

Design of a Water Purification System Using Modified Mineral Waste Materials (MMWM) and Activated Carbon Derived from Waste Materials

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Abstract: The study attempts to test and optimize the removal efficiency of haloacetic acids and antibiotics in a water purify device using a low-cost, modified mineral waste material (MMWM) accompanied with activated carbon (AC). There are four stages in this research to determine its maximum removal efficiency. In stage I, the concentration of haloacetic acids in different water samples were tested. Although it was within the acceptable limit, high attention must be paid due to its strong carcinogenicity. In the second stage, the adsorption ability of AC and MMWM for one type of haloacetic acids (dichloroacetic acid) was determined. Moreover, in order to achieve a maximum removal efficiency, the effects of temperature $(10^{\circ}C - 40^{\circ}C)$, size of the filling materials (40 mesh, 60 mesh, 200 mesh and 300 mesh), and ratio between AC and MMWM in the filling materials (1:4-4:1) on the removal of dichloroacetic acids (DCAA) and antibiotics were studied by an experimental design of L16(4)3 orthogonal array. The results indicated that the optimal conditions for removing DCAA in water samples are 30 °C, 20 mesh and ratio 3:2 of AC to MMWM. Consequently, the removal efficiency of the existed water purify device was improved from 29.26% to 71.46% after combing the optimum conditions.

Keywords: Activated Carbon (AC); Antibiotics; Haloacetic Acid; Modified Mineral Waste Materials (MMWM); Removal Efficiency

1. Introduction

Nowadays, water utilities are facing numerous challenges in protecting drinking water supplies from water-borne diseases and contaminants. To supply safe drinking water, chlorination is the most common disinfection method of drinking water. It is an effective way to kill many kinds of bacteria that may be harmful to human's health. However, during the process, the residual form of disinfectant can reacts with the natural organic matter (NOM) in water, leading to the formation of undesirable disinfection-by-product (DBPs) in water systems.

The natural organic matters in water contain the organic materials such as proteins, humic substances, hydrophilic acid or carbonates. According to some studies^[1], the residual chlorine can cause the formation of more than 600 DBPs in water supply systems. The of these two main classes compounds are trihalomethanes (THMs) and haloacetic acids (HAAs). Some of the compounds are considered potentially carcinogenic. There are nine species of HAAs, and five of them are currently regulated by EPA^[2]. They are MCAA (chloroacetic acid), DCAA (dichloroacetic acid), TCAA (trichloroacetic acid), MBAA (bromoacetic acid), and DBAA (dibromoacetic acid). In order to regulate

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those compounds, many agencies pay more attention to these compounds espousing for the public and they have placed limitations on their levels in drinking water.

Haloacetic acids are also found in industrial wastes and in other field like drugs, dyes and chemicals, they are colorless with a low volatility and dissolve easily in water, when consumed in drinking water; HAAs are rapidly absorbed into the bloodstream and they are carried throughout the body. Moreover, the different seasons, water sources, and drinking water treatment plants have different concentrations of HAAs. The more harmful Dichloroacetic acid (DCAA) and trichloroacetic acid (TCAA) make up about 80% of all HAAs, followed by BDCAA and BCAA, which make up around 15%. And this research mainly focuses on the DCAA and TCAA.

2. Materials and methods

This study is divided into four stages. In stage I, the concentrations of HAAs in drinking water, bottle water and tap water were detected by IC equipment (Doris. S, 2011)^[3]. The ability of AC and MMWM to remove HAAs in a specific water purification system was tested in stage II. After running of this stage, the results were analyzed and if both AC and MMWM have ability to remove HAAs, it was expected that combination of these two materials can improve the removal efficiency, therefore, the best radio between the use of AC and MMWM was found out during stage III. In the last stage, one of the most common water purify device would be tested and re-designed based on the result of stage III. It is expected that the removal ability for HAAs and antibiotics of the water purify device can be improved after combining the optimum conditions of AC and MMWM.

3. Results

3.1 Stage I

From the report: "EPA Method 557 Quantitation of Haloacetic Acid, Bromate, and Dalapon in Drinking Water Using Ion Chromatography and Tanderm Mass Spectrometry", the total amount of haloacetic acids for all nine HAAs of the tap water sample was 35.62ppb. And for the regulated HAA, the total amount was 30.21 ppb. Also, Su and Fang's experiment^[4] showed that there were no DCAA and TCAA in bottle water while tap water contain 6.43ppb DCAA and 18.29ppb TCAA. There are many references supported that the DCAA and TCAA can be detected in tap water. According to the test result, the concentration of DCAA in tap water was 0.0402 mg/L, and there was no DCAA detected in bottle water and drinking water. The reason why TCAA can not be detected may be explain as follows: the concentration of TCAA in water sample was too lower to be determined. Different equipment and methods have different detection limits. It was quite difficult to determine the detection limit values in this experiment; the retention time of TCAA was coincided with phosphate according to IC analysis; and it was difficult to separated them. Hence, no data of its concentration was obtained by using the calibration curve of TCAA.

Based on the result of stage I, the concentration of dichloroacetic acid was not exceeded the standard for drinking water quality in China (GB5479-2006)^[5]. Although it was within the acceptable limit. However, due to its strong carcinogenicity, people pay high attention to it.

3.2 Stage II

This stage tested whether AC and MMWM have removal ability for HAA. It is obvious to find that different sizes of filling materials had different removal efficiencies. The characteristics of the carbon materials, like the particle size, surface area, surface chemistry, density or hardness, all these factors have influence on the efficiency of adsorption. Compared with MMWM, AC had better removal ability. Moreover, the removal efficiency of AC (60 mesh) was the highest, with the number of 52.87%. And next is AC (180-200 mesh), its efficiency was up to 52.58% while AC (20-60 mesh) have the lowest efficiency. As for MMWM, it is a kind of green adsorption material from modified mineral waste. Its ability to remove the DCAA was not strong from the test results. The 1-0.3mm diameter MMWM can remove only 39.93% DCAA in a water sample. What's more, the removal efficiency of MMWM (0.3-0.075 mm) and MMWM (0.075-0.045 mm) were 41.33% and 41.42%, respectively.

Comprehensively, in this experiment, the removal efficiency of AC and MMWM for haloacetic acid was

remaining 40%-50%. From Li's report (2005)^[6], GAC (Granulated Activated Carbon) had ability to adsorb HAA with the removal efficiency between 20% and 30%, if it was operated with empty bed contract time ranging from 10-25min, the removal efficiency would increase to 48% and 61%. Therefore, it was expected that the combination of AC and MMWM would have a better removal ability than only one of the materials.

3.3 Stage III

According to the result, all removal efficiencies for DCAA were more than 80%, and the highest can reach up to 91.30%. Moreover, through the range analysis of the orthogonal test, the optimal level of each factor was determined, and the hierarchy of factor that affected the removal efficiency also obtained: the ratio between AC and MMWM > room temperature > size of AC and MMWM. After analysis, the optimal conditions for removing DCAA in water samples are 30 °C, 200 mesh and ratio 3:2 of AC to MMWM.

Additionally, through the variance analysis of the orthogonal test, it demonstrated that there was a significant difference among the temperature, size of AC and MMWM and ratio of AC/MMWM. The ratio between AC and MMWM had the most significant effect on improving removal efficiency of the purification column, followed by the temperature and size.

As for the antibiotics, those filling materials also have ability to remove different kinds of antibiotics in water samples. Many researches focus on investigating the concentrations of different kinds of antibiotics in drinking water and they found that terracycline antibiotics, macrolide antibiotics, sulfonamide antibiotics and penicillins were common in water samples (Qin, 2013)[7]. NFX (Norfloxacin) SMX and (Sulfadimethoxine) were chosen as analytes to test the removal ability of AC and MMWM in this experiment. After calculation, the adsorption condition of the two antibiotics. The removal efficiency of three parallel experiments of SMX were 91.4%, 90.2%, and 91.1%, respectively. And the removal efficiency of three parallel experiments of NFX were 81.5%, 81.1% and 81.1%, respectively. Based on the results in this stage, it indicated that the optimal condition have higher removal efficiency for SMX.

3.4 Stage IV

Taking the method of Chen's study $(2013)^{[8]}$, at stage III the optimum conditions (temperature: 30°C, size: 200 mesh, ratio of AC/MMWM: 3/2) were found for removing DCAA in water sample, after combining the optimum conditions with water purify device. The average removal efficiency was 71.46% with a standard deviation of 1.087. The result shows that the removal efficiency before was improved with the average number being just up to 29.26%.

Comparing the result with stage III, removal efficiency was lower than that of the adsorption purification unit setup. There were two main reasons: (1) The flow rates of water purify device was faster than purification unit setup. Although in this experiment the inner diameter, temperature, the size of AC and MMWM and ratio are controlled, and the flow rate was not taken into consideration. This factor may have impact on the test results. (2) There were other filling materials in water purify device. It contains a magnet, a filter and medical stone, which also influence the experimental data.

4. Conclusion

In this study, the main purpose was achieved. The optimum conditions of purification unit were found out through the orthogonal test and after combining the optimum conditions to the existing water purify device, the removal efficiency increased than before.

At stage I, small amount of DCAA was detected in tap water and the concentration was within acceptable limits. At stage II, study mainly focuse on the removal ability of different sizes of AC and MMWM, all of them can remove HAAs in water samples. Moreover, 60 mesh of AC in size and 0.075-0.045 mm of MMWM in size had the highest removal efficiency. And at stage III, the sequence of factors affected the removal efficiency was in the following order: the ratio between AC and MMWM > room temperature > size of AC and MMWM. Also, the optimal conditions obtained by using the orthogonal experimental design was shown as follows: ratio 3: 2 of AC and MMWM, 30 °C of temperature and 200 mesh of AC/MMWM in size. The optimal removal efficiency of purification column occurring in the orthogonal test was 91.30%. Moreover, the purification column also had removal for the two kinds of antibiotics, more than 90% of SMX can be removed in the optimal condition. The removal efficiency of NFX was lower than SMX, which is above 80%. After the existing water purify device was re-designed and improved at stage IV, the removal efficiency for HAA was increased from 29.26% to 71.46% in average.

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