

A Study on the Applicability of Torrefied Wood Flour Natural Material Based Coagulant to Removal of Dissolved Organic Matter and Turbidity¹

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ABSTRACT

With the emergence of abnormal climate due to the rapid industrialization, the importance of water quality management and management costs are increasing every year. In Korea, for the management of total phosphorus and total nitrogen, the major materials causing the water quality pollution, coagulants are injected in sewage treatment plants to process organic compounds. However, if the coagulant is injected in an excessive amount to PAC (Poly Aluminium Chloride), a secondary pollution problem might occur. As such, a study on the applicability of natural material-based coagulant is being conducted in Korea. Thus, this study aimed to evaluate the applicability of a mixed coagulant developed by analyzing water quality pollutants T-P, T-N as well as their turbidity, in order to derive the optimum mixing ratio between PAC and torrefied wood flour for the primary settling pond effluent. Under the condition where the content of PAC (10%) and torrefied wood flour is 1%, T-P showed the maximum removal efficiency of 92%, and T-N showed approximately 22%. This indicates that removal of T-N which includes numerous positively charged organic compounds that are equivalent to mixed coagulant is not well accomplished. Turbidity showed the removal efficiency of approximately 91%. As such, 1% of torrefied wood flour was determined to be the optimum addition. As a result of analyzing the removal efficiency for organic compounds by reducing PAC concentration to 7%, T-P showed a high maximum removal efficiency of 91%, T-N showed 32%, and turbidity showed the maximum of 90%. In addition, a coagulation process is performed by using the mixed coagulant based on 1% content of torrefied wood flour produced in this study by performing a coagulation performance comparative experiment with PAC (10%). As a result, PAC concentration was reduced to 30-50%, a similar performance with other coagulants in market was secured, PAC injection amount was reduced that an economic effect can be achieved, and it is considered to perform a stable water treatment that reduces the secondary pollution problem.

Keywords: torrefied wood flour, organic material remove efficiency, coagulation, water quality

1. INTRODUCTION

With the rapid industrialization and the occurrence of abnormal climate based on this, the importance of water quality in water-intake sources, streams and the

management costs are increasing every year (Hwang *et al.*, 2018). In Korea, for the management of total phosphorus and total nitrogen which are the major pollutants in water quality pollution, coagulants are injected in sewage treatment plants and the water-dis-

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solved organic compounds are processed by blending, coagulation, and sedimentation processes. The use of PAC accounts to 58% of all domestic coagulants. Although PAC has a very high content of impurities (heavy metals) on the quality standard, the standard is not properly reinforced. Thus, an excessive injection of coagulant increases the residual aluminum concentration, which leads to the secondary pollution (Park, 2009). While it is different for all the existing wastewater treatment plants based on incoming water quality, in general, 1ppm of 17% PAC coagulant is being injected sequentially, and up to 50ppm is being injected to reduce the total phosphorus of incoming water. For treated water with an excessive amount of coagulant in the same aluminum series with PAC, the water-dissolved aluminum ion could be flowed in drinking water. Moreover, if the dissolved aluminum ion is flowed in human body, it could cause neurological diseases such as Alzheimer's, etc. (Han *et al.*, 2016). In addition, rapid changes in environmental laws occurred after mid-2000 that include prohibition of ocean disposal, policy on pollution load management system, high level processing of sewage treatment plants, installation of total phosphorus processing facilities, and energy self-reliance projects, etc. As such, the domestic environmental medicine market is emphasizing the significance of coagulant application technology and technical management on changes in products and pollution load (MOE, 2017). Biomass is known to be an environmentally friendly resource that can be regenerated in the aspect of resource circulation (Gong *et al.*, 2016). As such, a study is being conducted on the applicability of coagulant that combines natural materials of vegetal and mineral substances, etc. (Nam *et al.*, 2016). If biomass (wood, etc.) is being used as a fuel that goes through thermolysis process and torrefaction process as natural material, the caloric value increases due to the increase in fixed carbon, and the water content is reduced due to thermol-

ysis of hydrophilic functional groups (Jung *et al.*, 2018; Priadi *et al.*, 2019; Chang *et al.*, 2019; Kim *et al.*, 2019; Kim *et al.*, 2018).

In addition, torrefaction is a heat treatment method which applies heat of relatively lower temperature of 200-300°C in anaerobic or nitrogen environment, to reduce contents of moisture, acid compounds with low calories, and hemicellulose that exist in wood biomasses (Lee *et al.*, 2016; Nam *et al.*, 2018).

As such, this study conducted two separate studies based on a previous study: Using a forest by-product of woods damaged by blight and harmful insects to analyze charge and absorption performances of Torrefied Wood Flour manufactured in a high-temperature treatment condition (Yang *et al.*, 2016), and a study on algae removal efficiency of paste coagulant manufactured by combining Torrefied Wood Flour showing a strong negative charge based on heat treatment and cationic polyacrylamide (Yang *et al.*, 2017). The previous study combined the cationic polymer coagulant with Torrefied Wood Flour to manufacture the combined coagulant showing cationic property through surface-modification process. In addition, the study focused on water pollutants according to the principle of removing organic compounds which cause water quality pollution, such as anionic organic compound T-P, etc. (Yang *et al.*, 2019).

Therefore, this study aimed to reduce the use of Torrefied Wood Flour used by the previous study and PAC used in the existing sewage treatment plants. In order to achieve this, Torrefied Wood Flour mixed PAC coagulant was used to analyze water pollutants removal efficiency based on the mixed coagulant concentration to evaluate the applicability of mixed coagulant. The evaluation items are turbidity, total phosphorus, and total nitrogen. This study also aimed to verify the optimal mixing ratio of mixed coagulant, as well as possibility of removing underwater pollutants through results derived from the evaluation.

2. MATERIALS and METHODS

2.1. Test material

2.1.1. Torrefied Wood Flour (TWF)

For Torrefied Wood Flour used as the test material, an oak tree chip for pulp with water content of 12% similar to the one used in the previous study was used. The material of torrefied chip was received from S corporation in Gumi which performed a rapid heat-treatment for 25 minutes at a temperature of 250 °C using superheated steam. The torrefied chip treated with superheated steam was grinded using a small high-speed pulverizer (Wonder Blender) in order to secure uniformity of particles. The used Torrefied Wood Flour was a portion (Zeta potential of more than -40 mV) with the size of 75 um ~ 106 um which is the maximum configuration of distribution based on particle size (Yang *et al.*, 2019).

2.1.2. Poly Aluminum Chloride (PAC)

As an inorganic polymer coagulant, PAC is configured with chemical formula of $(OH_m)Cl_{6-m}]_n$, and it is a coagulant in polymer series made by hydrochloric acid and aluminum. A colorless and a lemon-yellow transparent liquid coagulant were used. PAC is an inorganic polymer chemical compound with alkaline multinuclearized-ion structure having cationic charge, and it shows a strong coagulation (Choi *et al.*, 2016). A commonly used coagulant PAC was used to manufacture the Torrefied Wood Flour mixed coagulant.

2.1.3. Manufacturing mixed coagulant for analyzing coagulation function based on Torrefied Wood Flour content

In order to derive the optimal content of Torrefied Wood Flour during the manufacturing process of Torrefied Wood Flour mixed coagulant, this study manufactured mixed coagulant which combined Torrefied Wood Flour grinded through the superheated

Table 1. Ratio of mixed coagulant to TWF concentration

Division	0.5% TWF	1% TWF	5% TWF	10% TWF
PAC (10%)	500 ml			
TWF	1 g	5 g	25 g	50 g

steam treatment and PAC which is a coagulant in market by diluting them in certain concentration. The mixed coagulant was manufactured as shown in Table 1 below, by setting a mixing ratio with Torrefied Wood Flour contents of 0.5%, 1%, 5%, and 10% with the coagulant PAC (10%) in market. Moreover, the test material for this experiment was manufactured by adding Torrefied Wood Flour to 500 ml PAC (10%), and mixing at 800 rpm for approximately 5 hours (Yang *et al.*, 2016).

2.1.4. Manufacturing mixed coagulant for the analysis of coagulation performance based on PAC concentration

This study determined that 1% content of Torrefied Wood Flour is the optimal addition by considering the organic compound removal efficiency through the previous experiment for deriving the optimal content of Torrefied Wood Flour. In addition, in order to reduce content of PAC to secure economic feasibility and environmental stability, the experiment in this study manufactured the test material by diluting the Torrefied Wood Flour in PAC concentrations of 2%, 5%, 7%, and 10% based on 1% addition of Torrefied Wood Flour, and mixing it for approximately 5 hours at 800 rpm, as shown in Table 2.

Table 2. Ratio of mixed coagulant to PAC concentration

Division	2% PAC	5% PAC	7% PAC	10% PAC
TWF	5 g			
PAC	100 ml	250 ml	350 ml	500 ml

2.2. Experiment method

As shown in Table 1 and Table 2, in order to evaluate organic compound removal efficiency based on conditions of each mixed coagulant manufactured, the outflowing water of primary settling pond at Daejeon sewage treatment plant having a high content of organic compound in water was used. The collected primary settling pond's outflowing water was divided at 1L in 1L beaker using a volumetric flask, and Jar test was performed according to mixed coagulant conditions. Supernatant was collected by going through operating conditions of Jar test as follows: rapid max at 140 rpm 2 min, show-paced mix at 40 rpm 20 min, and sedimentation time of 30 min. After the supernatant was collected, it was filtered through a syringe filter with 45 um opening, and used as the analysis sample. Total phosphorus and total nitrogen were selected as the analysis items, which belong to turbidity and ion of general items in 「Water Pollution Standard Method (No. 2017-4)」.

2.2.1. Turbidity

In order to evaluate turbidity of the primary settling pond outflowing water and treated water, a turbidimeter (Turbidimeter TB 210 IR, Lovibond, Germany) was used for the measurement with 5 times repetition and the analysis was based on turbidity analysis criteria of 「Water Pollution Standard Method (No. 2017-4)」. The turbidimeter has the minimum detection limit of over 0.02 NTU (Nephelometric Turbidity Units), and it is equipped with a light source, and a photoelectric detector. In addition, the light source of tungsten filament operates at a temperature range of 2,200 K-3,000 K, the total passing distance of incident light and scattered light inside the measuring tube does not exceed 10cm, and the angle that absorbed light by the detector does not exceed (90±30)°C with respect to incident light.

2.2.2. Total phosphorus (T-P)

In order to evaluate the total phosphorus of the primary settling pond outflowing water and treated water, the total phosphorus of 「Water Pollution Standard Method (No. 2017-4)」 was measured by using a water quality analyzer (HS-3300, HUMAS) which complies with Ascorbic Acid Reductionism, etc. and it was measured 5 times in repetition.

2.2.3. Total nitrogen (T-N)

In order to evaluate the total nitrogen of the primary settling pond outflowing water and treated water, the total nitrogen of 「Water Pollution Standard Method (No. 2017-4)」 was measured by using a water quality analyzer (HS-3300, HUMAS) which complies with optical density measurement based on chromatography method, and it was measured 5 times in repetition.

2.2.4. Aluminum concentration

In order to evaluate the aluminum concentration of the primary settling pond outflowing water and treated water, the aluminum among heavy metals of 「Water Pollution Standard Method (No. 2017-4)」 was analyzed. The analysis of the sample was performed by FITI Testing & Research Institute, and it was measured 3 times in repetition according to inductively coupled plasma/atomic emission spectrometry.

3. RESULTS and DISCUSSION

3.1 Description of outflowing water of primary settling pond at Daejeon sewage treatment plant

In order to evaluate in-water organic compound removal efficiency and turbidity removal efficiency using polluted water collected from the primary settling pond at Daejeon sewage treatment plant, turbidity, total phosphorus, and total nitrogen items between

Table 3. Characteristics of primary settling tank effluent in DJ WTP

Division	Characteristics
Turbidity (unit : NTU)	60 ± 0.7
T-P (unit : mg/L)	4.11 ± 0.02
T-N (unit : mg/L)	33.785 ± 0.085

polluted and treated water were measured. The result of analyzing the quality of outflowing water of primary settling pond at Daejeon sewage treatment plant are shown in Table 3. In the case of turbidity, it was revealed to be not suitable with turbidity 15 NTU which belongs to SS (suspended solid), level 5 criteria among discharge water quality criteria (MOE, 2013). In the cases of T-P and T-N, it was revealed as exceeding 0.15 mg/L, level 5 criteria among discharge water quality criteria.

3.2. Result of analyzing coagulation performance based on content of Torrefied Wood Flour

T-P and T-N are the representative organic compounds that cause water pollution. The higher concen-

tration of these organic compounds results in facilitation of eutrophication due to the compounds or growth of microorganism in water. In addition, the water quality pollution is worsened due to the leak of toxic substances based on microorganism. The efficiency of removal of organic compound according to the content of torrefied wood and PAC(10%) is shown in Fig. 1. The contents of Torrefied Wood Flour were selected as 0.5%, 1%, 2%, and 5% by referring to the previously conducted study to manufacture the mixed coagulant. In addition, T-P, T-N, and turbidity removal efficiency based on the injection amount of the manufactured mixed coagulant was indicated.

As shown in Fig. 1, the removal efficiency of organic compounds in mixed coagulant containing 1% torrefied wood flour was relatively high. In the case of T-P, the maximum removal efficiency of approximately 92% was shown in PAC 10% coagulant 10 ml. In the case of T-N, approximately 22% was shown in TWF 1%+PAC 7% coagulant 10 ml. The organic compound constituting the total nitrogen reveals a positive charge and it showed a relatively less coagulation effect than the organic compound of total

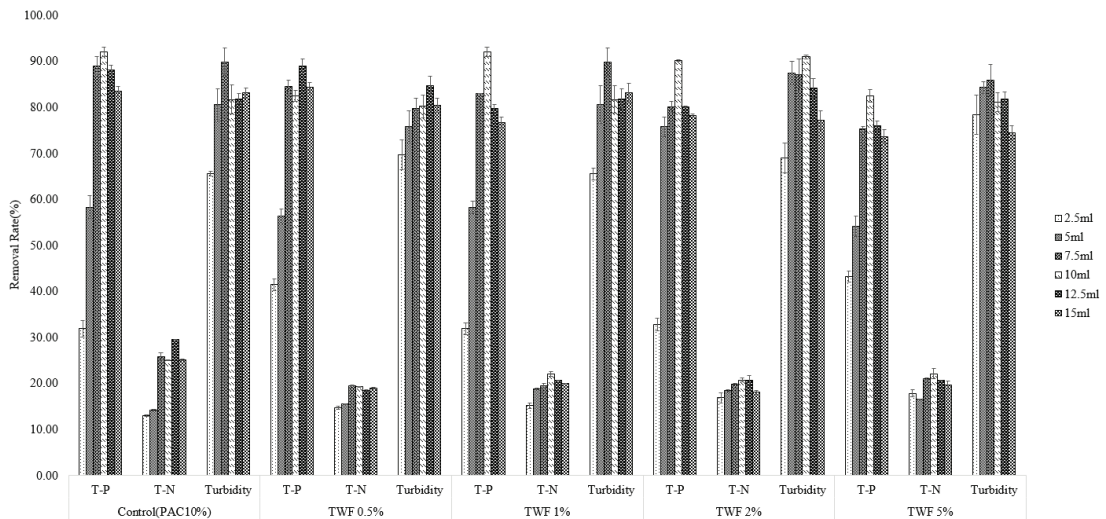


Fig. 1. Comparison of organic matter and turbidity change depending on TWF concentration and input conditions.

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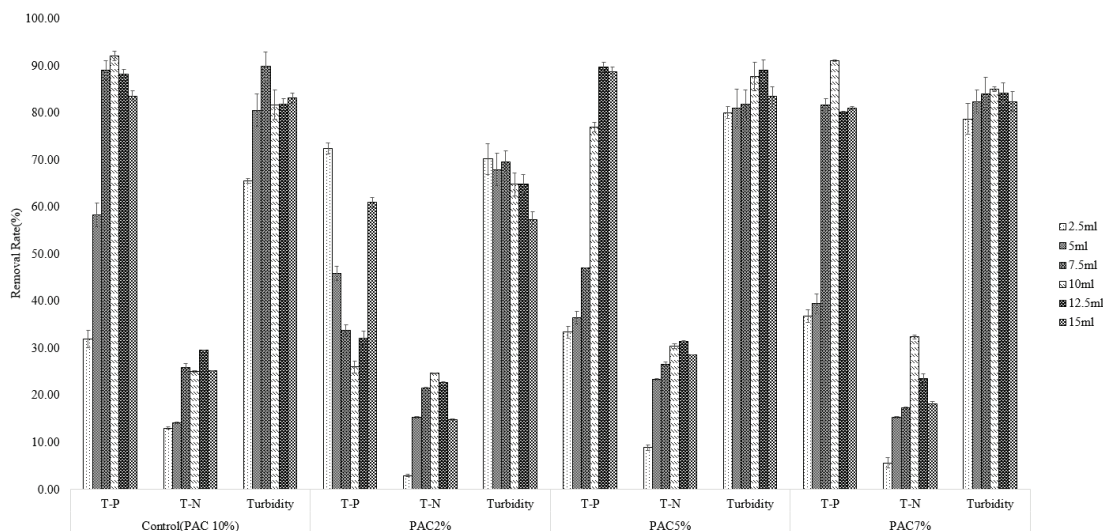


Fig. 2. Comparison of organic matter and turbidity change depending on PAC concentration and input conditions.

phosphorus showing a negative charge due to positive charge of PAC. In the case of turbidity, it can be seen that it shows a high removal efficiency of approximately 91% at maximum, if the injection amount of coagulant is 10 ml.

Furthermore, Jar test was performed to evaluate the coagulant performance. It was confirmed that if Torrefied Wood Flour is included, the flocs are generated quickly. Thus, it was determined that 1% content of Torrefied Wood Flour is the optimal addition at the time of manufacturing the mixed coagulant for removing the organic compounds.

3.3. Result of analyzing coagulation performance based on PAC content

Considering the previously performed organic compound removal efficiency result based on Torrefied Wood Flour content, 1% content of Torrefied Wood Flour was determined to be the optimal addition. Thus, this experiment aimed to analyze the organic compounds removal efficiency based on PAC concentration reduction, in the cases of PAC concentrations of 2%, 5%, 7%, and 10% with respect to 1% addition

of Torrefied Wood Flour. The mixed coagulant removal efficiency based on PAC concentration according to 1% content of Torrefied Wood Flour is shown in Fig. 2. In addition, it also indicates T-P, T-N, and turbidity removal efficiency based on the injection amount of the manufactured mixed coagulant.

As shown in Fig. 2, it can be seen that the organic compound removal efficiency is relatively higher when 1% of Torrefied Wood Flour is included and PAC concentration is 7%. In the case of T-P, it showed a high removal efficiency of maximum of 91% if the injection amount of coagulant is 10 ml. It can confirm that it shows a similar removal efficiency with the coagulation performance result of the mixed coagulant with 1% of Torrefied Wood Flour and 10% PAC concentration. In the case of T-N, it shows approximately 32% of removal efficiency with 10 ml of injection amount of coagulant. This result is presumed to be achieved by a similar cause with the previous experiment result for selecting the optimal content of Torrefied Wood Flour. In the case of turbidity, the maximum removal efficiency of approximately 90% was indicated when PAC concentration

was 7% based on 1% content of Torrefied Wood Flour, and 10 ml injection amount of coagulant. In addition, when 1% of Torrefied Wood Flour is included and PAC concentration was 5%, it shows a relatively less removal efficiency than when PAC concentration was 7%. However, with 12.5 ml injection amount of coagulant, T-P showed 90% of removal efficiency, whereas T-N showed approximately 32%, and turbidity showed 89%.

As a result of performing Jar test per concentration of mixed coagulant with reduced PAC concentration based on 1% content of Torrefied Wood Flour, it was confirmed that reducing PAC concentration by 30%-50% could secure a similar removal efficiency with when the existing PAC (10%) is injected.

3.4. Results of performance comparison of organic material removal of PAC and Torrefied Wood Flour Natural Material Based Coagulant

Based on the previous experiments, it was determined that the optimal removal effect of organic material was achieved at a concentration of 7% of PAC for 1% of Torrefied Wood Flour. An experiment was

conducted to compare and analyze the removal efficiency of organic compounds and PAC (10%) which is a market coagulant, with the final optimal efficiency. The efficiency of T-P, T-N and turbidity removal of the mixed coagulant which has 7% concentration and PAC (10%) coagulant based on 1% Torrefied Wood Flour content is shown in Fig. 3, Fig. 4, Fig. 5.

As shown in the following Fig. 3, it can be seen that Torrefied Wood Flour contains 1% and in the case of mixed coagulant which has PAC concentration of 7% shows T-P removal efficiency similar to PAC (10%). As a result of the previous experiment, the case of T-P showed a high removal efficiency of up to 91% when an injected concentration of coagulant was 10 ppm, and the case of PAC (10%) showed a removal efficiency similar to mixed coagulant with an efficiency of approximately 91% with the same coagulant injection.

The result of T-N removal efficiency according to the samples and input conditions of mixed coagulant and PAC (10%) coagulant is shown in Fig. 4. In the case of mixed coagulant, the removal efficiency was low as approximately 32% when the injection

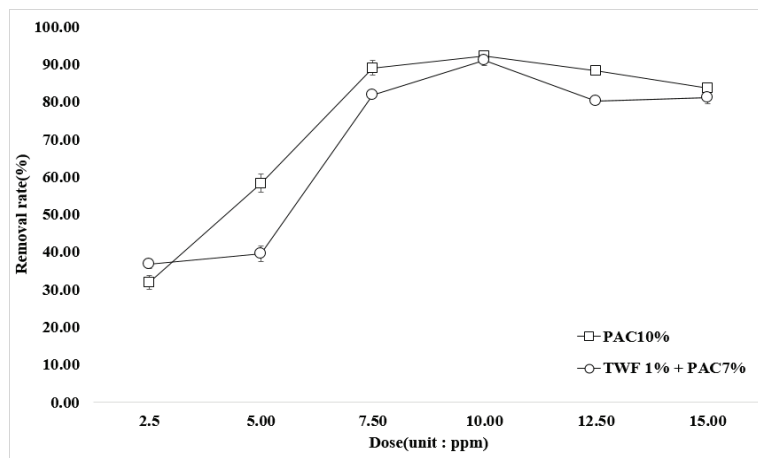


Fig. 3. Comparison of T-P Concentration change depending on samples and input conditions.

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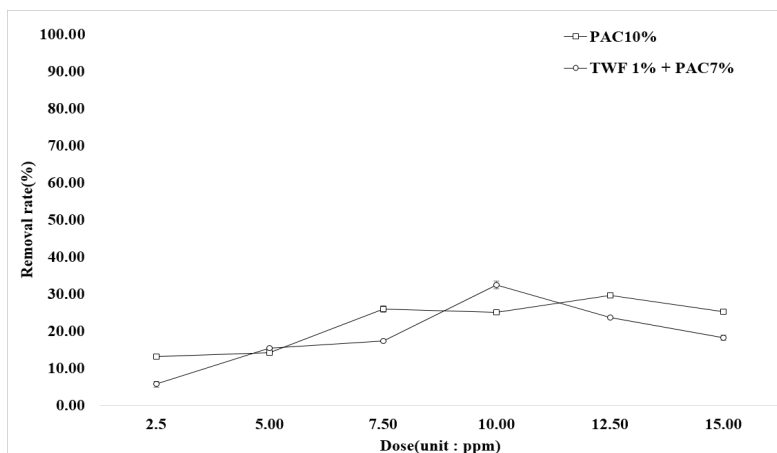


Fig. 4. Comparison of T-N Concentration change depending on samples and input conditions.

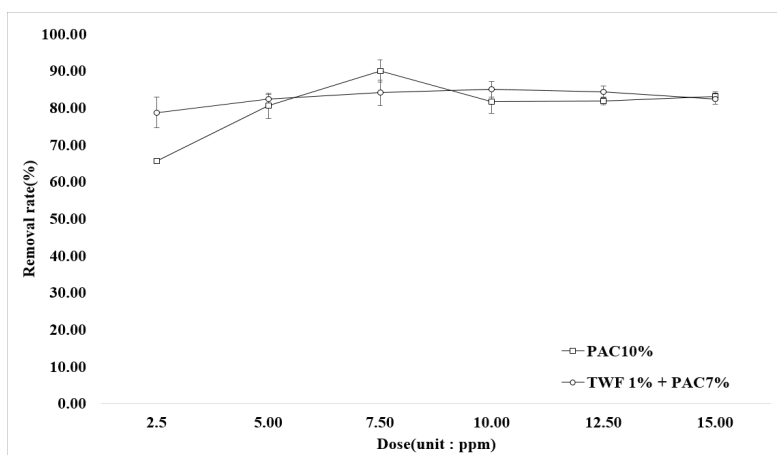


Fig. 5. Comparison of Turbidity Concentration change depending on samples and input conditions.

concentration was 10 ml and in the case of PAC (10%), the removal efficiency was approximately 35% when the injection concentration was 12.5 ml. This is determined to be occurred by a similar cause as the result of the previous experiment. It was judged that the coagulation effect appears to be low due to the low reactivity of the positively charged aluminum ion which is contained in the coagulation, and of the positively charged nitrogen compounds that make up

total nitrogen.

The result of turbidity removal efficiency according to the samples and input conditions of mixed coagulant and PAC (10%) coagulant. In the case of turbidity, the removal efficiency by injection amount was shown to be similar to that of mixed coagulant and PAC (10%), and approximately 90% removal efficiency was shown when mixed coagulant was 10 ml, and approximately 92% removal efficiency when PAC (10%) was 12.5 ml.

3.5. The concentration of aluminum in water

The change in the concentration of aluminum in water according to the treatment conditions of the existing water treatment matter PAC, Torrefied Wood Flour mixed coagulant is shown in Fig. 6. Here, Control means the aluminum concentration of the primary settling pond's outflowing water, and the concentration of aluminum remaining in the water when injecting and disposing of coagulants of TWF 1%+PAC 5%, TWF 1%+PAC 7%, and PAC 10%, respectively. The concentration of aluminum in the water of PAC 10% was approximately 3200 mg/L. It shows that under the condition of TWF 1%+5% PAC, the result was 930 mg/L, and 597 mg/L under the condition of 1% TWF+7% TWF+5% PAC. It shows that compared to PAC 10%, TWF 1%+PAC 7% were reduced by 3 times, and TWF 1%+PAC 5% were reduced by 5 times. In addition, according to concentration dilution, 2,240 mg/L and 1,600 mg/L underwater aluminum concentration shall be indicated. However, the difference from the dilution concentration is determined to be due to the Torrefied Wood Flour micropore which was activated by the carbonation process and the potential by absorption per-

formance expressed.

The results of this study showed that the coagulation process was carried out using the mixed coagulant manufactured according to the optimal Torrefied Wood Flour content derived from the experiment and the appropriate concentration of PAC, resulting in removal efficiency similar to the injection amount of PAC (10%). The use of PACs for the management of total phosphorus is increasing, and according to 2017 sewage statistics, the domestic sewage treatment plants' sewage system supply rate is 94%, which means the number of domestic sewage treatment plants is currently 660. Considering that the annual capacity of the domestic sewage treatment plant is 2,304,381 meters, the cost of injecting coagulant into the domestic sewage treatment plant is approximately 56,270,000,000 KRW per year. If the existing PAC is replaced by TWF 1%+PAC 7% which was developed through this study, it is determined that the existing coagulant injection will be reduced by 30%, which means 16.8 billion won can be secured as benefits of 30%, thereby securing the economic efficiency. Furthermore, the injection of PAC mixed with Torrefied Wood Flour in the primary settling pond can increase the size of floc by the coagulation with the underwater contaminants

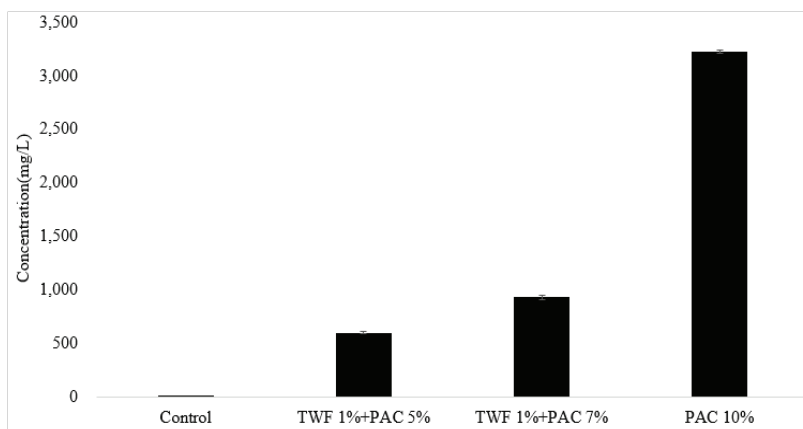


Fig. 6. Comparison of Aluminum Concentration change depending on samples.

by Torrefied Wood Flour. Thus, the sedimentation velocity of underwater suspended solids can be increased. It is judged that due to the reduction of sedimentation time, the time to stay in the primary settling pond in the sewage treatment process could be reduced. As a result, it is concluded that environmental stability can be secured by reducing the concentration of aluminum in the water as the coagulant injection amount increases.

4. CONCLUSION

This study derived the optimal mixing ratio of mixed coagulant manufactured by combining PAC which is a coagulant in market with Torrefied Wood Flour that was overheat steam treated. In order to review the applicability of the mixed coagulant as a water treatment material, in-water organic compounds of T-P, T-N, and turbidity removal efficiency was analyzed when using the mixed coagulant. The followings results were drawn:

- 1) The optimal condition is the 1% addition rate of Torrefied Wood Flour in the market coagulant PAC (10%), and the analysis result of the removal efficiency of organic compound showed that when injecting 10 ml, T-P was up to 92%, T-N was approximately 22%, and turbidity was up to 91%. It was thus confirmed that the mixed coagulant developed in this study was applicable to the treatment of in-water organic compounds in the process of coagulation and blending.
- 2) According to the analysis of removal efficiency of organic compounds based on PAC concentration of 1%, which is the optimal amount of Torrefied Wood Flour, the removal efficiency of T-P 91%, T-N 32%, and the turbidity up to 90% were shown when 10 ml of 7% PAC concentration was injected.
- 3) PAC (10%) and the coagulation performance

comparison experiment allowed the PAC concentration to be reduced by 30% to 50% when the coagulation process was carried out using a mixed coagulant based on 1% of Torrefied Wood Flour manufactured in this study. It has been confirmed that if used as a water treatment material using a mixed coagulant, the removal efficiency can be secured similar to that of the market coagulant PAC (10%).

Therefore, it can be determined that water treatment process by mixing and coagulation using Torrefied Wood Flour mixed coagulant developed in this study will secure economic and environmental effects, as well as removal efficiency similar to the existing coagulant PAC (10%). Although the removal efficiency has been verified, further studies need to be conducted on the size of floc and the principle of the substance removal according to the addition of Torrefied Wood Flour.

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APPENDIX

(Korean Version)

용존성 유기물질 및 탁도 제거를 위한 반탄화목분 천연재료 혼합응집제의 적용성에 관한 연구

초록 : 급격한 산업화로 인한 이상기후의 발생으로 매년 수질관리 중요성과 관리비용이 증대되고 있는 실정이다. 이에 국내의 경우 수질오염의 주요 원인물질인 총인, 총질소의 관리를 위해 하수처리장에서 응집제를 투입하여 유기물질을 처리하고 있다. 그러나 PAC(Poly Aluminium Chloride)의 경우 응집제 과다 주입 시 2차 오염문제가 발생될 수 있다. 이에 국내에서는 천연재료를 혼합한 응집제를 이용한 응집제의 적용성에 관한 연구가 진행되고 있다. 따라서 본 연구에서는 하수처리장의 1차 침전지 유출수를 대상으로 PAC와 반탄화목분의 최적 혼합비율을 도출하기 위해 수질 오염물질인 T-P, T-N 및 탁도를 분석하여 개발한 혼합응집제의 적용성 평가를 하고자 한다. PAC(10%)와 반탄화목분 함유량이 1%가 되는 조건에서 T-P의 경우 최대 92%의 제거효율이 나타났으며, T-N의 경우 약 22%의 제거효율이 나타났다. 이는 혼합응집제와 동일한 양전하성 유기물질이 다수 포함된 T-N의 제거가 잘 이루어지지 않은 것으로 판단된다. 탁도는 약 91%의 제거효율이 나타났다. 이에 반탄화목분 1%가 최적 첨가량이라고 판단하였으며, PAC 농도를 저감하여 유기물질 제거효율을 분석한 결과, PAC 농도 7%에서 T-P의 경우 최대 91%의 높은 제거효율이 나타났으며, T-N의 경우 32%의 제거효율이 나타났다. 탁도의 경우 최대 90%의 제거효율이 나타났다. 또한, PAC(10%)와 응집성능 비교실험을 수행하여 본 연구에서 제조한 반탄화목분 1% 함유량 기준에서 혼합응집제를 이용하여 응집공정을 수행할 경우 PAC 농도를 30-50%로 저감할 수 있으며, 시판 응집제와 유사한 성능을 확보할 수 있을 뿐만 아니라 PAC 주입량이 저감되어 경제적 효과를 확보하고, 2차 오염 문제를 저감하는 안정적인 수처리를 수행할 수 있을 것이라고 사료된다.

1. 서 론

급격한 산업화와 이로 인한 이상기후의 발생으로 매년 취수원 및 하천의 수질관리의 중요성과 관리비용이 증대되고 있다(Hwang et al., 2018). 이에 국내의 경우 수질오염의 주요 원인물질인 총인, 총질소의 관리를 위해 하수처리장에서 응집제 투입으로 수중 용존성 유기물질을 혼화-응집-침전공정을 통해 처리되고 있다. 국내 응집제는 PAC의 사용량이 58%를 차지하고 있다. PAC의 경우 품질기준상 불순물(중금속) 함량이 매우 높음에도 불구하고, 기준 강도가 제대로 이루어지지 않아 응집제 과다 주입 시 잔류 알루미늄 농도의 증가로 인해 2차 오염에 대한 문제가 대두되고 있다(Park, 2009). 기존의 하수종말처리장에서는 유입수질에 따라 다르지만 일반적으로 17% PAC 응집제를 1ppm씩 연속식으로 주입하여 처리하고 있으며 유입수의 총인 저감을 위하여 최대 50ppm까지 주입되고 있다. 투입되는 PAC와 같은 알루미늄 계열의 응집제가 과다 투입된 처리수의 경우 수중 용존 Aluminium 이온이 식수로 유입될 가능성이 높으며, 용존된 Aluminium 이온이 사람에게 유입될 경우 알츠하이머 등과 같은 신경계 질환을 유발할 수 있다(Han et al., 2016). 또한 국내 환경약품 시장은 2000년 중반 이후 해양투기 금지, 오염 총량제 관리, 하수처리장의 고도처리 및 총인처리시설 설치, 에너지자립화사업 등 급격한 환경 법규 변화에 의해 응집제 적용 기술 및 제품의 변화와 오염 부하량에 대한 기술적 관리의 중요성이 강조되고 있다(MOE, 2017). 바이오매스는 자원순환 측면에서 재생가능한 친환경적인 자원으로 알려져 있다(Gong et al., 2016). 이에 국내에서는 식물성 및 광물성 물질 등의 천연재료를 혼합한 응집제를 이용한 응집제의 적용성에 관한 연구가 진행되고 있다(Nam et al., 2016). 대표적인 천연재료로 열분해 과정을 거치는 것으로 바이오매스(목재 등)가 연료로 사용될 때 전처리 과정으로 반탄화 과정을 거치면, 고정탄소의 증가로 발열량이 증가하고, 친수성 작용기들의 열분해로 인해 소수성을 띄어 고유수분함량이 줄어들게 된다(Jung et al., 2018; Priadi et al., 2019; Chang et al., 2019; Kim et al., 2019; Kim et al., 2018). 또한, 반탄화는 무산소 또는 질소 환경에서 200-300°C의 상대적으로 낮은 온도의 열을 가하여 목질계 바이오매스 내 존재하는 수분과 저열량의 산 성분 및 헤미셀룰로오스 함량을 낮춰 에너지 밀도를 증가시키는 열처리방법이다(Lee et al., 2016; Nam et al., 2018).

이에 본 연구에서는 기 수행된 연구를 기반으로 산림 부산물인 병충해 피해목을 이용하여 고열처리조건에서 제조된 반탄화목분의 전하, 흡착성능에 대한 연구(Yang et al., 2016)와 열처리에 의한 강한 음전하를 띄는 반탄화목분과 양이온성 Polyacrylamide를

혼합하여 제조된 paste상 응집제의 녹조제거효율에 대한 연구(Yang et al., 2017)가 진행되었다. 선행연구는 반탄화목분에 양이온성 고분자 응집제를 혼합하여 표면개질처리를 통해 양이온성을 띠는 혼합 응집제로 제조하여, 음이온성 유기물질 T-P 등 수질오염을 유발하는 유기물질을 제거하는 원리에서 착안하여 수중 오염물질에 대한 연구이다(Yang et al., 2019).

따라서 본 연구에서는 기존의 선행연구에서 사용된 반탄화목분과 기존의 하수종말처리장에서 사용되는 PAC의 사용량을 저감하고자 반탄화목분이 혼합된 PAC 응집제를 이용하여 혼합응집제 농도에 따른 수중 오염물질 제거효과를 분석하여 혼합응집제의 적용 가능성을 평가하고자 한다. 평가항목으로는 탁도, 총인, 총질소를 평가하고자 한다. 평가를 통하여 도출된 결과를 통하여 혼합응집제의 최적 혼합비율과 수중 오염물질 제거 가능성에 대해서 검증하고자 한다.

2. 재료 및 방법

2.1. 공시재료

2.1.1. 반탄화목분(Torrefied Wood Flour, TWF)

공시재료로 사용된 반탄화목분은 기 수행된 연구와 유사하게 함수율 12%의 펄프용 참나무 칩을 사용하였으며, 구미소재의 S사에 의뢰하여 과열 증기를 이용하여 250°C에서 25분간 고온 급속 열처리한 반탄화 칩을 분양받아 사용하였다. 과열증기처리한 반탄화 칩은 입자의 균일성을 확보하기 위하여 실험실용 소형 고속분쇄기(Wonder Blender)를 이용하여 분쇄를 하였다. 분쇄된 반탄화목분은 입자크기에 따른 분급시 최대 구성인 75 μm ~ 106 μm 크기 반탄화목분(Zeta potential -40 mV 이상)으로 제조하여 사용하였다(Yang et al., 2019).

2.1.2. Poly Aluminum Chloride(이하 PAC)

PAC는 무기 고분자 응집제의 하나로 화학식 $[\text{Al}(\text{OH})\text{Cl}_6\text{-m}]_n$ 로 구성되어 있으며, 염산과 알루미늄으로 만든 폴리머 계열의 응집제이다. 무색 내지 담황색의 투명한 액상 응집제를 사용하였다. PAC는 양이온성 전하를 가지고 있는 염기성 다핵착이온의 구조를 갖는 무기고분자 화합물로 강력한 응집작용을 나타내는 수처리용 응집제이다(Choi et al., 2016). 반탄화목분의 혼합응집제를 제조하기 위해 보편적으로 사용되는 응집제 PAC를 사용하여 혼합응집제를 제조하였다.

2.1.3. 반탄화목분 함유량에 따른 응집성능 분석을 위한 혼합응집제 제조

본 연구에서는 일차적으로 반탄화목분 혼합응집제 제조 시 반탄화목분의 최적함유량을 도출하기 위해 앞서 과열증기처리를 통해 분쇄한 반탄화목분과 시판 응집제인 PAC를 일정농도로 희석하여 혼합한 혼합응집제를 제조하였다. 혼합응집제는 1차로 시판응집제 PAC(10%)에 반탄화목분 0.5%, 1%, 5%, 10%의 함유량으로 혼합비를 설정하여 아래 Table 1에 나타난 바와 같이 제조하였고, 500 ml PAC(10%)에 반탄화목분을 첨가하여 800 rpm에서 약 5시간 교반을 통해 제조하여 본 실험의 공시재료로 사용하였다(Yang et al., 2016).

2.1.4. PAC농도에 따른 응집성능 분석을 위한 혼합응집제 제조

본 연구에서는 앞서 반탄화목분의 최적함유량을 도출 실험을 통해 유기물질 제거효율 결과를 고려하여 반탄화목분 함유량이 1%가 최적 첨가량이라고 판단하였고, 이차적으로 PAC의 함유량을 저감하여 경제성 및 환경적 안정성을 확보하기 위해 본 실험에서는 반탄화목분 1%첨가량을 기준으로 PAC 2%, 5%, 7%, 10%의 농도로 희석하여 800 rpm에서 약 5시간 교반을 통해 아래 Table 2에 나타난 바와 같이 제조하여 본 실험의 공시재료로 사용하였다.

2.2. 실험방법

Table 1과 Table 2에 나타난 바와 같이 제조한 각 혼합응집제의 조건에 따른 유기물질 제거효율 평가를 위해 수중 유기물질 함유량이 높은 대전하수처리장 1차침전지 유출수를 대상으로 채수하여 사용하였다. 채수된 1차 침전지 유출수는 유기물질 제거효율 평가를 위해 1L비커에 부피플라스크를 이용하여 1L씩 분할하였고, 혼합응집제 조건에 따라 Jar test를 수행하였다. Jar test 운전 조건은 급속교반 140 rpm 2 min, 완속교반 40 rpm 20 min, 침전시간 30 min을 거쳐 상등액을 채취하였다. 상등액 채취 시 45 μm 공극을 가진 실린지 필터를 통해 여과시킨 후 분석시료로 사용하였다. 분석항목은 「수질오염공정시험기준(제 2017-4호)」 중 일반항목에 해당하는 탁도와 이온류에 해당하는 총인, 총질소에 대해 측정하였다.

2.2.1. 탁도(Turbidity)

A Study on the Applicability of Torrefied Wood Flour Natural Material Based Coagulant to Removal of Dissolved Organic Matter and Turbidity

1차 침전지 유출수와 처리수의 탁도를 평가하기 위해 「수질오염공정시험기준(제 2017-4호)」의 탁도 분석기준에 의거하여 광원부와 광전자식 검출기를 갖추고 있으며, 최저검출한계가 0.02NTU 이상인 NTU(Nephelometric Turbidity Units) 탁도계로서 광원인 텅스텐필라멘트가 2,200 K-3,000 K 온도에서 작동하고 측정튜브내의 투사광과 산란광의 총 통과거리가 10 cm가 넘지 않으며, 검출기에 의해 빛을 흡수하는 각도가 투사광에 대해 (90±30)°C를 넘지 않는 탁도계(Turbidimeter TB 210 IR, Lovibond, Germany)를 이용하여 측정하였으며, 이때 반복수는 5회이다.

2.2.2. 총인(T-P)

1차 침전지 유출수와 처리수의 총인을 평가하기 위해 「수질오염공정시험기준(제 2017-4호)」의 총인의 경우 아스코빈산 환원법 등을 기본으로 하는 측정기를 이용하여 수질분석기기(HS-3300, HUMAS)를 이용하여 측정하였으며, 이때 반복수는 5회이다.

2.2.3. 총질소(T-N)

1차 침전지 유출수와 처리수의 총인을 평가하기 위해 「수질오염공정시험기준(제 2017-4호)」의 총질소의 경우 크로마토그래피 기법에 의한 흡광도 측정을 기본으로 하는 측정기를 이용하여 수질분석기기(HS-3300, HUMAS)를 이용하여 측정하였으며, 이때 반복수는 5회이다.

2.2.4. 알루미늄 농도

1차 침전지 유출수와 처리수의 알루미늄 농도 평가를 위하여 「수질오염공정시험기준(제 2017-4호)」의 중급속류 중 알루미늄을 분석하였으며, 분석은 FITI 시험연구원에 시료분석 의뢰하였으며, 유도결합플라즈마/원자발광분광법에 따라 3회 반복하여 측정하였다.

3. 결과 및 고찰

3.1. 대전하수처리장 1차침전지 유출수 성상

대전하수처리장 1차침전지 유출수에서 채취한 오염수를 이용하여 수중 유기물질의 제거효과와 탁도 제거효과 평가를 위해 오염수와 처리수 간의 탁도, 총인, 총질소 항목을 측정하였다. 대전하수처리장 1차침전지 유출수의 수질분석 결과는 다음 Table 3과 같다. 탁도의 경우 하천의 방류수질기준 중 5등급 기준인 SS(부유물질) 기준에 해당되는 탁도 15 NTU에 적합하지 않은 것으로 나타났다(MOE, 2013). T-P와 T-N의 경우 하천방류수질기준 중 5등급 기준인 0.15 mg/L를 초과하는 것으로 나타났다.

3.2. 반탄화목분 함유량에 따른 응집성능 분석결과

수중 오염을 유발하는 유기물질 중 대표적인 유기물질은 T-P와 T-N이 존재하며, 해당 유기물질의 농도가 높을 경우 유기물질로 인한 부영양화 또는 수중 미생물의 성장을 촉진하여, 미생물로 인한 독성물질 유출로 인해 수질오염이 심각해진다. 아래 Fig. 1은 시판 응집제인 PAC(10%)에 반탄화목분의 함유량에 따른 유기물질 제거효율에 대해 나타난 것이다. 반탄화목분의 함유량은 기 수행된 선행연구를 참고하여 0.5%, 1%, 2%, 5%의 함유량으로 선정하여 혼합응집제를 제조하였으며, 제조한 혼합응집제의 주입량에 따른 T-P, T-N, 탁도의 제거효율을 나타내었다.

다음 Fig. 1에 나타난 바와 같이 반탄화목분이 1% 포함된 혼합응집제의 유기물질 제거효율이 상대적으로 높은 결과를 나타냈다. T-P의 경우 Control인 PAC 10% 응집제 10 ml에서 최대 약 92%의 제거효율, T-N의 경우 TWF 1%+PAC 7% 응집제 10 ml에서 약 22%의 제거효율을 나타내는데 이는 총 질소를 구성하는 유기물질이 양전하성을 나타내며 상대적으로 PAC의 양전하성에 의해 음전하성을 띠는 총인의 유기물질과 달리 상대적인 응집제 제거 효과가 낮게 나타난 것으로 판단된다. 탁도의 경우 전체 조건 혼합응집제 조건에서 응집제 주입량 10 ml일 경우 최대 약 91%의 높은 제거효율을 나타낸 것을 알 수 있다.

또한, 응집제 성능평가를 위해 Jar test를 수행하였는데, 반탄화목분이 함유될 경우 플러의 크기와 형성이 빠르게 생성되는 것을 확인할 수 있었다. 이에 반탄화목분을 첨가한 혼합응집제 제조 시 반탄화목분 함유량이 1%일 경우 최적 첨가량으로 유기물질 제거에 효과적이라고 판단하였다.

3.3. PAC 함유량에 따른 응집성능 분석결과

앞서 수행한 반탄화목분 함유량에 따른 유기물질 제거효율 결과를 고려하여 반탄화목분 함유량이 1%가 최적 첨가량이라고 판단하였고, 이에 본 실험에서는 반탄화목분 1% 첨가량에 대해 PAC 농도 2%, 5%, 7%, 10%일 경우 PAC 농도 감소에 따른 유기물질 제거효과에 대해 분석하고자 한다. 아래 Fig. 2는 반탄화목분의 1% 함유량을 기준으로 PAC 농도에 따른 혼합 응집제 제거효율에 대해 나타낸 것이며, 제조한 혼합응집제의 주입량에 따른 T-P, T-N, 탁도의 제거효율을 나타내었다.

다음 Fig. 2에 나타난 바와 같이 반탄화목분이 1% 포함되고, PAC 농도가 7%일 경우 유기물질 제거효율이 상대적으로 높은 것을 알 수 있으며, T-P의 경우 응집제 주입량이 10 ml 일 경우 최대 91%로 높은 제거효율이 나타났다. 이는 PAC 반탄화목분이 1% 포함되고, PAC 농도가 10%인 혼합응집제의 응집성능 결과와 유사한 제거효율을 나타내는 것을 알 수 있다. T-N의 경우 응집제 주입량 10 ml 약 32%의 제거율을 나타내는데 이는 앞서 수행된 반탄화목분의 최적 함유량 선정 실험결과와 유사한 원인에 의한 것으로 판단된다. 탁도의 경우 반탄화목분 1% 함유량 기준 PAC 농도가 7%일 경우 응집제 주입량 10 ml에서 최대 약 90%의 제거효율을 나타냈다. 또한, 반탄화목분이 1% 포함되고, PAC 농도가 5%일 경우 PAC 7%일 경우보다 다소 제거효율이 낮게 나타났지만, 응집제 주입량 12.5 ml에서 T-P의 경우 90%의 제거효율이 나타났고, T-N의 경우 약 32%로 제거효율이 나타났으며, 탁도는 89%의 제거효율이 나타났다. 이에 반탄화목분 1% 함유량 기준에서 PAC 농도가 저감된 혼합 응집제의 농도별 Jar test 실험 결과 PAC 농도를 30%-50%로 저감시켜 기존의 PAC(10%)를 주입할 경우와 유사한 제거효율을 확보할 수 있음을 확인하였다.

3.4. PAC와 반탄화목분 혼합응집제의 유기물질 제거 성능 비교 결과

앞서 수행한 실험을 통해 반탄화목분 1% 첨가량에 대해 PAC 7%의 농도일 경우 최적의 유기물질 제거효과가 나타났다고 판단하였으며, 최종적으로 최적 효율이 나타난 혼합 응집제와 시판 응집제인 PAC(10%)의 유기물질 제거효율을 비교·분석하기 위한 실험을 수행하였다. 아래 Fig. 3, Fig. 4, Fig. 5는 반탄화목분의 1% 함유량을 기준으로 PAC 농도가 7%인 혼합응집제와 PAC(10%) 응집제의 T-P, T-N, 탁도 제거효율을 나타낸 그림이다.

다음 Fig. 3에 나타난 바와 같이 반탄화목분이 1% 포함되고, PAC 농도가 7%인 혼합응집제의 경우 응집제 주입량이 증가함에 따라 PAC(10%)와 유사한 T-P 제거효율을 나타내는 것을 알 수 있다. 앞서 실험을 통해 도출된 결과로 T-P의 경우 응집제 주입농도 10 ppm일 경우 최대 약 91%로 높은 제거효율이 나타났으며, PAC(10%)의 경우 동일한 응집제 주입량에서 약 91%의 제거효율로 혼합응집제와 유사한 제거효율이 나타났다.

Fig. 4는 혼합응집제와 PAC(10%) 응집제 주입량에 따른 T-N 제거효율을 나타낸 그래프이다. 혼합응집제의 경우 주입량 10 ml에서 약 32%로 낮은 제거효율이 나타났고, PAC(10%)는 주입량 12.5 ml에서 약 35%의 제거효율이 나타났다. 이는 앞서 도출된 실험결과와 유사한 원인에 의한 것으로 판단된다. 응집제에 포함되어 있는 양전하성 알루미늄이온과 총질소를 구성하는 양전하성 질소화합물의 반응성이 낮아 응집효과가 낮게 나타난 것으로 판단된다.

Fig. 5는 혼합응집제와 PAC(10%) 응집제 주입량에 따른 탁도 제거효율을 나타낸 그래프이다. 탁도의 경우 주입량에 따른 제거효율이 혼합응집제와 PAC(10%) 유사한 경향으로 나타났으며, 혼합응집제의 경우 10 ml에서 약 90%의 제거효율이 나타났으며, PAC(10%)의 경우 12.5 ml에서 약 92%의 제거효율이 나타났다.

3.5. 수중 알루미늄 농도

기존 수처리제인 PAC, 반탄화목분 혼합응집제 처리 조건에 따른 수중의 알루미늄 농도 변화는 Fig. 6과 같으며, Control은 1차 침전지 유출수의 알루미늄 농도를 의미하며, TWF 1%+PAC 5%, TWF 1%+PAC 7%, PAC 10% 응집제를 각각 10 ppm 주입 및 처리 시 수중에 잔류하는 알루미늄 농도를 의미한다. PAC 10%의 수중 알루미늄 농도는 약 3200 mg/L이며, TWF 1%+PAC 7% 조건에서는 930 mg/L, TWF 1%+PAC 5% 조건에서는 597 mg/L의 수중 알루미늄 농도 결과를 나타냈다. PAC 10% 대비 TWF 1%+PAC 7%는 3배 저감, TWF 1%+PAC 5% 5.5배 저감된 결과를 나타냈으며, 농도 희석에 따르면 2,240 mg/L, 1,600 mg/L의 수중 알루미늄 농도를 나타내야하지만 희석 농도와와의 차이는 탄화공정에 의해 활성화된 반탄화목분 미세공극 및 전위에 의해 발현된 흡착성능에 의한 것으로 판단된다.

이러한 결과로 본 연구에서 실험을 통해 도출한 최적 반탄화목분 함유량과 PAC 적정 농도에 따라 제조한 혼합응집제를 이용하여 응집공정을 수행할 경우 PAC(10%) 주입량과 유사한 제거효율을 확보할 수 있었다. 총인처리를 위하여 점차 증가하는 PAC 사용량이 증가되고 있으며 2017년 하수도 통계 기준 국내 하수처리장의 하수도 보급률은 94%로 현재 국내 하수처리장은 660개소에 달하고 있다. 전국의 연간 하수처리장 처리용량은 20,304,381m³인 것을 고려한다면, 연간 국내 하수처리장에서

A Study on the Applicability of Torrefied Wood Flour Natural Material Based Coagulant to Removal of Dissolved Organic Matter and Turbidity

응집제 투입량에 따른 소요비용은 약 56,027 백만원에 달하는 막대한 비용이 매년 소요되고 있다. 기존 PAC에서 본 연구에서 개발된 TWF 1% + PAC 7%로 대체할 경우 기존 응집제 투입량을 30%로 절감시켜 30%의 편익으로 168억원을 확보하여 경제적 효율성을 확보할 수 있을 것으로 판단된다. 경제적 효과를 확보할 수 있을 뿐만 아니라 1차 침전지에서 반탄화목분이 혼합된 PAC를 투입함으로써 반탄화목분에 의하여 수중 오염물질과의 응집에 의하여 floc의 크기를 증가시켜 수중 부유물질의 침강속도를 증가시킬 수 있다. 침전 시간의 감소로 하수처리공정의 1차침전지 체류시간을 감소시킬 수 있을 것으로 판단된다. 이로 인해 응집제 투입량이 증가함에 따라 높아지는 수중 알루미늄 농도 저감을 통해 환경적 안정성을 확보할 수 있을 것으로 판단된다.

4. 결론

본 연구에서는 시판 응집제인 PAC와 과열증기처리한 반탄화목분을 혼합하여 제조한 혼합응집제의 최적 혼합비율을 도출하였으며, 혼합응집제의 수처리제로써의 적용성을 검토하기 위해 혼합응집제 사용 시 수중 유기물질인 T-P, T-N 및 탁도 제거효과에 대해 분석하였으며, 다음과 같은 결론을 도출하였다.

- 1) 시판 응집제인 PAC(10%)에 반탄화목분 혼합 비율 중 1% 첨가비율이 최적조건이며, 이때의 유기물질 제거효율 분석 결과 10ml 주입시 유기물 제거효율은 T-P 최대 92%, T-N 약 22%, 탁도 최대 91% 결과를 나타냈다. 이에 본 연구에서 개발한 혼합응집제가 화학적 응집·혼화 처리공정에서 수중 유기물질 처리에 적용 가능함을 확인하였다.
- 2) 최적의 반탄화목분 투입량인 1%를 기준으로 PAC 농도에 따라 유기물질 제거효율 분석 결과 PAC 농도 7% 10ml 주입시 T-P 91%, T-N 32%, 탁도 최대 90%의 제거효율이 나타났다.
- 3) PAC(10%)와 응집성능 비교 실험을 통해 본 연구에서 제조한 반탄화목분 1% 함유량 기준에서 혼합응집제를 이용하여 응집공정을 수행할 경우 PAC 농도를 30%-50%로 저감할 수 있으며, 혼합응집제를 이용하여 수처리제로 사용할 경우 시판 응집제인 PAC(10%)를 사용할 경우와 유사한 제거효율을 확보할 수 있음을 확인하였다.

따라서, 본 연구에서 개발한 반탄화목분 혼합응집제를 이용하여 혼화-응집 공정의 수처리를 수행할 경우 기존의 응집제인 PAC(10%)와 유사한 제거효율 확보는 물론 경제 및 환경적 효과도 확보할 수 있을 것으로 판단된다. 제거효율에 대하여 검증하였으나 반탄화목분 첨가에 따른 floc의 크기 및 물질제거 원리 등에 대한 추가적인 연구가 필요할 것으로 판단된다.