

A novel electro-catalytic degradation method of phenol wastewater with Ti/IrO₂-Ta₂O₅ anodes in high-gravity fields

Jing Gao, Junjuan Yan, Youzhi Liu, Jiacheng Zhang and Zhiyuan Guo

ABSTRACT

In the electro-catalytic degradation process of phenol wastewater, bubbles and mass transfer limitation will result in the decrease in wastewater degradation efficiency, a long electrolysis time and a high energy consumption. Self-made Ti/IrO₂-Ta₂O₅ anodes and a high-gravity electro-catalytic reactor were used to improve them. The Ti/IrO₂-Ta₂O₅ anode was prepared with a thermal decomposition method and characterized by scanning electron microscopy (SEM). Under optimum conditions, the removal efficiencies of phenol, total organic carbon (TOC) and chemical oxygen demand (COD) respectively reached 94.77%, 50.96% and 41.2% after 2 h electrolysis in the high-gravity field, which were respectively 10.93%, 16.72% and 24.84% higher than those in the normal gravity field. For about the same removal efficiencies, the electrolysis time and energy consumed in the high-gravity field were 33.3% and 15.4% lower than those consumed in the normal gravity field, respectively. The degradation pathway of phenol detected by high performance liquid chromatography (HPLC) was unchanged in the high-gravity field, but the degradation rate of phenol increased. The Ti/IrO₂-Ta₂O₅ anode provided good stability because the removal efficiencies of phenol and TOC decreased slightly and the surface morphology of the coating was almost unchanged when it had been used in electrolysis for 11 months, about 1,200 h, in the high-gravity field. Results indicated that the phenol wastewater degradation efficiency was improved, the time was shortened, and the energy consumption was reduced in the high-gravity field.

Key words | degradation pathway, electro-catalysis, high-gravity, phenol wastewater, Ti/IrO₂-Ta₂O₅ anode

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INTRODUCTION

Phenol, a kind of hazardous water pollutant, commonly exists in wastewater originating from oil refineries, cokeries, plastic plants, paper plants, synthetic chemicals, petrochemicals, pharmaceuticals, and other enterprises (Luo *et al.* 2015). Phenol exhibits high toxicity, carcinogenicity, and mutagenicity to ocean life, livestock, and human beings (Wang *et al.* 2014). Therefore, various technologies have been developed to degrade phenol wastewater, such as physical adsorption (Damjanovic *et al.* 2010), biological treatment (Fang *et al.* 2006), wet oxidation (Santos *et al.* 2006), photochemical oxidation (Shi *et al.* 2015), ultrasonic reaction (Kidak & Ince 2006) and electro-catalytic oxidation (Feng *et al.* 2010). The electro-catalytic oxidation method has

a unique advantage that electrical energy is used instead of chemical reagents to degrade organic compounds. Electro-catalytic oxidation of wastewater is a clean process, and the treatment devices are simple in operation and have a small footprint (Abaci *et al.* 2005; Steter *et al.* 2014). However, bubbles adhere to the electrode surface in the process of electro-catalytic oxidation, resulting in a small contact area between the electrolyte and electrode and limiting the amount of active particles generated. Meanwhile, mass transfer is limited by the slow diffusion rate of ions; the different ionic concentrations between the bulk solution and electrode surface creates concentration polarization (Lu *et al.* 2012), resulting in decreasing wastewater degradation

efficiency, long electrolysis time and high energy consumption.

Previous studies were mainly focused on the study of electrodes with high oxygen evolution potential for alleviating the influence of bubbles, especially the titanium-based metal-coated oxide electrodes (Park et al. 2012; Shestakova et al. 2014; Zhao et al. 2014). Among metal oxide electrodes, a Ti/IrO₂ electrode, which has the appropriate IrO₂ loading, can possess high electro-catalytic activity (Calderon et al. 2010). Inert oxide incorporated into the electrode could increase the stability of the IrO₂ coating (Rocha et al. 2014), for example a Ti/IrO₂-Ta₂O₅ electrode, which exhibits good electro-catalytic activity and electrochemical stability (Da Silva et al. 2004; Tolba et al. 2010; Aris et al. 2015). However, the mass transfer cannot be improved effectively. The high-gravity technology is an intensification of the mass transfer process and has been applied in wastewater treatment (Liu et al. 2010; Gao et al. 2012), gas purification, dust removal (Liu 2009), water electrolysis (Cheng & Scott 2003; Lao et al. 2011), chlor-alkali electrolysis (Cheng et al. 2002; Wang et al. 2008), and material preparation (Eftekhari 2003, 2004; Mahito et al. 2004). The interphase buoyancy factor $\Delta\rho g$ is large and the interphase slip rate is fast in the high-gravity field (Liu 2009), while bubbles can be easily separated from the electrode surface and electrolytes in the electro-catalytic oxidation process of wastewater. In this way, the interference of bubbles is weakened. At the same time, the renewal speed of the electrode surface and interphase contact area can be increased, concentration polarization can be reduced, and the mass transfer process can be intensified in the high-gravity field (Liu 2009). This will be beneficial to the pollutants in wastewater that contact the electrode surface, increase the rate of electro-catalytic oxidation and reduce the reaction time. So, the degradation efficiency of wastewater can be improved, and the electrolysis time and energy consumption can be reduced in the high-gravity field.

In this paper, the electro-catalytic oxidation process of phenol wastewater in a high-gravity field with self-made catalytic anodes of Ti/IrO₂-Ta₂O₅ and a self-made high-gravity electro-catalytic reactor were studied. The process was optimized. The phenol wastewater degradation pathways, efficiency, electrolysis time, energy consumption and stability were researched in the high gravity field and compared with those in the normal gravity field to discuss whether the electro-catalytic oxidation process of phenol wastewater can be improved in the high-gravity field.

EXPERIMENTAL

Preparation and characterization of Ti/IrO₂-Ta₂O₅ anodes

The Ti/IrO₂-Ta₂O₅ anodes were prepared through thermal decomposition of IrO₂-Ta₂O₅ coating solutions on the cylindrical titanium substrate. Before coating, the titanium substrate underwent sandblasting, 5 min ultrasonic cleaning in 0.2 mol/L NaHCO₃ solution using an ultrasonic cleaner (SG7200HDT, GuTel, China, 59 KHz, 350 W), and 2 h etching in 10.0% boiled oxalic acid. Then, the titanium cylinder was ultrasonically cleaned in distilled water. According to the Ir:Ta molar ratio of 0.5:0.5, H₂IrCl₆·6H₂O and TaCl₅ were respectively dissolved in hydrochloric acid and ethanol to prepare the coating solution. Then, the coating solution was brushed evenly onto the titanium substrate. The solvent was evaporated at 100 °C for 10 min and the residue was calcined at 550 °C for 10 min in a pre-heated oven. Afterwards, the coatings were cooled to room temperature naturally. The same procedure was repeated until the desired oxide load was obtained. The surface morphology of the coating was observed with scanning electron microscopy (SEM; SU3500, Hitachi, Japan) before and after use.

High-gravity electro-catalytic reactor

A high-gravity electro-catalytic reactor was designed and used in this study. As shown in Figure 1, the concentric cylindrical anodes and cathodes are respectively connected with an anode plate and a cathode plate, which are arranged alternately to form a multistage electro-catalytic reactor. The volume of the reactor is 3 L and the effective circular volume is 5 L. The inter-electrode spacing is 1.0 cm. Cathodes and

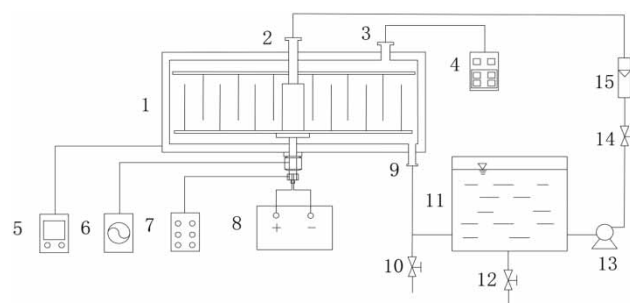


Figure 1 | Flow diagram for phenol wastewater treatment by the high-gravity electro-catalytic reactor. (1) high-gravity electro-catalytic reactor; (2) wastewater inlet; (3) gas outlet; (4) gas detection system; (5) temperature control system; (6) high-gravity control system; (7) power and signal transmission system; (8) DC power supply; (9) wastewater outlet; (10, 12, 14) valve; (11) wastewater storage tank; (13) pump; (15) flowmeter.

cathode plates were relatively static to the setup shell. The center of the anode plate was connected with the shaft and could rotate to form different high-gravity fields. The intensity of the high-gravity field was calculated with the high-gravity factor (β) (Liu 2009). The high-gravity factor (β) is expressed as:

$$\beta = \frac{rN^2}{900}, \quad (1)$$

where β is the high-gravity factor; N is the rotating speed (r/min); r is the average radius of the anodes (m).

Wastewater in the storage tank entered the high-gravity electro-catalytic reactor via a pump and a flowmeter. Treated water could be directly discharged from the outlet via a valve or discharged into the wastewater storage tank for recycling. The total current in the system was supplied by a DC-regulated power supply.

Electro-catalytic oxidation of phenol

The cylindrical Ti/IrO₂-Ta₂O₅ electrode described above was used as the anode and a cylindrical stainless steel electrode was used as the cathode. Firstly, 5.0 L of sample solution containing 500 mg/L phenol and NaCl was used as a supporting electrolyte. The phenol concentration was detected by high performance liquid chromatography (HPLC) (UltiMate 3000, Thermo Fisher, USA) with a C₁₈ reversed-phase column (Accucore XL C₁₈, 250 × 4.6 mm, 4 μm) at 25 °C. The mobile phase was methanol/water (60:40, v/v) and the flow rate was 1.0 mL/min. The detection wavelength was 270 nm and the injection volume was 20 μL.

The total organic carbon (TOC) was detected by TOC tester (1030 W, OI, USA). The chemical oxygen demand (COD) was determined spectrophotometrically by a COD analyzer (5B-3A, Lianhua, China) according to a Chinese national standard method (HJ/T 399-2007). The phenol, TOC and COD removal efficiency (η) was defined as:

$$\eta = \frac{c_0 - c_t}{c_0} \times 100\% \quad (2)$$

where c_0 and c_t are the concentration of phenol or TOC or COD in solution before and after electrolysis at time t (mg/L), respectively.

The energy consumption E (kWh/kgCOD) was calculated using Equation (3)

$$E = \frac{U \cdot I \cdot \Delta t}{1000 \cdot V \cdot \Delta COD} \times 100\% \quad (3)$$

where U is the voltage (V), I is the applied current (A), Δt is the duration of the electrolysis (h), V is the volume of feed

wastewater (m³), and ΔCOD results from the removed COD during the time Δt (kg/m³).

RESULTS AND DISCUSSION

Electro-catalytic oxidation of phenol

The effects of current density, high-gravity factor, circulating liquid flow rate, NaCl electrolyte concentration, initial pH and electrolysis time on the removal efficiencies were studied with the self-made catalytic anodes Ti/IrO₂-Ta₂O₅ and the self-made high-gravity electro-catalytic reactor.

Effect of current density

Current density is a particularly important parameter for controlling phenol removal. As shown in Figure 2, the removal efficiencies of phenol and TOC increase with the increase in current density and increase slowly at a current density of 500 A/m². A high current density can promote electron transfer in the system and ·OH generation on the electrode surface, resulting in promoting the electro-catalytic oxidation of phenol and intermediate products. But the higher current density leads to a higher cell voltage and energy consumption. Therefore, the optimum current density was 500 A/m².

Effect of high-gravity factor

The effects of the high-gravity factor on phenol and TOC removal efficiencies are shown in Figure 3. The removal

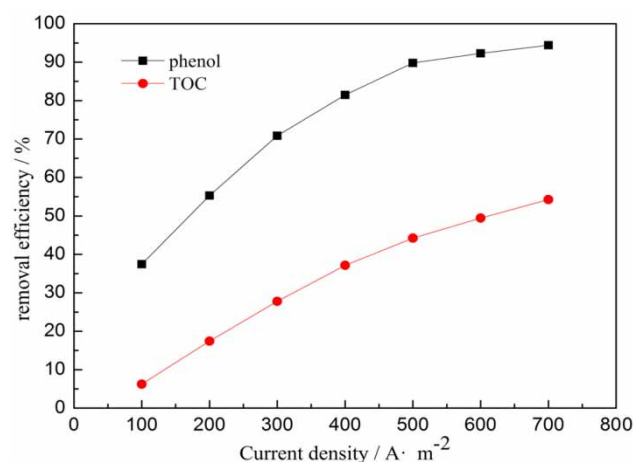


Figure 2 | Effects of the current density on the removal efficiency (phenol concentration: 500 mg/L; NaCl concentration: 8.5 g/L; pH: 4; high-gravity factor: 20; circulating liquid flow rate: 50 L/h; degradation time: 2 h).

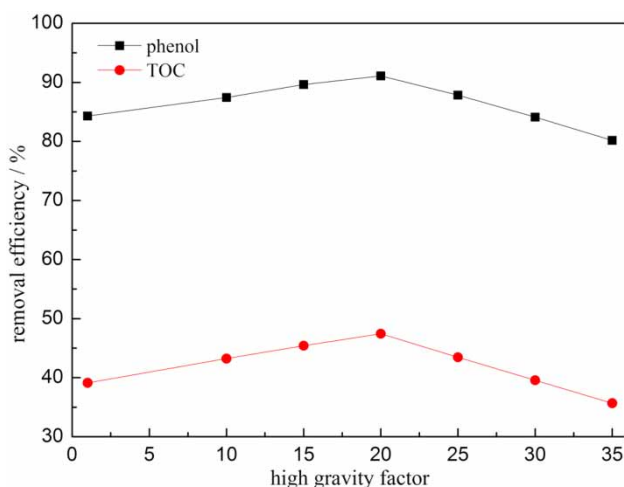


Figure 3 | Effect of the high-gravity factor on the removal efficiency (phenol concentration: 500 mg/L; NaCl concentration: 8.5 g/L; current density: 500 A/m²; pH: 4; circulating liquid flow rate: 50 L/h; degradation time: 2 h).

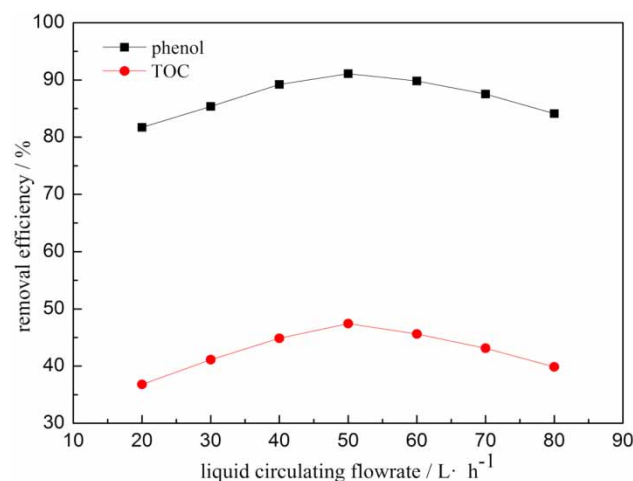


Figure 4 | Effect of the circulating liquid flow rate on the removal efficiency (phenol concentration: 500 mg/L; NaCl concentration: 8.5 g/L; current density: 500 A/m²; pH: 4; high-gravity factor: 20; degradation time: 2 h).

efficiencies of both phenol and TOC increase with the increase in the high-gravity factor, and are highest under a high-gravity factor of 20 because the system mass transfer efficiency is low when the high-gravity factor is small or one (a normal gravity field). In the normal gravity field, polarization of the interface layer would be generated, so that phenol and other organic compounds could not spread to the electrode surface for direct oxidation reaction. The diffusion rate of active particles in the system was also affected by the high-gravity factor. Therefore, the diffusion rate affected the indirect oxidation reaction. In addition, the bubbles could not be separated from the electrode surface in time, so the contact area between electrolyte and electrode would decrease. Therefore, the removal efficiencies of phenol and TOC decreased.

However, the removal efficiencies of phenol and TOC decreased with the further increase in the high-gravity factor. When the high-gravity factor was too large, the organic compounds could not completely react with the electrode surface and active particles in the solution, thus resulting in decreases in the removal efficiencies of both phenol and TOC. The high removal efficiencies could be obtained when the gravity factor was 20.

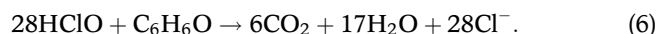
Effect of circulating liquid flow rate

Figure 4 shows the effect of circulating liquid flow rate on the removal efficiencies of phenol and TOC. When the high-gravity factor was constant, the circulating liquid flow rate was the main influencing factor of equipment liquid volume

and the contact area between electrolyte and electrode. With the increase in the circulating liquid flow rate, the liquid volume and the contact area between the electrolyte and electrode increased. At the same time, the removal efficiencies of phenol and TOC increased. However, too large a flow rate would lead to shorter contact time between the organic compounds in solution and the electrode. A lot of organic compounds in the solution were replaced by the new solution before reacting with the electrode or active particles, so the removal efficiencies decreased significantly. Therefore, the optimum circulating liquid flow rate was 50 L/h.

Effect of NaCl electrolyte concentration

The effect of electrolyte concentration on phenol and TOC removal is shown in Figure 5. The phenol and TOC removal efficiencies increase with the increase in the concentration of NaCl electrolyte, and the increase rate gradually decreases when the concentration of NaCl is more than 9.5 g/L. The possible influencing reactions of removal efficiencies are expressed as follows:



In Equation (4), Cl⁻ may be oxidized into Cl₂ on the anode surface and Cl₂ can be hydrolyzed to produce

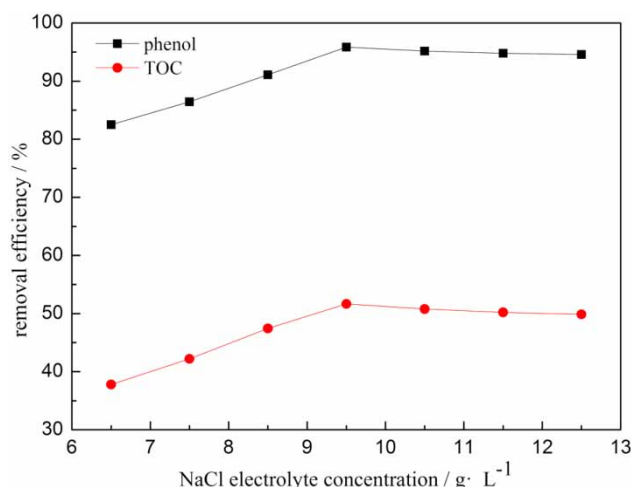


Figure 5 | Effect of NaCl electrolyte concentration on the removal efficiency % (phenol concentration: 500 mg/L; current density: 500 A/m²; pH: 4; high-gravity factor: 20; circulating liquid flow rate: 50 L/h; degradation time: 2 h).

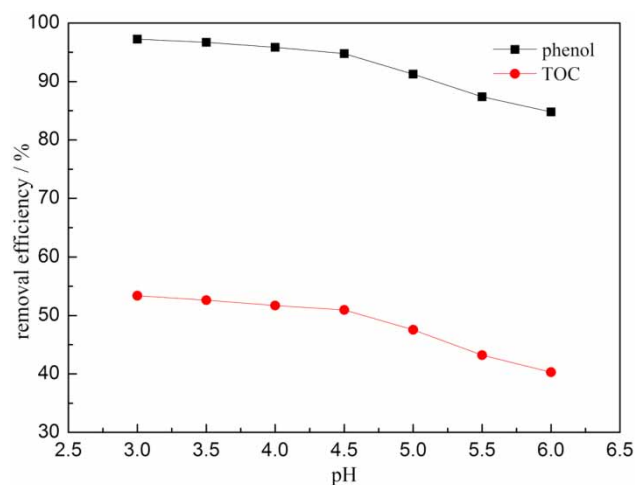


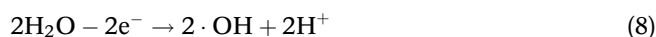
Figure 6 | Effect of pH on the removal efficiency (phenol concentration: 500 mg/L; NaCl concentration: 9.5 g/L; current density: 500 A/m²; high-gravity factor: 20; circulating liquid flow rate: 50 L/h; degradation time: 2 h).

HClO in Equation (5). Then, phenol and other organic compounds can be reduced by HClO to generate Cl⁻ and form the Cl⁻ cycle (Equation (6)).

The phenol oxidation process involves the reaction between phenol and oxidants such as ·OH and HClO. Therefore, the concentration of Cl⁻ directly affects the removal efficiency of phenol. With the increase in the concentration of Cl⁻, the amount of ClO⁻ increases and the removal efficiencies of phenol and TOC increase. However, under the constant current power supply and phenol concentration, the presence of too much Cl⁻ would lead to the collision among ions, thus affecting the indirect oxidation process of phenol and decreasing the removal efficiencies of phenol and TOC. The suitable concentration of NaCl was 9.5 g/L.

Effect of initial pH

The effect of initial pH on phenol and TOC removal is shown in Figure 6. The removal efficiencies of phenol and TOC gradually decrease with the increase in pH value and decrease fast over a pH value of 4 because Cl₂ can be hydrolyzed to produce HClO in solution and the ionization equilibrium of HClO exists in solution (Equation (7)). The equation reveals that it is favorable to the formation of HClO in an acidic field. In addition, the ·OH can be produced by H₂O in an acidic field (Equation (8)). Therefore, the removal efficiencies of phenol and TOC can be improved by the oxidation of HClO and ·OH.



However, too much acid affects the yield of ·OH, which will decrease the removal efficiencies of phenol and TOC and increase the cost. Considering the above factors, the optimal pH value was 4.

Effect of electrolysis time

The effect of different electrolysis times on the removal efficiencies of phenol, TOC and COD in the normal gravity field and high-gravity field is shown in Figure 7. The

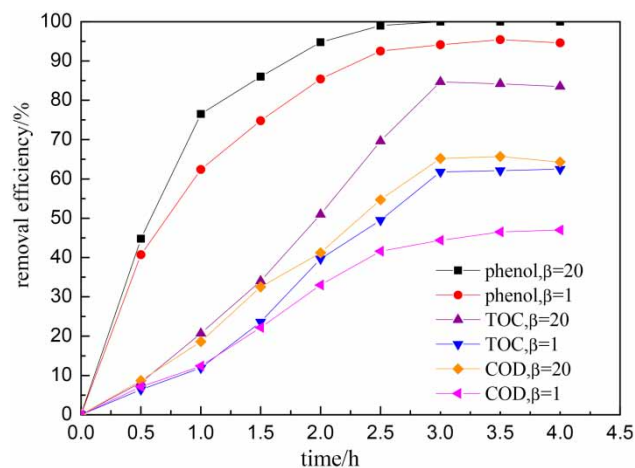


Figure 7 | Effect of electrolysis time on the removal efficiency in high-gravity field and normal gravity field (phenol concentration: 500 mg/L; NaCl concentration: 9.5 g/L; current density: 500 A/m²; pH: 4; circulating liquid flow rate: 50 L/h).

removal efficiencies of phenol, TOC and COD increase with the increase in electrolysis time. However, the increasing rate of phenol removal efficiency reduces gradually and the increasing rate of TOC and COD removal efficiencies increase gradually after 2 h electrolysis. From the kinetic point of view, the higher initial concentration of phenol leads to the faster degradation rate. Therefore, the phenol removal efficiency was significantly higher than TOC and COD removal efficiencies. As the electrolysis time was prolonged, the concentration of phenol gradually decreased and the concentration of intermediate products increased gradually. As a consequence, most of the active particles were mainly used in oxidation of intermediate products, so that the increasing rate of phenol removal efficiency tended to decrease and the increasing rate of TOC and COD removal efficiencies tended to increase.

It was also observed that the removal efficiencies of phenol, TOC and COD in the high-gravity field were higher than those obtained in the normal gravity field because the update rate of the electrode surface could be accelerated in the high-gravity field so as to increase the contact area between electrolyte and electrode. The mass transfer among the ions in the solution was enhanced, thus decreasing the concentration polarization, increasing the interphase slip rate, weakening the bubbles effect and improving the removal efficiencies. The removal efficiencies of phenol, TOC and COD were respectively 94.77%, 50.96% and 41.2% in the high-gravity field, which were 10.93%, 16.72% and 24.84% higher than those in the normal gravity field after 2 h electrolysis, and reached 100%, 84.72% and 65.23% in the high-gravity field, which were higher than the 94.12%, 60.80% and 44.4% removal efficiencies in the normal gravity field after 3 h electrolysis, respectively. So, for about the same removal efficiencies, the electrolysis time was shortened in the high-gravity field. The electrolysis time consumed in the high-gravity field was 33.3% shorter than that consumed in the normal gravity field.

Degradation pathway analysis

The electro-catalytic oxidation process of phenol with Ti/IrO₂-Ta₂O₅ anodes was monitored by recording its HPLC chromatograms for different degradation times in the high-gravity field and normal gravity field (Figure 8). A single well-defined peak of phenol gradually diminished, while some new peaks appeared. According to the retention

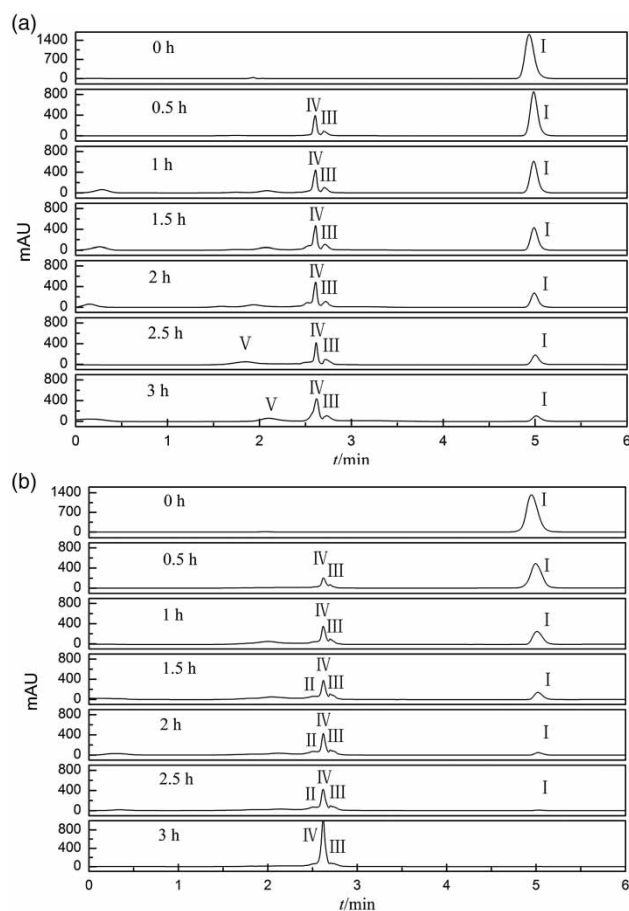


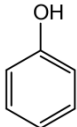
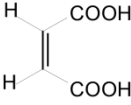
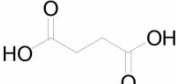
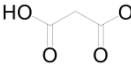
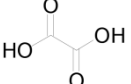
Figure 8 | HPLC chromatograms of different electro-catalytic degradation time: (a) in high-gravity field; (b) in normal gravity field.

time of standard products, we obtained the possible degradation intermediate products (Table 1).

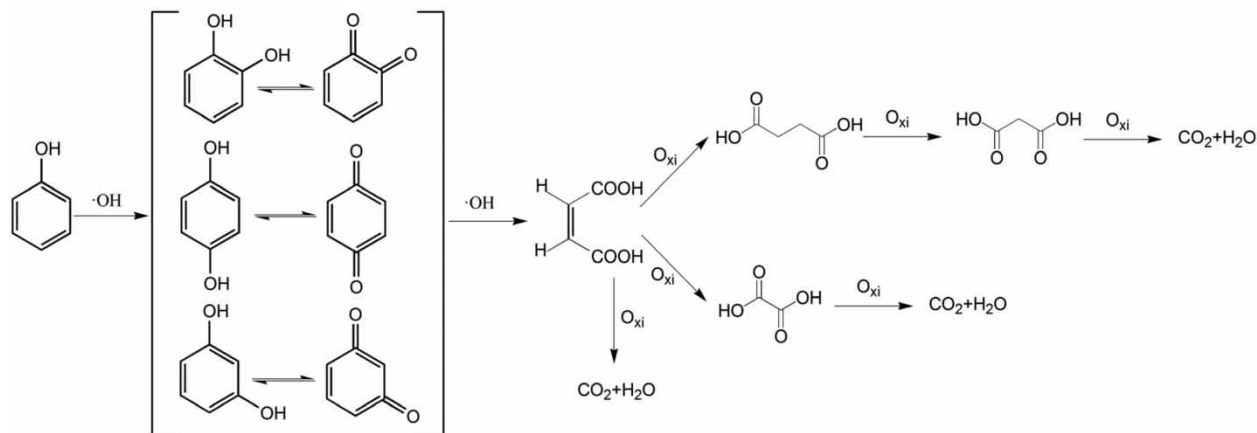
As shown in Figure 8 and Table 1, a simple pathway of phenol degradation was proposed, as illustrated in Figure 9. In the early reaction stage, $\cdot\text{OH}$ attacked benzene molecules and substances, such as catechol, resorcinol, hydroquinone, and benzoquinone were generated. However, these benzene molecules and substances could not be observed in HPLC chromatograms because the benzene ring-opening reaction was fast. In the second stage, the benzene ring was further oxidized to produce carboxylic compounds. In the final stage, these compounds were decomposed into CO₂, H₂O and other small molecules due to the interaction of $\cdot\text{OH}$ and HClO.

The degradation process and intermediate products were similar whether in the high-gravity field or in the normal gravity field, as shown in Figures 8 and 9. However, after the same degradation time, the peak of phenol in the high-gravity field diminished faster than that in the

Table 1 | The main degradation intermediates of HPLC chromatograms

Chromatographic peak	t_R /min	Material	Structure
I	5.007	Phenol	
II	2.512	Maleic acid	
III	2.698	Succinic acid	
IV	2.622	Malonic acid	
V	2.047/1.853	Oxalic acid	

normal gravity field. The results indicated that the degradation pathway of wastewater was unchanged in the high-gravity field, but the degradation efficiency of wastewater was improved.

**Figure 9** | Possible phenol degradation pathway by Ti/IrO₂-Ta₂O₅ anodes.**Table 2** | Effect of using time of Ti/IrO₂-Ta₂O₅ anode on the removal efficiency in the high-gravity field (phenol concentration: 500 mg/L; NaCl concentration: 9.5 g/L; current density: 500 A/m²; pH: 4; circulating liquid flow rate: 50 L/h; degradation time: 3 h)

Time	First time	1 month	3 months	5 months	11 months
Removal efficiency of phenol	100%	100%	100%	100%	98.6%
Removal efficiency of TOC	84.7%	84.5%	82.3%	81.1%	80.4%

Stability of anode in the high gravity field

To assess the stability of the Ti/IrO₂-Ta₂O₅ anode, the surface morphology of the Ti/IrO₂-Ta₂O₅ coating and degradation efficiencies of wastewater were analysed during use of the anode in the high-gravity field. When the Ti/IrO₂-Ta₂O₅ anode was used in electrolysis for 11 months, about 1,200 h, in the high gravity field, the removal efficiencies of phenol and TOC decreased slightly, as shown in Table 2. And the surface morphology of the Ti/IrO₂-Ta₂O₅ coating was almost unchanged, the coating showed no cracking or pull off, as shown in Figures 10 and 11. The performance of the Ti/IrO₂-Ta₂O₅ anode does not get worse. The anode can provide good stability for phenol wastewater degradation in the high-gravity field.

Energy consumption analysis

The main operational cost of the electro-catalytic oxidation process is related to the energy consumption. The total energy consumption (E_{total}) is mainly caused by the DC power supply ($E_{DC \text{ power supply}}$), pump (E_{pump}) and motor (E_{motor}) in the high-gravity field and is only caused by the DC power supply in the normal gravity field. Table 3 shows the energy consumptions in the normal gravity field

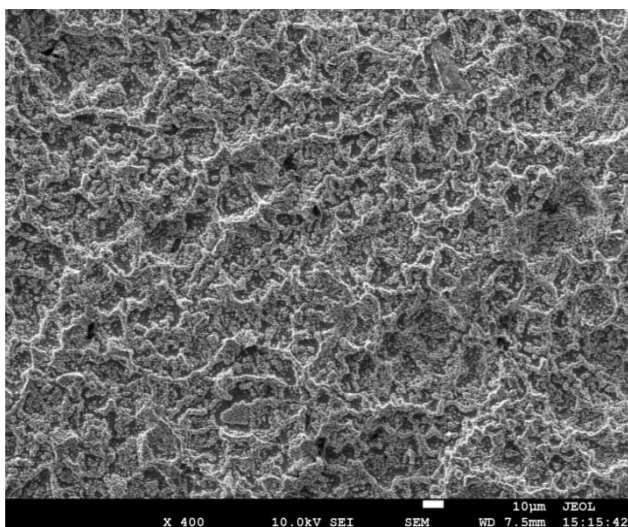


Figure 10 | SEM images of Ti/IrO₂-Ta₂O₅ anode (before electrolysis, ×400).

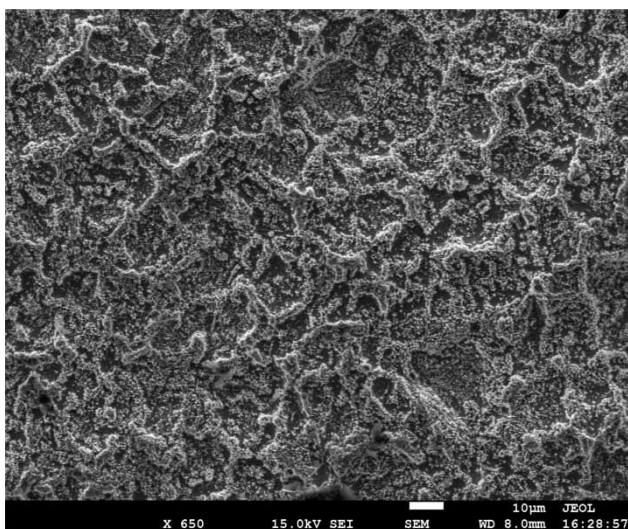


Figure 11 | SEM images of Ti/IrO₂-Ta₂O₅ anode (after electrolysis for 11 months in the high gravity field, ×650).

and in the high-gravity field when the differences in phenol, TOC and COD removal efficiencies were small, 3 h and 2 h were taken for electrolysis respectively. The E_{DC} power supply

and E_{total} in the high-gravity field were 33.3% and 15.4% lower than those in the normal gravity field, respectively. For about the same removal efficiencies, the required degradation time was shortened and the energy consumption was reduced in the high-gravity field.

CONCLUSIONS

The self-made Ti/IrO₂-Ta₂O₅ anodes and self-made high-gravity electro-catalytic reactor were used for electro-catalytic degradation of phenol wastewater. Under the optimum conditions of a phenol concentration of 500 mg/L, NaCl concentration of 9.5 g/L, current density of 500 A/m², high-gravity factor of 20, liquid circulation flow rate of 50 L/h and initial pH value of 4, the removal efficiencies of phenol, TOC and COD were higher than those in the normal gravity condition. For about the same removal efficiencies, the electrolysis time and energy consumed in the high-gravity field were lower than those consumed in the normal gravity field.

The degradation pathway of phenol wastewater detected by HPLC was proposed to be that phenol was firstly converted into aromatic compounds, such as pyrocatechol, hydroquinone, resorcinol, and benzoquinone, then into organic acids, such as maleic acid, succinic acid, malonic acid, and oxalic acid, and finally into CO₂ and H₂O. The degradation pathway of phenol wastewater was unchanged, but the degradation efficiency of phenol wastewater was improved in the high-gravity field.

The performance and stability of the Ti/IrO₂-Ta₂O₅ anode did not get worse when the Ti/IrO₂-Ta₂O₅ anode was used in electrolysis for 11 months, about 1,200 h, in the high gravity field because the removal efficiencies decreased slightly and the surface morphology of the Ti/IrO₂-Ta₂O₅ coating was almost unchanged.

The study provided a novel, effective and clean method for electro-catalytic degradation of phenol wastewater.

Table 3 | Energy consumption analysis in normal gravity field and in high-gravity field (phenol concentration: 500 mg/L; NaCl concentration: 9.5 g/L; current density: 500 A/m²; pH: 4; circulating liquid flow rate: 50 L/h; In the normal gravity field: $\beta = 1$; in the high-gravity field: $\beta = 20$)

Item	Time (h)	E_{DC} power supply (kWh/kgCOD)	E_{pump} (kWh/kgCOD)	E_{motor} (kWh/kgCOD)	E_{total} (kWh/kgCOD)
In the normal gravity field	3	39.6	null	null	39.6
In the high-gravity field	2	26.4	2.5	4.6	33.5
Comparison	33.3%	33.3%	null	null	15.4%

ACKNOWLEDGEMENTS

The study was financially supported by the National Natural Science Foundation of China (Grant No. 21376229) and the Natural Science Foundation for Young Scientists of Shanxi Province, China (Grant No. 2015021033).

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First received 12 September 2016; accepted in revised form 19 April 2017. Available online 4 May 2017