surface after NaOH hydrothermal treatment were consistent with that before the reaction. Deactivation of NaTaO<sub>3</sub> in the coupling reaction system can be assumed to be mainly caused by the loss of surface Na.

Furthermore, as shown in our previous report (8), when NaTaO<sub>3</sub> photocatalyst was soaked in 4 M HCl solution for 12 h, surface Na<sup>+</sup> was exchanged with H<sup>+</sup>, which led to the This article is protected by copyright. All rights reserved.

evident decrease in pinacol generation rate, as shown in Figure S4. An expected pleasure, the photocatalytic activity of NaTaO<sub>3</sub> returned to its original level after hydrothermal treatment with 10 M NaOH. This further emphasized the significant role of surface Na<sup>+</sup> on the photocatalytic activity of NaTaO<sub>3</sub> (Figure 5).

#### Regeneration and stability of deactivated NaTaO<sub>3</sub> in coupling reaction system

The NaTaO<sub>3</sub> photocatalyst regenerated with NaOH hydrothermal treatment was used in photocatalytic coupling reaction system. Repetitive regenerated 10-cycle (T) operation test, the regenerated NaTaO<sub>3</sub> consistently showed excellent reactant conversion rate and target product pinacol selectivity (Table 4). This result further verified that in the coupling reaction system, loss of surface Na on NaTaO<sub>3</sub> was the main reason for its deactivation. NaOH hydrothermal treatment could regenerate deactivated NaTaO<sub>3</sub> photocatalyst. Thus, a simple and efficient photocatalyst regeneration method was successfully demonstrated.

# CONCLUSION

In NaTaO<sub>3</sub> photocatalytic isopropanol dehydrogenation coupling and acetone hydrogenation coupling reaction systems, NaTaO<sub>3</sub> photocatalyst lost its photo-activity after three cycles. Results indicated that runoff of Na on the surface was the main reason for the deactivation of NaTaO<sub>3</sub> photocatalyst. After hydrothermal treatment of the deactivated NaTaO<sub>3</sub> with 10 M NaOH at 180°C for 12 h, the photocatalytic activity returned to the original level. The photocatalytic activity of NaTaO<sub>3</sub> remained stable even after 10 cycles of regenerated by 10 M NaOH. This study developed a simple and efficient photocatalyst regeneration method.

#### ACKNOWLEDGEMENTS

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#### SUPPORTING INFORMATION

Additional Supporting Information is available in the online version of this article:

Figure S1. A schematic of the employed photocatalysis reactor.

Figure S2. The XPS full spectra of the NaTaO<sub>3</sub> photocatalyst at different conditions.

**Figure S3.** GC-MS spectra of the coupling reaction system (a) 2,5-hexanedione, (b) pinacol. **Figure S4.** Response of DMB formation to an exchange of Na<sup>+</sup> ions on NaTaO<sub>3</sub> surfaces with HCl and to a subsequent re-exchange with NaOH.

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### FIGURE CAPTIONS

Figure 1. XRD pattern of NaTaO<sub>3</sub> photocatalyst before and after reaction.

Figure 2. FTIR spectra of NaTaO<sub>3</sub> photocatalyst before and after reaction.

Figure 3. UV-vis spectra of NaTaO<sub>3</sub> photocatalyst before and after reaction.

**Figure 4.** XPS spectra of the samples: (a) Ta 4f of NaTaO<sub>3</sub> before photocatlytic reaction; (b) Ta 4f of NaTaO<sub>3</sub> after photocatlytic reaction; (c) Ta 4 f of NaTaO<sub>3</sub> and Ta<sub>2</sub>O<sub>5</sub> after photocatlytic reaction, then hydrothermal treated in 10 M NaOH solution at  $180^{\circ}$ C for 12 h; (d) Na 1s of NaTaO<sub>3</sub> before photocatlytic reaction; (e) Na 1s of NaTaO<sub>3</sub> after photocatlytic reaction; (f) Na 1s of NaTaO<sub>3</sub> after photocatlytic reaction, then treated in 10 M NaOH solution at  $180^{\circ}$ C for 12 h.

**Figure 5.** The deactivated NaTaO<sub>3</sub> photocatlyst was regenerated by hydro-thermal treatment of 10 M NaOH at 180°C for 12 h.

Cuel	Conv	Conversion (%)		Selectivity (%)	
Cycle	Acetone	Isopropanol	DMB	HXD	
1T	61.25	53.12	91.25	3.32	
2T	58.02	19.18	82.67	15.60	
3T	55.01	7.60	77.31	19.02	
a Departion	anditions: 1.0 a NaTaO	nhotoootolyat 200 mI	of reastant aquaque	solution (0.25 mol	

	Table 1.	The evaluation	of stability	of NaTaO <sub>3</sub>	photocatalys	$t^{[a]}$ .
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<sup>[a]</sup>Reaction conditions: 1.0 g NaTaO<sub>3</sub> photocatalyst, 200 mL of reactant aqueous solution (0.25 mol acetone and 0.25 mol isopropanol); light, 300 W high pressure Hg lamp ( $\lambda$ =365 nm); temperature, ~25°C; reaction duration, 12 h. (2,3-dimethyl-2,3-butanediol abbreviated as DMB, 2, 5-hexanedione abbreviated as HXD).

Table 2. Surface atom content of NaTaO<sub>3</sub> photocatalyst under different conditions.

Conditions -	Sur	face atom content (100	0%)
Conditions	Na	Ta	0
Before reaction	21.41	12.64	48.43
After reaction	17.58	12.95	52.11
Treated with NaOH	20.58	13.01	47.49

**Table 3.** The reaction activity of regenerated NaTaO<sub>3</sub> photocatalyst<sup>[a]</sup>.

Catalyst	<b>Conversion</b> (%)		Selectivity (%)	
Regeneration	Acetone	Isopropanol	DMB	HXD
<sup>[b]</sup> Ultrasonic and roasting	61.25	53.12	91.25	3.32
<sup>[c]</sup> Hydrothermal treatment	58.02	19.18	82.67	15.60
<sup>[d]</sup> Treated with 10 M NaOH	55.01	7.60	77.31	19.02

<sup>[a]</sup>Reaction conditions: 1.0 g NaTaO<sub>3</sub> photocatalyst, 200 mL of reactant aqueous solution (0.25 mol acetone and 0.25 mol isopropanol); light, 300 W high pressure Hg lamp ( $\lambda$ =365 nm); temperature, ~25°C; reaction duration, 12 h. <sup>[b]</sup>The deactivated NaTaO<sub>3</sub> was treated by ultrasonic washing with acetone, and then it was roasted in a muffle furnace under 500°C; The deactivated NaTaO<sub>3</sub> was treated by hydrothermal methods at 180°C. <sup>[d]</sup>The deactivated NaTaO<sub>3</sub> was hydrotherma treated by 10 M NaOH at 180°C for 12 h.

Cycles (T)	Conversion (%)		Selectivity (%)	
Cycles (1)	Acetone	Isopropanol	DMB	HXD
1T	61.25	53.12	91.25	3.21
2T	60.69	53.36	90.91	3.71
3T	61.36	53.87	90.83	4.01
4T	66.72	51.66	90.97	3.44
5T	62.60	54.37	90.61	3.55
6T	63.30	56.18	91.23	3.15
7T	66.72	56.82	90.99	3.30
8T	68.71	59.12	90.76	3.89
9T	64.02	61.28	91.25	3.00
10T	64.58	61.28	90.99	3.45

Table 4. The stability evaluation of regenerated NaTaO<sub>3</sub> photocatalyst.

<sup>[a]</sup>Reaction conditions: 1.0 g NaTaO<sub>3</sub> photocatalyst, 200 mL of reactant aqueous solution (0.25 mol acetone and 0.25 mol isopropanol); light, 300 W high pressure Hg lamp ( $\lambda$ =365 nm); temperature, ~25°C; reaction duration, 12 h.





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