

Removal of disinfection byproduct precursors by a combined electrochemical and adsorption process

Department of Civil and Environmental Engineering, Waseda University. Student Member ○Wei Lu

Department of Civil and Environmental Engineering, Waseda University. Regular Member Yutaka Sakakibara

1. Introduction

Disinfection by-products in drinking water may pose health risks, therefore the control or removal of their precursors is a major challenge in water supply. As conventional coagulation and sand filtration system was not always effective in removing the precursors, different innovative processes have been studied¹⁾. In this study, a new electrochemical process was proposed. The removal performances of humic acid (HA) under continuous operation were studied using two different electrodes.

2. Materials and Methods

Fig. 1 shows the apparatus used in this study. Electrolytic cell reactor (Height: 89.0mm, Inner Diameter: 35.6mm) is the main part of the experiment apparatus. SnO_2 electrodes (2.3cm^2) were prepared according to a procedure reported by Cui et al. (2009)²⁾. A stainless mesh electrode (2.3cm^2) was also prepared. Then a solid polymer electrolyte (SPE) membrane was sandwiched by SnO_2 electrode and stainless mesh electrode, while granular cathodes (Pt/Ti or mixture of Pt/Ti and granular activated carbon) were packed in four compartments connecting to the anode. The size of GAC was about 0.25~0.50 cm. Before being used as cathodes, GAC was dipped into HA solution to reach an adsorption saturation state. Electric current passing to each compartment was controlled by the electric power supply (Kikusui, PMC 160-0.4mA).

Synthetic HA solution was prepared according to the following procedure: 0.1g of HA powder was dissolved into 1L of NaOH solution (0.01mol/L), diluted 10 times, and HCl was added to adjust pH around 7.5. After that, this solution was filtered by 0.45 μm membrane filters (Toyo, A045) and used for continuous experiment. During the experiment, syringe filter unit was used to take samples. TOC and UV_{254} were used as the main monitoring indicators. Samples were periodically taken to measure TOC and UV_{254} . TOC was analyzed with TOC analyzer (Shimadzu TOC-5000), while UV_{254} with Spectrophotometer (Shimadzu UV-160).

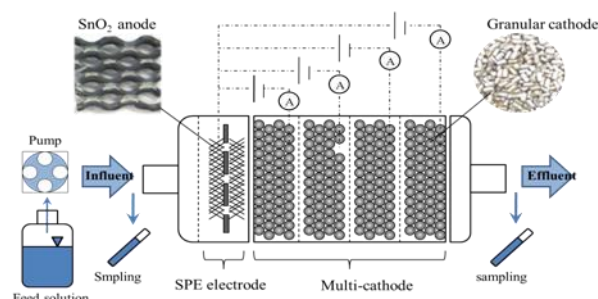
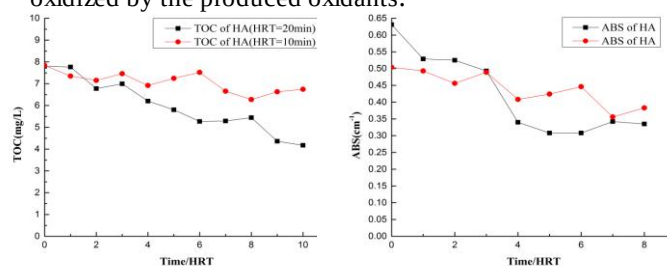


Fig. 1 Experiment Apparatus

3. Results and Discussion

Fig. 2 shows experimental results of continuous treatments using granular Pt/Ti cathodes. HRT was set at 10 and 20 minutes, respectively. Other parameters were kept constant (i.e. pH=7.5, Current Density=22mA/cm²). As shown in Fig. 2(a) and Fig. 2(b), effluent TOC concentrations and UV_{254} became smaller than influent. Effluent TOC concentrations and UV_{254} were smaller at longer HRT. In the reactor, ozone was produced from SnO_2 anode and reduced to OH radicals at granular cathodes^{3,4)}. Humic acid could be directly oxidized at anode, or be oxidized by the produced oxidants.



(a) TOC Removal (b) UV_{254} Removal

Fig. 2 Removal of HA (Pt/Ti Cathodes)

Fig. 3 shows experimental results of the combined process using mixed Pt/Ti and GAC granular cathodes at different HRTs, i.e. 10, 20 and 30 minutes, respectively. Other parameters were constant (pH=7.5, Current Density=22mA/cm²). The results show that the removal performance of the combined process was obviously enhanced, and removal efficiencies increased with the

Keyword: Disinfection byproduct precursors, Electrochemical and adsorption process, Granular activated carbon

Address: 3-4-1 Okubo, Shinjuku, Tokyo, Japan. Email:sakaki@waseda.jp

increase of HRT. That is, compared with Pt/Ti cathodes, effluent TOC concentrations and UV_{254} were smaller in the case of the mixed cathodes of Pt/Ti and GAC. It was supposed that HA was adsorbed onto the surface of GAC, and at the same time, OH radicals produced from cathodes reacted effectively with absorbed humic acid. This humic acid adsorption or acclimation on GAC as well as concurrent OH radical formation from GAC might enhance the removal performance.

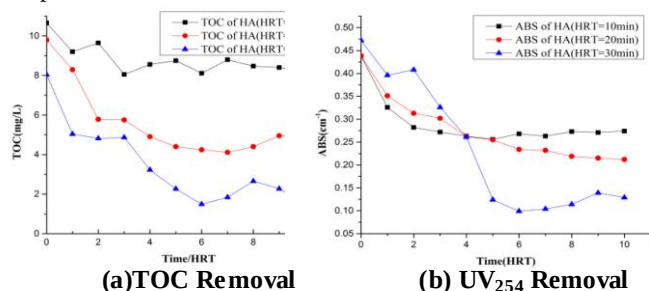


Fig. 3 Removal of HA at different HRTs

(Pt/Ti & GAC cathodes)

Fig. 4 shows experimental results of the combined process at different current densities, 11, 22 and 33 mA/cm², respectively. Other parameters were kept constant (pH=7.5, HRT=20 minutes). Results show that removals of HA were better at larger current densities. This is because higher concentration of ozone and OH radicals were produced at larger current densities.

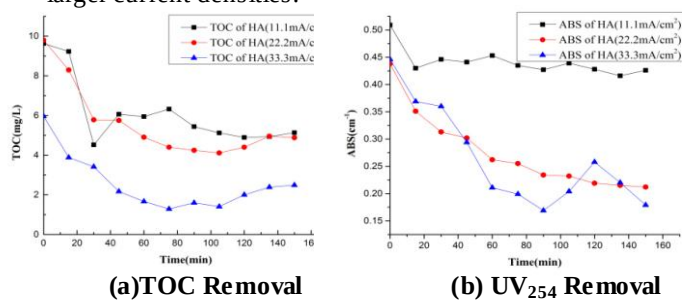
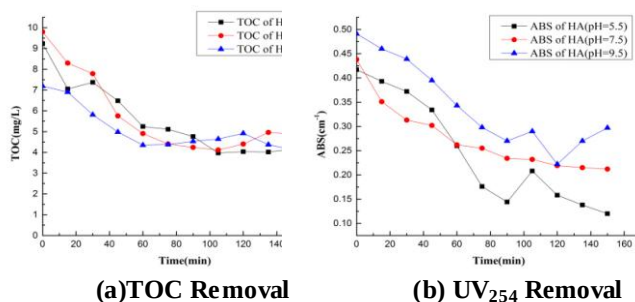


Fig. 4 Removal of HA at different current densities

(Pt/Ti & GAC cathodes)

Fig. 5 shows experimental results of the combined process at different pH, 5.5, 7.5 and 9.5, respectively. Other parameters were the same (HRT=20 minutes, Current Density=22 mA/cm²). From the results, it can be seen that the removal efficiency decreased at higher pH. Some reasons may account for this result. Firstly, former research showed that the production efficiency of ozone decreased with the increase of pH⁵⁾. Moreover, the concentration of carbonate and bicarbonate ions increased at higher pH. These chemicals were scavengers of OH radicals⁶⁾. So the oxidation of HA was influenced at higher pH.



(a) TOC Removal

(b) UV₂₅₄ Removal

Fig. 5 Removal of HA at different pHs

4. Conclusions

Experimental results demonstrated that the present electrochemical process could remove and decompose HA. Removal performance of HA increased with increasing HRT and current densities. It was also demonstrated that the use of GAC is effective to enhance removal performance. A further study on kinetics and long-term experiments will be needed to evaluate the removal performances.

Acknowledgement

This research was supported in part by the Grant-in-Aid for Scientific Research (B) (No. 24360219), the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

Reference

- 1) T. Bond, E. H. Goslan, S. A. Parson, B. Jefferson: Treatment of Disinfection Byproduct Precursors, *Environmental Technology*, **32**(1), 1-25, 2011.
- 2) Yuhong Cui, Yunhai Wang, Bin Wang, Haihui Zhou, Kwong-Yu Chan, and Xian-Yao Li: Electrochemical Generation of Ozone in a Membrane Electrode Assembly Cell with Convective Flow, *Journal of Electrochemical Society*, **156**(4), E75-E80, 2009.
- 3) Kishimoto, N., Morita, Y., Tsuno, H., and Oomura, T.: Advanced Oxidation Effect of Ozonation Combined with Electrolysis, *Water Research*, **39**, 4661-4672, 2005.
- 4) M. Shimura, M.H. Uddin, Y. Sakakibara, and T. Tanaka: An advanced Oxidation Process Using A 3-Dimensional Electrochemical Reactor Generating and Reducing O₃, *J.JSCE, Ser. G (Environmental Research)*, **48**(7), 697-703, 2011.
- 5) Christensen P. A., Yonar T, Zakaria K.: The Electrochemical Generation of Ozone: A Review, *Ozone: Science & Engineering*, **35**(3), 149-167, 2013.
- 6) D. Vione, S. Khanra, S. C. Man, P. R. Maddigapu, R. Das et al: Inhibition vs. enhancement of the nitrate-induced phototransformation of organic substrates by the OH scavengers bicarbonate and carbonate, *Water Research*, **43**(18), 4718-4728, 2009.