Decontamination of Wastewater Containing Acetone by Continuous Supercritical

Water Oxidation

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Abstract

This study was conducted to investigate the destruction removing efficiency (DRE) of wastewater containing acetone by supercritical water oxidation (SCWO). A 200mL Inconel 625 reactor with continuous flow of pressurized hot water was used in this study. The experimental conditions were controlled at sub- and supercritical state of water. Reactor's temperature was set in the ranges of 350~480 and pressure in between 23~27Mpa. The DRE of acetone depended upon both reactor temperature and acetone concentration. The DRE was evaluated by the removal efficiency of chemical oxygen demand (COD) and total organic carbon (TOC) of acetone solution. Acetone at the $4 \sim 15\%$ (v/v) and reaction temperature at 350 , the COD and TOC removal are around $40 \sim 60\%$. As reaction temperature raise to 480 , the DRE of TOC and COD were 98%. For SCWO system presently used in our lab, optimum concentration for acetone destruction were in between 8~10%. Higher reaction temperature will be helpful to promote DRE. As reaction temperature above the auto ignition temperature of acetone, oxidation of acetone could be greatly enhanced and resulted in highly 465 destruction efficiency by SCWO.

Key words: Supercritical water oxidation, Acetone, Destruction removing efficiency

Introduction

In the past decade, the use of pressurized water above its critical state provides a green process for wastewater treatment. Water at room temperature possesses high dielectric constant with excellent solvating power for inorganic salts. However, hot and compressed water behaves much more like organic solvent because of the diminishing of hydrogen bonding strength. The superior characteristic in the supercritical phase, the mass transfer resistance between oxidant and organic is eliminated, therefore, the oxidation kinetic becomes extremely fast. Water near or above its critical point (Tc=375 , Pc=22.1MPa) can cause highly oxidized efficiency. Advantages of SCWO are environmentally benign with clean discharge. Recently, SCWO has been proven as a powerful technology for destruction organic compounds from a wide variety of wastes [1-8].

Acetone is a manufactured chemical and is widely used to make plastic, fibers, drugs and organic solvent to clean the organic residuals on electronic wafers. Breathing moderate to high levels of acetone for a short period of time can cause eye, nose, throat, and lung irritation. Other harmful symptoms include headache, confusion, vomiting, unconsciousness, and possibly coma. Acetone miscible with water is difficult to degrade by traditional wastewater

treatment. In this work, a preliminary study was done for water contains high concentration of acetone. The acetone was destructed by SCWO in a continuous flow system with a tube reactor. The effects of temperature and pressure on the destruction removing efficiency were investigated. The experimental results indicated that the obtained reaction parameters in supercritical water may provide useful information to scale up design for treating of spent acetone in water.

Materials and methods

A continuous flow SCWO system is shown in Figure 1. The equipment was designed and assembled by Asia Giant cooperation in Taiwan. Acetone is mixed with water to simulate the waste and the acetone concentration in reactor ranges from 4 to 15%. The removal of chemical oxygen demand (COD) and total organic carbon (TOC) are used to evaluate the DRE of acetone. The acetone is then pressurized and flows into the flow system by pump P1. The wasted feeding stream is cooled by water before entering the reactor to prevent plugging due to the potential precipitation of inorganic salt and metal oxides in the wastes and the formation of various tars in reactor tube. The pre-heated water is pressurized and fed into the reactor by pump P2. Compressed air supplied from a10 hp air cylinder (10~90mL/min) is pressurized by a booster P3 (AGT 14/62, 17~42mL/min). A constant system pressure in the reactor was maintained at 23~27MPa through this experiment and sufficient air is used as the oxidant in this work. The pre-heated water, air and acetone are mixed in reactor, which is made of Inconel-625 with internal volume of 200mL in which submerged in a furnace. The reaction temperature mainly between 350 to 480 was applied to the reactor. The treated solution effused from the reactor is cooled and flows into a gas-liquid separator T1 where the mixture of carbon dioxide generated in the process and the residue of the excess air is separated from the liquid effluent. A control valve installed on the gas effluent at the top of the separator regulates the pressure of the system, and a needle valve connected at the bottom of the separator constantly discharges the liquid effluent to maintain the liquid level in the separator. The process water leaving the reactor after SCWO was collected and characterized by its pH value, and percent DRE based on TOC (Apollo 9000 TOC combustion analyzer) and COD (dichromate oxidation method) measurement of treated water. The DRE of acetone is then calculated as $(COD_i COD_f)/CODi$ × 100%.

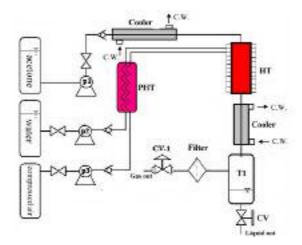


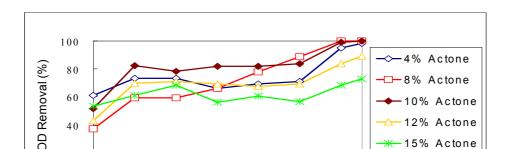
Figure 1 Illustration of the continuous flow system of SCWO

Results and discussion

The initial COD, TOC, and pH of the acetone solution with five different levels of concentration are shown in Table 1. Based on COD and TOC analysis the results of DRE of acetone by SCWO with five different concentration at eight levels of temperature are summarized in Figure 2 and 3. The reproducibility of SCWO data was evaluated by the percent relative standard deviation (RSD) and the RSD of these data is typically $\pm 2.3\%$. It is known that temperature, temperature, and sufficient amount of air supply are the most important parameters in SCWO and they have both theoretical and practical implications for the destruction process. The experimental result showed that the efficiency of decomposition steeply improve under supercritical condition as opposed to sub-critical region. For example, At sub critical (i.e., 350), a 10% acetone solution with 51% DOC removal while 83% COD removal was obtained at supercritical temperature at 380 . As the temperature increases to 480, a >98% COD removal was obtained. At the sub-critical temperature such as 350 the DRE of acetone was generally poor. A very interesting result was found that there is no clear trend in enhancing the DRE of acetone in the temperature range between 370~450 . This phenomenon is related to the auto-ignition temperature (AIT) of acetone. Because the AIT for acetone is 465 , The DRE of acetone was found greatly enhanced as the SCW temperature higher than its auto-ignition temperature. Degradation of acetone is often not complete, as it is in sub-critical water oxidation, with some compounds, such as acetic acid, being particular formed. The completeness of degradation of acetone in water could also be evaluated by the pH alter of the aqueous solution. Higher DRE resulted in little bit decreasing the pH of solution, owing to the acetic acid is produced. Nevertheless, the acetic acid is often relatively benign or biodegradable. Figure 4 shows the change pH of aqueous acetone solution with SCW temperature. The measured initial pH of acetone aqueous solution is around 4.88, after the SCWO the pH drops to 2.8~3.3.

Aqueous acetone	4%	8%	10%	12%	15%
COD (mg/L)	91,956	126,095	193,108	202,303	257,477
TOC (mg/L)	23,513	35,845	49,716	54,972	75,913
pН	4.88	4.92	4.95	4.98	5.00

 Table 1
 The initial COD, TOC, and pH of the acetone solution with five different levels of concentration



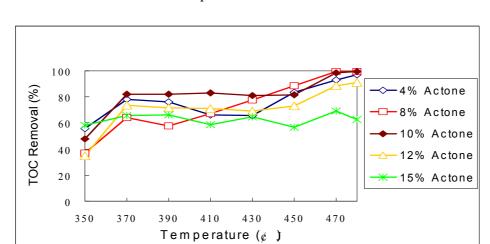


Figure 2 The COD removal of acetone solution *versus* reaction temperature.

Figure 3 The TOC removal of acetone solution *versus* reaction temperature.

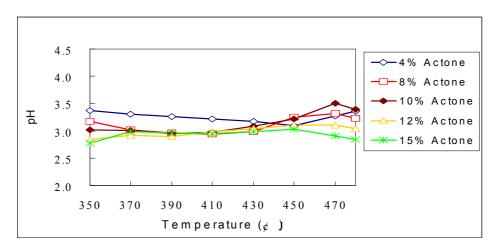


Figure 4 The pH variation of acetone solution *versus* reaction temperature.

Conclusions

In this preliminary work, the treatment of wastewater containing acetone by SCWO is found feasible. The concentration of acetone in simulated water is from $4\sim15\%$ and the experiments are conducted in a continuous flow system. The reaction temperature and pressure are set in between 350~480 and 23~27MPa with overwhelming excess air . Two stages of enhanced DRE were observed, DRE increases significantly at the critical point and auto-ignition temperature (465) of acetone. For the system presently used in our lab the optimum concentration ranges for acetone degradation were in between 8~10%.

Acknowledgements

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