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Improved mixing of hydrogen peroxide injection in advanced oxidation process treatment using computational fluid dynamics

Pyonghwa Jang[a], Hyunjun Kim[a], Gyusun Kyung[b], Heekyong Oh[c,*]

[a]*R&D Center, OCI, Seongnam si 13212, Korea*
[b]*Seoul Water Research Institute, Seoul 04981, Korea*
[c]*Engineering Solution Team, DAEWOO E&C, Seoul 04548, Korea, Tel. +82-2-2288-1263; Fax: +82-2-2288-4096; email: heekyong.oh@daewooenc.com*

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ABSTRACT

The main issues with ozone-based advanced oxidation processes (AOPs) occur in the type of poor mixing, residual oxidizing agents, inadequate concentration control, and by-product formation. This study focused on improving the mixing effect of hydrogen peroxide by slightly modifying the existing injection facility by performing computational fluid dynamics (CFD) on the existing hydrogen peroxide injection system of the ozone-based AOP. CFD analysis showed that conventional diffuser systems were inefficient for mixing oxidizer and feed water. The mixing ratio was low since the momentum of the feed water could not be overcome by the hydrogen peroxide injection flow, and the length of the contact pipeline was not sufficient for the oxidizer to mix with the feed water. The problem could be overcome by increasing the number of diffusers to increase the injection flow of hydrogen peroxide. This study excluded dissociation and reaction between hydrogen peroxide and contaminants or scavengers, so further studies are needed.

Keywords: Advanced oxidation process; Computational fluid dynamics; Diffuser; Dispersion; Mixing; Hydrogen peroxide

1. Introduction

Advanced oxidation processes (AOPs) are considered to be attractive oxidation technologies to degrade natural organic matter, disinfection by-products (DBPs) precursors, and other non-degradable micropollutants and to disinfect pathogens [1–3]. Theoretically, AOPs can mineralize organic compounds to carbon dioxide and water, according to Eq. (1). AOPs have many options to select and combine oxidants such as ozone, hydrogen peroxide, photocatalyst, and electron beam to produce highly reactive $^{\bullet}$OH radicals [4–6].

$$R-H+OH^{\bullet}\rightarrow H_2O+R^{\bullet} \qquad (1)$$

AOPs used in a drinking water treatment plant can be classified into four groups: ozone-based processes (O_3/high pH, O_3/UV, O_3/H_2O_2, O_3/Fe(II), O_3/TiO_2, metal oxide catalyst, O_3/ultrasound) [7]; photocatalytic process (H_2O_2/UV, O_3/UV/H_2O_2, TiO_2/UV, H_2O_2/TiO_2/UV, O_3/TiO_2/UV) [8]; the Fenton reaction-based processes (Fe^{2+}/H_2O_2, $Fe^{2+}/H_2O_2//H_2O_2$) [9]; other processes which include ionizing radiation, electron beam technology [10]. Among them, ozone has been widely applied in water treatment since it is a strongly reactive and unstable allotrope of oxygen.

In Korea, the ozone-based AOP and ozone oxidation process are generally combined with the subsequent

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* Corresponding author.

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adsorption or bio-adsorption processes in drinking water treatments as shown in Table 1 [11,12]. The application of advanced water treatment in Korea's drinking water treatment plants is more than 43%, and granular activated carbon (GAC) or BAC are recommended to be positioned at the final process before post-disinfection. GAC processes have combined with ozone oxidation for improving bio-adsorption of macro organic compounds [13]. Eventually, a GAC turns into a BAC with both adsorption and biodegradation functions over operation times. Recently, GAC sandwich slow sand filtration (named FA – filtration adsorption in Korea), has been applied for removing taste and odor-causing compounds (2-MIB, Geosmin), DBP precursors, and emerging concern compounds [14,15]. Besides, only ozone oxidation before the BAC process was enhanced with other oxidants with UV or hydrogen peroxide to oxidize the emerging micropollutants.

Ozone was dosed with fine diffusers in the ozone contactors with 15~20 min of hydraulic retention time when ozone was first introduced into a water treatment plant. Recently, more than 70% of the ozone dose system has been injected using the side stream or full stream method into the feed pipeline linked to the ozone contact basin [16]. Also, most of the ozone retrofit project includes the injection system. The ozone dose was decided based on total organic carbon (TOC), T&O causing compounds, and water temperature. When ozone is used only for oxidants, ozone dose is controlled between 2 and 3 mg/L, and ozone dose is decreased by 25%~50% when combined with the other oxidant. AOP can be designed for the first time or modified to the existing ozone system when the non-degradable contaminants are contained in the raw water or cannot be controlled by the existing treatment process.

Oxidizing agents used for ozone-based AOP of a water treatment plant should be selected in consideration of target compound removal, mixing efficiency, work safety, the convenience of O&M, and economics. The main issues in the AOP operation using an oxidizing agent are poor mixing, residual oxidizer, inadequate concentration control, and by-product formation, which might be resulted from the limitations in scaling up to actual facilities and lower operation rate less than design capacity [17]. The success of the AOP operation depends on the activation of the agent in water. Oxidation of contaminants by ozone has two pathways; direct oxidation and indirect oxidation for target microcontaminants shown in Fig. 1. $^{\bullet}$OH initiates ozone decomposition, and the initiation of ozone decomposition can be accelerated by adding hydrogen peroxide. This reaction is fast and important particularly when the $^{\bullet}$OH scavenger concentration is low in the water. It leads to the consumption of ozone and $^{\bullet}$OH, lowering the oxidation capacity of the system. A low concentration of hydrogen peroxide tends to be inactive, however, it can be a scavenger in high concentration to ozone. It is important to control the proper concentration of agents in treatment water. For that reason, rapid mixing and accurate injection are required to secure instantaneous contact between oxidizing agents and organics in water.

There have been many studies that evaluate the removal performance of contaminant using a laboratory experiment. They are indirect and qualitative approaches but they do not reflect actual conditions and give quantitive data such as mixing time or uniformity of agent. Moreover, the operating plant has a huge scale and the real testing experiment itself is risky and extensive. For that reason, the numerical experiment can be an alternative to predict mixing, mass transfer and contaminant removal in the oxidation process is fundamental for process development and scale-up [18]. Computational fluid dynamics is one of the parts of fluid mechanics and calculating numerical methods are solving

Table 1
Combination processes of advanced water treatment system in Korea

Water resource	Combination processes	Capacity (m^3/d)	Ozone dosage method
Han River	S/F → O_3 → BAC	1,950,000	2 mg/L, injector
	S/F → H_2O_2/O_3 → BAC	2,100,000	1.5 mg/L, injector
	MF → H_2O_2/O_3 → BAC	50,000	1.5 mg/L, injector
Paldang Dam	S/F → O_3 → BAC	1,496,000	2~3 mg/L, injector
	S/F → O_3 → FA	530,000	2~3 mg/L, injector
	S/F-UV/O_3 → FA	110,000	
	S/F → GAC	145,000	
Nakdong River	O_3 → S/F → O_3 → BAC	4,208,000	2~3 mg/L, diffuser
	S/F → O_3 → BAC	422,000	2~2.5 mg/L, diffuser
	S/F → H_2O_2/O_3 → FA	120,000	3 mg/L, diffuser
	S/F → GAC	70,000	2 mg/L, diffuser
	S/F → H_2O_2/O_3 → BAC	36,000	3 mg/L, diffuser
	MF → GAC	16,000	
Geum River	S/F → GAC	100,000	
	S/F → GAC	28,000	
Nam River and related dam	S/F → O_3 → BAC	433,000	2 mg/L, diffuser
	S/F → GAC	661,000	

S/F: sand filtration; MF: microfiltration; FA: filtration adsorption; GAC: granular activated carbon; sandwich slow sand filtration.

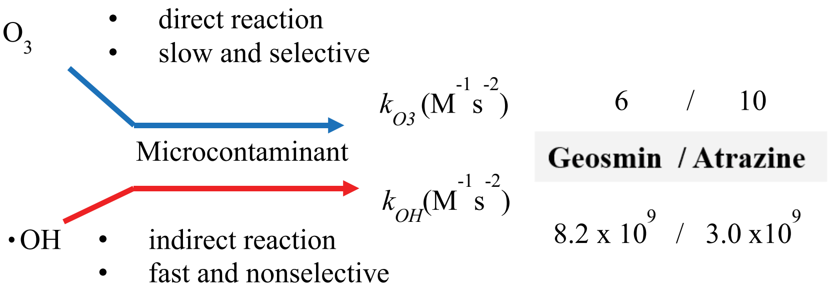

Fig. 1. Oxidation pathways of microcontaminants by ozone.

the various flows of fluid [19]. Two-dimensional models, which is to solve the flow rate of fluid and analyze the mathematical operations. This analysis process can show the effects of temperature and static pressure, velocity on the mixing or dispersion of oxidant before designing and constructing the injection device at the field.

When the operation rate of a water treatment plant decreased less than the initial design capacity, the hydraulic condition of the oxidant injection system can be changed. Therefore, in this study, a computational fluid analysis for the hydrogen peroxide injection system was performed to analyze and improve the existing mixing effect of hydrogen peroxide in the ozone-based AOP process. Finally, the computational fluid dynamics (CFD) simulation of hydrogen peroxide injection could suggest improved mixing approaches for the injection system with minor modification.

2. Material and methods

2.1. Testbed for the AOP

The water treatment plants using the Han River are supplying tap water of advanced water treatment to citizens as shown in Table 1 by 100% of advanced water treatment as shown in Table 1. The testbed where the CFD was performed is a drinking water treatment plant with ozone-based AOP facilities. The process of water purification plant, where the AOP was designed by hydrogen peroxide and ozone. Hydrogen peroxide was injected into the pipeline and then mixed into the side stream system of ozone, causing an immediate mixing reaction in seconds.

Table 2 shows the goal of final water after advanced water treatment combined with AOP and BAC. The main issues of water quality in river water are T&O causing compound and DBP and the concentration of them should be controlled by; less than 8 ng/L of Geosmin and 2-MIB, less than 1.0 mg/L of TOC, and less than 30 μg/L of trihalomethane (THM). Besides, the goal of *Giardia lamblia*, was less than 0.5 log removal at a reservoir because ozone has a role in disinfection.

Table 2
Goal of final water after advanced water treatment at the testbed

Parameters	Goal
Geosmin, 2-MIB	≤ 8 ng/L
Turbidity	≤ 0.08 NTU
Giardia lamblia	0.5 log removal
THM	≤ 30 μg/L
HAA	≤ 10 μg/L
TOC	≤ 1.0 mg/L

THM: trihalomethane; TOC: total organic carbon.

The AOP process was operated by combining ozone and hydrogen peroxide as the primary and secondary oxidizing agents, respectively, shown in Fig. 3. Hydrogen peroxide was diluted with feed water of sand filtrate within a static mixer, then injected into the diffuser. The dilute H_2O_2 solution flew out of the main pipe after contact of 3.5 s after injection in case of a conventional diffuser and it diffused slowly with a parallel streamline.

2.2. CFD analysis procedure

The numerical analysis of the hydrogen peroxide injection and mixing was performed using commercial software ANSYS Fluent Ver. 16.0. The flow transfer of fluid in 3D

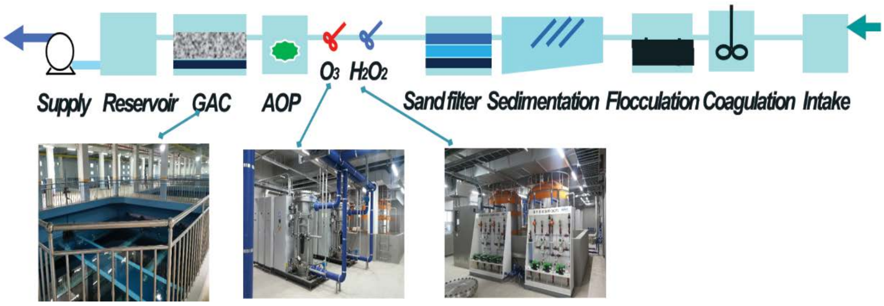

Fig. 2. Process of water treatment plant as a testbed.

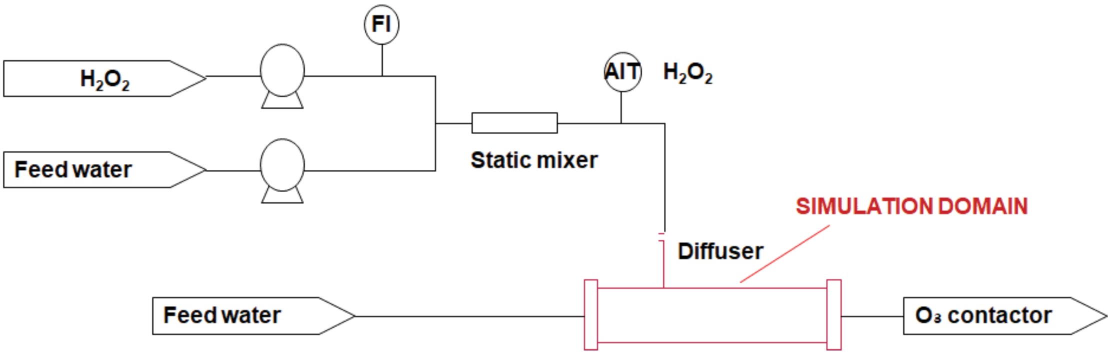

Fig. 3. Diffuser of hydrogen peroxide in the AOP system.

Table 3
Analysis conditions of the AOP system

Items	Specifications
Fluid temperature	25°C
Fluid pressure	1 bar
Feed flow rate of filtrate	9,375 m^3/h
Flow rate of H_2O_2	63 m^3/h

space used the realized k–ε turbulence model. It considered the momentum transfer and mass diffusion by turbulence where the mass conservation equation and the momentum conservation equation were used. Meanwhile, a mass of fluid is conserved and the mass conservation equations for simulation are described as followed.

For mass conservation:

$$\frac{\partial \rho}{\partial t} + \nabla\left(\rho \vec{v}\right) = S_m \tag{2}$$

where S_m is the source from the dispersed second phase and to be added to the continuous phase.

For conservation momentum:

$$\frac{\partial}{\partial t}\left(\rho \vec{v}\right) + \nabla\left(\rho \vec{v}\vec{v}\right) = -\nabla P + \nabla\left(\bar{\bar{\tau}}\right) + \rho \vec{g} + \vec{P} \tag{3}$$

where $\bar{\bar{\tau}}$ is stress tensor, P is static pressure, $\rho \vec{g}$ is gravitational body force and $\vec{P}$ is external body forces.

In ANSYS Fluent, realized k–ε model performs the best of all versions of k–ε model from several validations of flow. Therefore, the realized k–ε model has been implemented during this numerical modeling.

For realized k–ε model:

$$\frac{\partial \rho}{\partial t}\left(\rho k\right) + \frac{\partial}{\partial x_i}\left(\rho k u_i\right) = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_i}{\sigma_k}\right)\frac{\partial k}{\partial x_j}\right] + G_k + G_b - \rho\varepsilon - Y_M + S_k \tag{4}$$

$$\frac{\partial}{\partial t}\left(\rho \varepsilon\right) + \frac{\partial}{\partial x_i}\left(\rho k u_i\right) = \frac{\partial}{\partial x_j}\left[\left(\mu + \frac{\mu_i}{\sigma_k}\right)\frac{\partial \varepsilon}{\partial x_j}\right] + C_{1\varepsilon}\frac{\varepsilon}{k}\left(G_k + G_{3\varepsilon}G_b\right) - C_{2\varepsilon}\rho\frac{\varepsilon^2}{k} + S_\varepsilon \tag{5}$$

where G_k is the production of turbulence energy due to mean velocity gradients.

For chemical conservation:

$$\frac{\partial}{\partial t}\left(\rho^q \alpha^q Y_l^q\right) + \nabla\left(\rho^q \alpha^q \vec{v}^q Y_l^q\right) = -\nabla \alpha^q \vec{J}_l^q + \alpha^q S_i^q + \sum_{p=1}^{n}\left(\dot{m}p^i q^j - \dot{m}q^j p^i\right) \tag{6}$$

where p is static pressure, Y_l is the local mass fraction for the species, $\dot{m}q^j p^i$ is mass transfer source between species, α^q is volume fraction for phase q, and S_i^q is the rate of creation by addition from the dispersed.

The distribution and mixing between target substances were determined based on the qualification of species mixing, individually. Therefore, the degree of mixing of the target component was determined as 'uniformity', which is calculated by the volume fraction efficiency of the target at the cross-section, and the closer to 1 means a uniform distribution.

For uniformity:

$$\text{uniformity} = 1 - \frac{\sum_{i=1}^{N}\left[\sqrt{\left(x_i - \bar{x}\right)^2} A_i\right]}{2\bar{x}A} \tag{7}$$

where A_i is surface area at the i-th cell, x_i is variable (fraction or concentration) at the i-th cell, and $\bar{x}$ is the area-weighted variable of surface,

The assumption of analysis is provided as follows.

- Dilute H_2O_2 is the perfect mixture.
- Diluted hydrogen peroxide contains H_2O_2 and feed water of sand filtrate.
- Physical properties of density, molecular weight are based on 25°C, 1 bar condition of pure water.
- Feedwater flow has been distributed at the main solution feed pipe.
- Chemical reactions or heat transfer are not considered.

3. Results and discussions

3.1. Operation conditions of AOP

The dose of oxidizing agents was calculated based on the water quality, especially the target of Geosmin and

2-MIB. The dosing table of ozone and peroxide was recommended based on the seasonal main event; algal booming, water temperature. The hydrogen peroxide concentration was determined at a ratio of 1:0.5 based on the ozone dose concentration, and in the low-temperature season below 5°C, the injection concentration was made to be 1:5 due to the low reactivity of oxidant. Table 4 is a table of basic oxidizer injection for AOP process operation, but the oxidant injection rate could be flexibly adjusted according to the site conditions such as raw water quality and residual oxidant concentration.

The H_2O_2 stock solution used 35% concentration. The injection flow of hydrogen peroxide was 0.12~0.5 L/min based on the dosing conditions and 450,000 m^3/d of design capacity. Since hydrogen peroxide was injected into the two main pipelines linked to the ozone contact basins, two tube pumps were selected and covered 62~311 mL/min flow rater, respectively. Table 5 shows the specifications of the diffuser for injecting the diluted hydrogen peroxide into the main pipe. However, operation flow was 225,000 m^3/d (9,375 m^3/h) at the testbed, therefore the velocity of feed water was reduced by 1.22 m/s, that of the diluted H_2O_2 was reduced by 2.23 m/s (63 m^3/h of flow rate), and the velocity was 15 m/s at the orifice as shown in Table 5. Feedwater which should be treated flows and it would dominate the flow in the solution main pipe. The ratio of flow rate between feed water and dilute H_2O_2 was about 149. It is necessary to inject oxidant with enough pressure into the feed water instantly.

3.2. Analysis of existing hydrogen peroxide injection device

Hydrogen peroxide with a concentration of 35% was diluted with the sand filtrate. The dilute H_2O_2 was injected into the main pipeline at a lower flow rate less than 149 times of feed water as shown in Fig. 5. The feed water dominated the whole movement of fluid as described by the volume fraction of hydrogen peroxide in Fig. 6. It was considered that there were no other disturbance elements for the flow other than the diffuser in the main pipe. The retention time of oxidant flow was about 3.5 s in the main pipeline. The oxidant dispersed after radial injection with the same direction gradually and the streamlines of the diluted H_2O_2 solution showed parallel flow in the main pipeline (Fig. 7).

The volume fraction of dilute H_2O_2 at cross-sections along flow direction is plotted in Fig. 8. The mixture gradually formed a uniform distribution from 0.02 to 0.56 as diffusion toward flow direction occurred. Besides, the path distance of hydrogen peroxide flow was not enough in the main pipeline due to low injection pressure as shown in Fig. 9. Fig. 10 shows there was a poor mixing area between injection points.

In the existing injection device, the distribution range of dilute H_2O_2 fraction was from 0% to 3% at the discharge surface and its uniformity at the outlet of the orifice was

Table 5
Specification of hydrogen peroxide diffusers at the design capacity

Orifice velocity (m/s)	12
Hole size (mm)	21.6
No. of holes	4
Flow rate (m^3/s)	0.018
Head loss (m)	9.06
Head loss (m)	9.06
Diameter of pipe (mm)	100

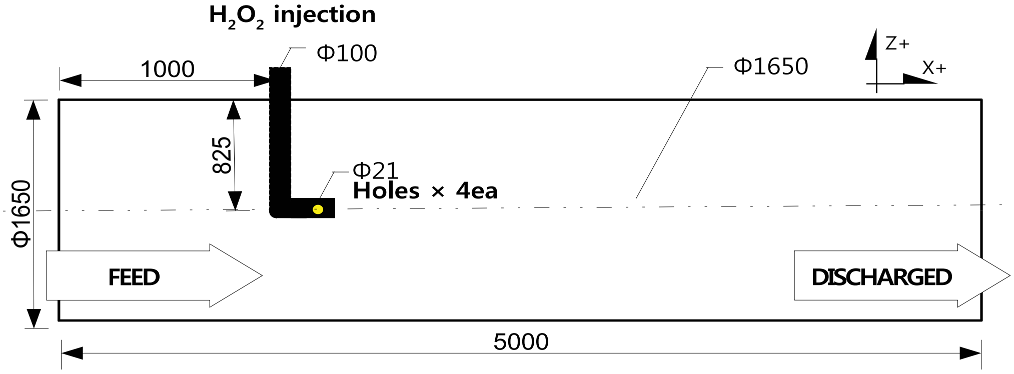

Fig. 4. CFD domain design and conditions.

Table 4
Dosing conditions of oxidizing agents based on the AOP modes

Modes	Oxidizing agents	Dry	Normal	Cold	Emergency
Only ozone	O_3 (mg/L)	1.0~1.5	0.5~1.0	0.1~0.5	1.5~2.0
AOP	O_3 (mg/L)	0.5~0.75	0.25~0.5	0.1~0.3	0.7~1.0
AOP	H_2O_2 (mg/L)	0.25~0.30	0.15~0.25	0.3~0.5	0.35~0.5

Dry: Feb.~May; Normal: June, Oct.~Nov.; Cold: Dec.~Jan.; Emergency: July~Sep.

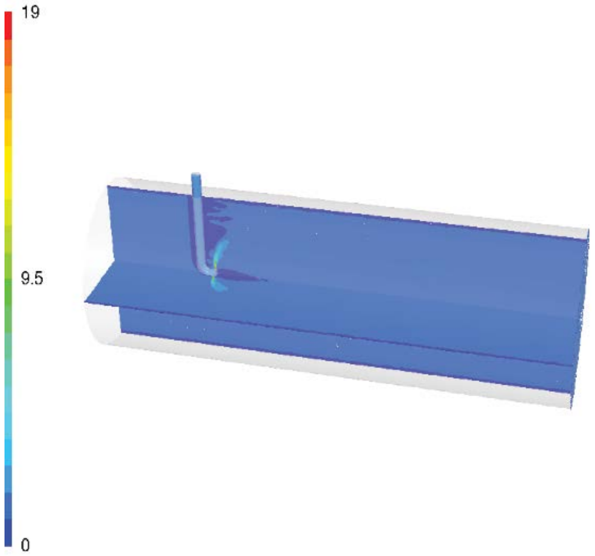

Fig. 5. Velocity magnitude (m/s).

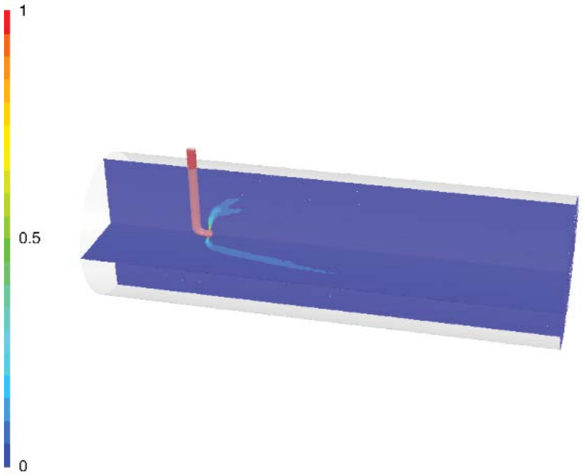

Fig. 6. Volume fraction of H_2O_2 fluid.

calculated by 56%. The volume fraction and the uniformity value of dilute H_2O_2 at cross-sections along flow direction are plotted in Fig. 10.

3.3. Analysis of modified hydrogen peroxide injection device

The injection conditions of the oxidant were examined to enhance the injection pressure or to disperse the oxidizer in the radial direction for atomization and diffusion of the solution. The case studies for modification on diffusers were reviewed to divide the stream of oxidant flow into several separate parts.

The orifice hole number of the diffuser was doubled from 90° interval to 45° interval. The number of holes was doubled, but the individual hole diameter was reduced from Ø 21 mm to Ø 15 mm so that the total hole area is the same as the previous diffuser. That is, the velocity at the orifice hole was kept as the previous design by

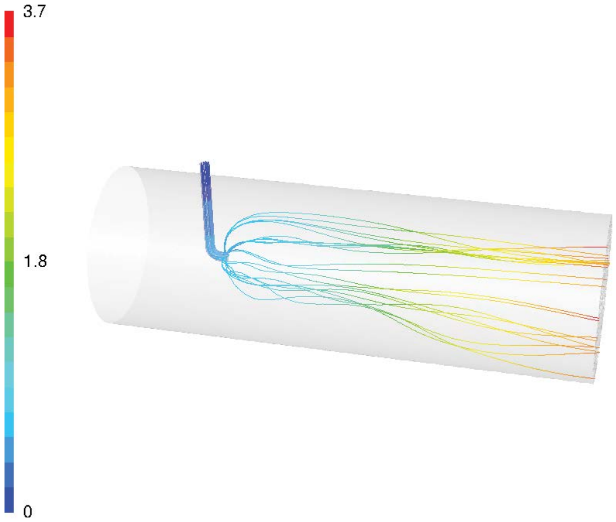

Fig. 7. Streamline of dilute H_2O_2 and retention time.

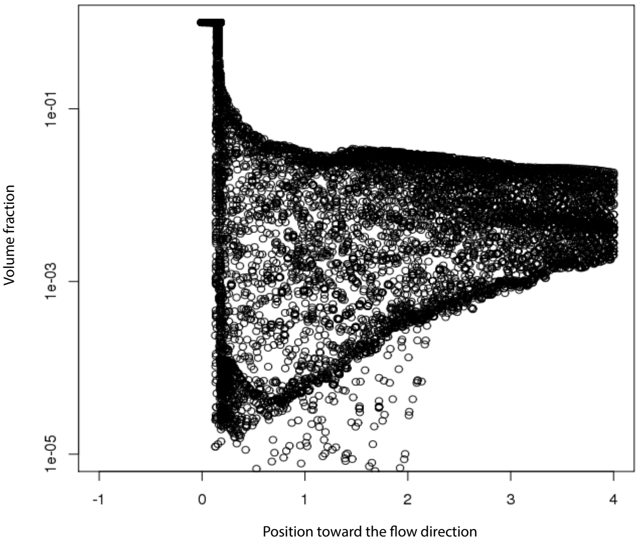

Fig. 8. Log scaled volume fraction of H_2O_2 along the flow direction.

adjusting the hole size. Rather, the result showed that the stream did not have enough radial momentum, causing it tended to converge at the center of the injection pipe. As a result, the dilute H_2O_2 solution was swept away by feed water and it made the uniformity decreased. The final uniformity of dilute H_2O_2 fraction was calculated at 35% at the discharge surface of the diffuser as shown in Fig. 11.

To split the stream of hydrogen peroxide, the number of existing diffusers was increased. Each diffuser had four holes, but one had + arrangement and the other had × arrangement. Here, the length of the entire pipe was extended to set the distance from the downstream side to the outlet, equally. The fluid conditions were the same as the previous experiment, but the dilute oxidant was divided into two diffusers with the same flow rate. The flow rate of the oxidant injected

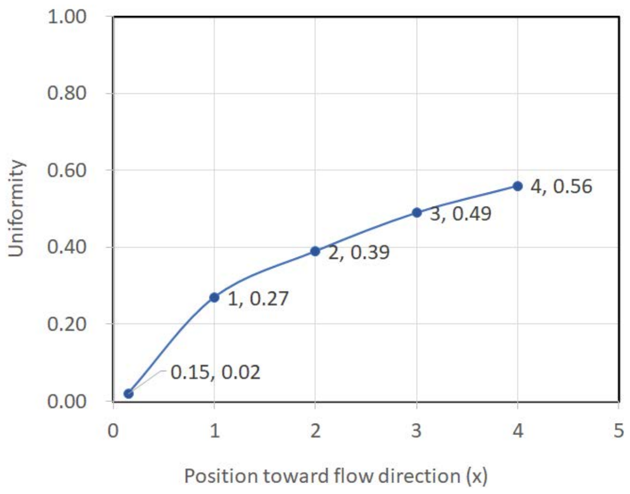

Fig. 9. Uniformity of H_2O_2 along the flow direction.

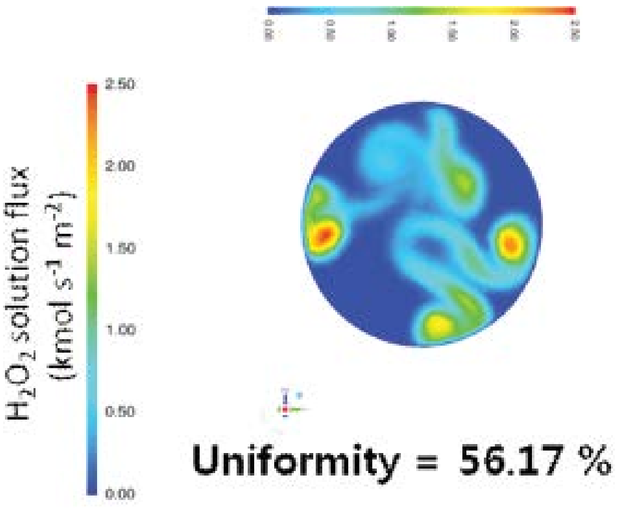

Fig. 10. Flux and uniformity of dilute H_2O_2 at the discharge the surface.

into one diffuser was reduced in half, and it resulted in a decrease of uniformity when the number of the diffuser was doubled. It was considered that it was because the stream did not have enough radial momentum, causing the tendency of converging at the center of the diffuser as shown in Fig. 12. The split of the injection stream resulted in lessening the momentum of fluid. Finally, it was swept away by feed water and converged in the central region of the diffuser. The fraction of diluted H_2O_2 obtained a worse uniformity of 34% at the diffuser outlet surface as shown in Fig. 13.

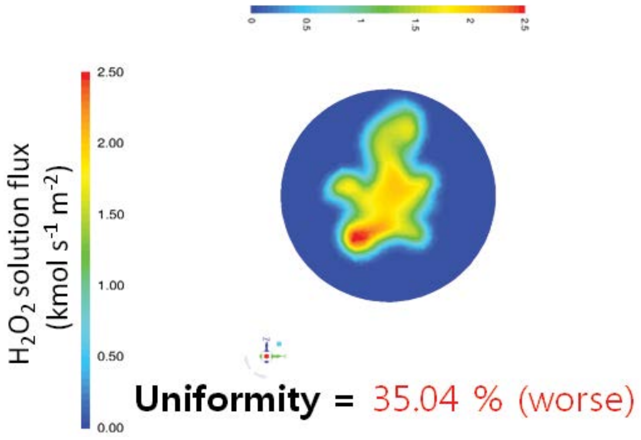

Fig. 11. Flux and uniformity of dilute H_2O_2 at the diffuser with doubled holes.

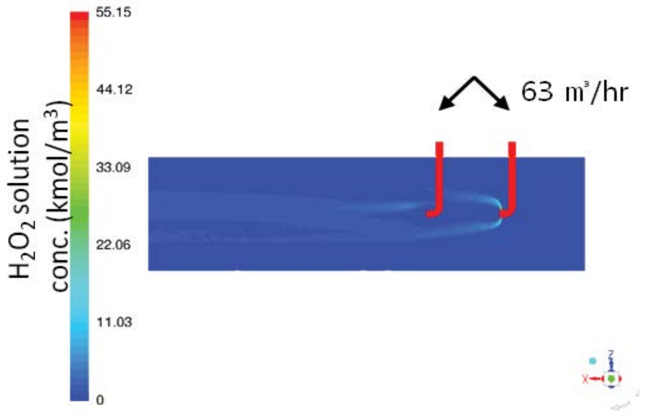

Fig. 12. Concentration of dilute H_2O_2 with dual diffusers.

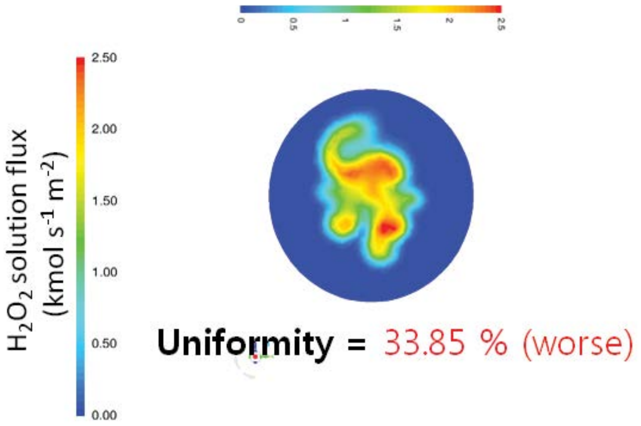

Fig. 13. Flux and uniformity of dilute H_2O_2 with doubled diffusers.

To increase the injection velocity of the oxidizing agent for turbulent flow in the pipe, the dilute ratio of H_2O_2 was doubled and the same designs related to the diffuser were applied. When the flow rate of the dilute H_2O_2 was doubled and the pressure at the nozzle was strengthened, the uniformity of the outlet was improved up to 79%. Besides, a diffuser with 8 holes and a doubled flow rate of H_2O_2 improved the mixing rate up to 83% as shown in Fig. 14. It showed sufficient hydraulic pressure should be secured first for efficient mass transfer with dispersion, and radially distributed orifice can lessen dead zone in mixing.

In this study, the dispersion of oxidants was reviewed, focusing on the oxidizer injection conditions to improve the mixing rate within a short contact time of 5 s in the pipe. To increase oxidation efficiency in the AOP unit, it is

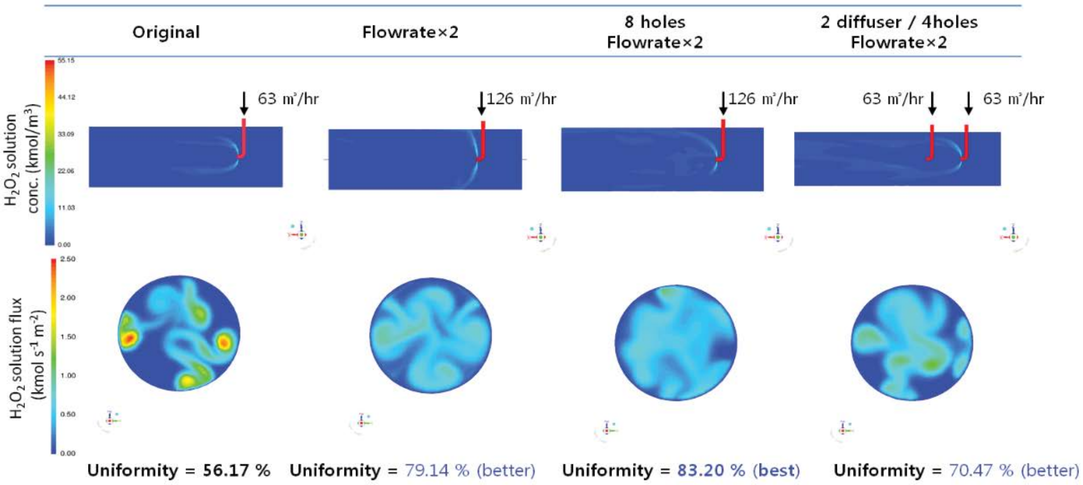

Fig. 14. Uniformity comparison between existing and modified diffusers.

necessary to maintain sufficient water pressure and to be injected at dispersed points in the pipe.

4. Conclusions

When the AOP process is operated in a water purification plant, the mixing effect of the oxidant injection device may be degraded due to the lower operation rate than the initial design capacity. Therefore, this study focused on upgrading the mixing performance of hydrogen peroxide in the existing ozone-based AOP system using the CFD. The analysis results showed that the existing design of the mixing system was inadequate for efficient mixing between oxidant and treated water; The injection flow of hydrogen peroxide could not overcome the momentum of the feed water, and the main pipe length was short to provide the contact time for the oxidant and feed water. The mixing effect of the oxidant could be improved through modification of the injection conditions, and an increase of flow rate and dispersion injection points could maintain sufficient injection pressure in the main pipeline. However, dissociation and reactivity of hydrogen peroxide with pollutants or scavengers should be further analyzed using the CFD analysis.

Acknowledgments

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