

# **Drinking Water Treatment using the Combined Ozonation and Membrane Ultrafiltration Processes**

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## **Abstract**

Ozonation and membrane ultrafiltration are used in a new drinking water treatment technology to remove water contaminants from surface waters. The production of microbubbles in this technology prevents membrane fouling, contributing to their continuous cleaning by several mechanisms including scrubbing and detachment of deposits through the self-collapse of microbubbles. The transmembrane pressure was maintained below the critical membrane pressure of 35 psi for 12 to 18 months during full-scale operations without chemical backwashing. The technology removes turbidity, color, organic contaminants and pathogenic substances according to stringent environmental standards. Currently, in Canada there are eight full-scale drinking water treatment plants using this technology.

**Key words:** Ozone; Ultrafiltration Membrane; Drinking Water Treatment; Microbubbles; Membrane Fouling Prevention

## **Introduction**

Strict regulations have been introduced in most countries including Canada for the production of drinking water from surface waters in an effort to reduce the risk to consumers. The treatment objectives in the production of drinking water include the removal of pathogenic substances, i.e. bacteria, viruses and protozoa, suspended and colloidal material, color, smell and taste and reductions in the concentrations of naturally occurring organic compounds such as humic acid and algal metabolites as well as inorganic contaminants. New drinking water regulations are emphasizing the need for the removal or reductions in the concentrations of trace contaminants or micro-pollutants that are often found in rivers and lakes through the discharge of untreated or inadequately treated wastewaters.

The use of ozone in the treatment of drinking water is gaining attention because of its effective disinfection and chemical oxidation reactions and its strength in the removal of micro-pollutants

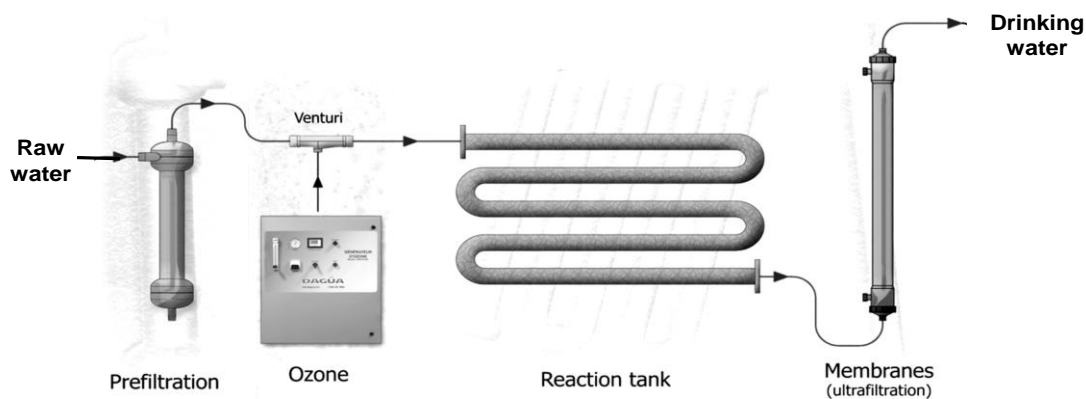
that result from pharmaceutical or industrial operations. Esplugas et al. (2007) investigated the application of advanced oxidation processes (AOP) and ozonation to remove micro-pollutants including endocrine disrupting chemicals (EDCs), pharmaceuticals and personal care products (PPCPs) in water and reported successful results with the use of ozonation alone.

This paper discusses an innovative use of ozonation followed by ultrafiltration membrane process in a drinking water treatment technology, called DaguaFlo, and presents the treatment results of this technology at full-scale operations. The generation of microbubbles and their contribution to continuous cleaning of membranes and prevention of membrane fouling is also discussed. The performance results of this technology at pilot-scale operations were reported by Niquette et al. (2007).

## Materials and Methods

### Design and Operation of the DaguaFlo Technology

In the DaguaFlo technology, the raw water from lakes and rivers passes through a pre-filtration unit before being ozonated, as presented in Figure 1. Pre-filtration commonly uses a rotary sieve with a pore size that depends on the quality of water and can be around 400 microns. Ozone is then injected into the water. A venturi injector is commonly used in full-scale operations for the injection of ozone into the water to ensure high mass transfer and efficient dissolution of ozone in water. Ozone is thoroughly mixed with water in a reaction chamber for a few minutes to ensure proper disinfection and oxidation of contaminants. The gas-saturated water then passes through ultrafiltration membranes for a complete decontamination and production of drinking water that emerges downstream of the membranes.



**Figure 1.** Schematic diagram of the DaguaFlo technology

The DaguaFlo technology does not use chemicals for coagulation. In addition, acid/base cleaning of membranes in this technology (using citric acid and sodium hydroxide) is carried out on an infrequent basis, once every 12 to 18 months, as opposed to conventional membrane-based

treatment technologies that need frequent use of acid/base for membrane cleaning and de-clogging. Moreover, using ozone as the primary disinfectant ensures the removal or considerable reductions in the concentrations of precursors of disinfection by-products (DBPs), notably trihalomethanes (THMs) and haloacetic acids (HAAs), thus preventing the generation of these hazardous compounds during the secondary disinfection process that commonly uses chlorine.

The two major contaminant removal processes in the DaguaFlo technology are ozonation and ultrafiltration membrane processes. Ozone is used for disinfection and chemical oxidation of contaminants. As an exceptionally powerful natural disinfectant gas, more active than chlorine, ozone removes pathogenic substances including viruses, bacteria and Giardia and Cryptosporidium protozoa. As a powerful oxidant, ozone oxidizes organic and inorganic contaminants, promotes coagulation of suspended and colloidal particles, and removes taste, color and smell that are referred to as organoleptic properties of water, and improves water clarification. Ozone also removes or reduces the concentrations of trace contaminants that result from industrial and pharmaceutical operations and gasoline additives, and reduces the content of iron and manganese in water. The follow-up ultrafiltration membranes remove suspended particles and organic materials and further improve the turbidity and color of water.

The removal of organic matter in water by ozonation results from their breakdown through direct or indirect reactions with ozone and subsequent interactions of organic constituents with metal ions that are usually present in surface waters, leading to the removal of complex organics from the liquid phase by precipitation, coagulation, or sorption onto metal hydroxide flocs. The removal of colloidal particles is materialized by several mechanisms that have been suggested for the effect of ozone on the destabilization, aggregation and coagulation of colloidal particles. These mechanisms target desorption of natural organic matter (NOM) from the surface of colloidal particles and prevention of NOM sorption that is known to increase steric and electrostatic barriers to coagulation and inhibit the aggregation of colloidal particles. The suggested mechanisms include breakdown of natural organic matter (NOM) and association of NOM constituents with existing metals followed by the precipitation of resulting complexes and their inhibited adsorption onto the particle surface, breakdown of sorbed NOM and desorption of stabilized organic coatings from the colloidal particles, polymerization of natural organic matter leading to particle aggregation via bridging reactions, breakdown of iron and manganese complexes leading to in situ production of metal coagulants, and production of coagulating polymers and flocculant aid by lysing algae and releasing biopolymers into the water (Jekel, 1994; Mysore et al., 1996).

## **Microbubble Generation and their Effect on Membrane Cleaning**

As a unique attribute, the DaguaFlo technology benefits from the generation of microbubbles and their use to prevent membrane fouling, contributing to continuous cleaning of membranes. The microbubbles are generated by passing the ozonated water through a gas-liquid separation unit such as a centrifuge to remove undissolved gases before water enters the membranes. Consequently, when the gas-saturated water passes through the membranes, the transmembrane

pressure gradient transforms the dissolved gases into microbubbles. The microbubbles contribute to the prevention of fouling and continuous cleaning of membranes by several mechanisms including scrubbing and self-collapse that creates pressure waves, contributing to the detachment of deposits. Microbubbles benefit from a large surface area, high gas-dissolving capability and enhanced mass transfer efficiency. In addition, they have the tendency of shrinking under water due to the surface tension and dissolution of gas in the surrounding water. According to the Young-Laplace equation (Agrawal et al., 2012; Takahashi et al., 2003), the shrinking of microbubbles leads to the progressive increase in their internal pressure until they collapse. The high pressure spot created at the final stage of microbubbles collapse produces pressure waves that will be distributed in the vicinity of a collapsing bubble, and will promote the detachment of deposits from the membranes. As reported by Agrawal et al., (2012), microbubbling is more efficient than chemical cleaning with NaOCl solution for the removal of extracellular polymeric matrix of biofilms. Furthermore, collapsing microbubbles have been shown to generate free radicals (Agrawal et al., 2011, 2012; Takahashi et al., 2003, 2007), which react rapidly and non-selectively with the chemicals in the water and cause their rapid disintegration. The strong oxidation effects of free radicals are particularly important for the removal of micro-pollutants that have recently become a central focus of regulatory organizations for the treatment of drinking water.

In addition to oxygen and nitrogen (if air is used for ozone generation), the microbubbles also contain ozone, promoting the oxidation of organic matter and enhancing coagulation of colloidal particles, further preventing the attachment of fouling material to the membrane capillaries.

Microbubbles are effective in de-clogging and prevention of membrane fouling because membrane fouling occurs primarily due to the formation of biofilm and entrapment of organic molecules and colloidal particles in the capillaries. The larger particulate matters do not commonly contribute to the fouling process as they are removed at the surface of membranes and their entrance into the capillaries is prevented.

The continuous cleaning of membranes by the action of microbubbles prevents the necessity of frequent backwashing by chemicals, thus prolonging the active life of membranes and avoiding additional maintenance and control requirements. Frequent backwashing using air and filtered water in the DaguaFlo technology ensures that the membrane permeate is ready for potable use while the reject stream can be directly discharged into the environment (subject to local regulation).

## **Operating Conditions and Sample Analysis**

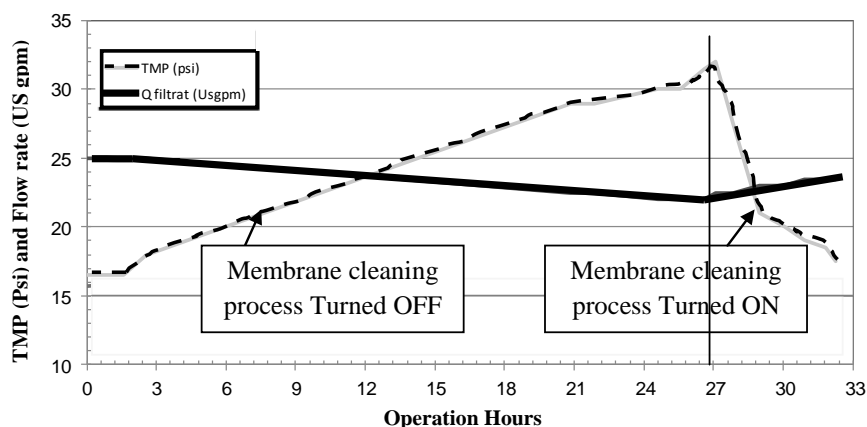
The full-scale DaguaFlo plants use Microza filtration modules containing polyvinylidene difluoride (PVDF) hollow fiber membranes developed by Asahi Kasei Chemicals in Tokyo, Japan. Each module has a diameter of 15.2 cm (OD), height of 203.2 cm, contains 6000 hollow fibers at 1.3 mm internal diameter and operates at a filtration flux of 93 to 121 L/m<sup>2</sup>/h (at 20 °C). The operating pressure of the treatment process is 30-40 psi. Ozone is generated from oxygen or

air by using a commercial ozone generator such as Pinnacle Plasma QuadBlock<sup>®</sup> ozone generators (Florida, USA), producing 8-10% ozone (w/w). Ozone is injected in the water through venturi-type ozone injectors from Mazzei (California, USA). The peak ozone use is around 12 kg/h in the summer for the treatment of 20,000 m<sup>3</sup>/day raw water. Lower ozone consumptions result in the winter because of the lower pollution and higher ozone dissolution rates.

The analyses of samples were conducted by an accredited laboratory in Quebec, Canada, according to the methods recommended by the Centre d'expertise en analyse environnement du Québec (CEAEQ) ([http://www.ceaeq.gouv.qc.ca/methodes/methode\\_para.htm](http://www.ceaeq.gouv.qc.ca/methodes/methode_para.htm)).

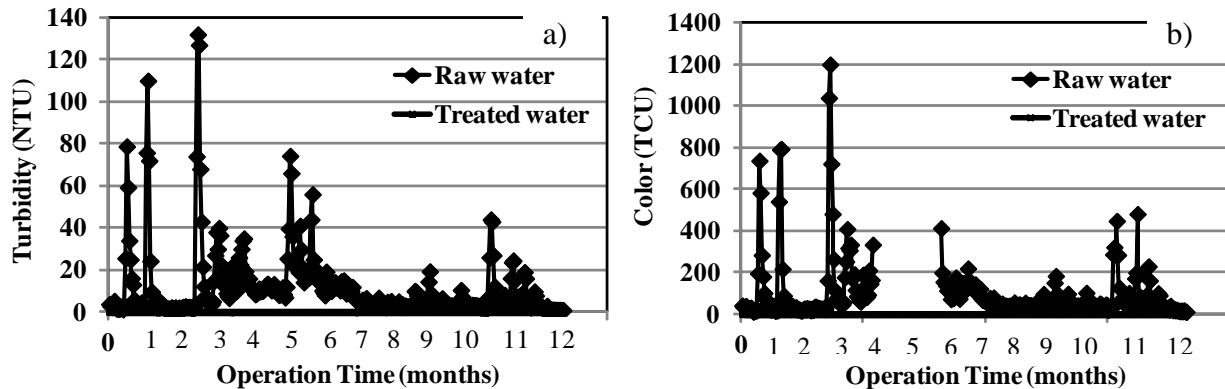
## Results and Discussion

The effect of continuous membrane cleaning process was examined by introducing the ozonated water into the membranes without degassing, allowing the accumulation of pollutants for 27 hours that increased the transmembrane pressure (TMP) (Figure 2). After the resumption of membrane cleaning process through the generation of microbubbles, the membrane pressure returned to normal levels within a few hours. The maintenance of transmembrane pressure (TMP) below the critical membrane pressure of 35 psi has been experienced during all full-scale operations of the DaguaFlo technology that have been treating water at flow rates of 640 m<sup>3</sup>/d to 20,000 m<sup>3</sup>/d. The frequency of chemical backwash and membrane cleaning by using acid and base has ranged from 12 to 18 months in full-scale operations of the DaguaFlo technology. These results present the effective membrane cleaning and fouling prevention mechanism in the DaguaFlo technology that has been materialized by the generation and use of microbubbles. This is in contrast to conventional membrane-based water treatment technologies that require frequent chemical backwashing of a few times per month. Moreover, repeated chemical cleaning will contribute to the increase in operating cost of the treatment process for chemical use and maintenance and control requirements while lowering the useful life of membranes, requiring their regular replacement.



**Figure 2.** The effect of continuous membrane cleaning process on transmembrane pressure (TMP)

Figure 3 (a and b) presents the turbidity and color of raw water and treated water at a DaguaFlo plant during the treatment of 20,000 m<sup>3</sup>/d in 2013, and shows that the values of these contaminants were consistently reduced below 0.5 NTU and 1 TCU, respectively, at the outlet of the treatment system. These values are within the standard limits set by the government of Quebec. During the reported operation period, water turbidity as high as 132 NTU (Figure 3a) and color as high as 1200 TCU (Figure 3b) were observed and properly removed.



**Figure 3.** Turbidity (a) and color (b) in raw water and treated water during the operation of full-scale DaguaFlo plant at 20,000 m<sup>3</sup>/d in 2013

The treated water samples from full-scale operations are routinely analyzed for 42 organic compounds, including volatile organic compounds (VOCs), phenolic compounds, pesticides and trihalomethanes (THMs), as well as 14 inorganic compounds and pathogenic substances to ensure compliance with environmental standards. Table 1 presents the average concentrations of faecal coliforms, total coliforms and cyanobacteria, indicators of pathogen content of drinking water, at the process outlet. These values show that the pathogen content of water was adequately reduced to levels below the detection limit of the analytical method used, well within the standards set by public health organizations.

**Table 1.** Average bacteria concentrations in raw water and treated water

Type of Bacteria	Raw Water (CFU/100 mL)	Treated Water (CFU/100 mL)
Cyanobacteria	80 - 60,000	<1
Faecal coliforms	100 - 20,000	<1
Total coliforms	3,700 - 180,000	<1

## Full-scale DaguaFlo Plants

Currently, there are eight commercial full-scale installations with the capacity of 640 m<sup>3</sup>/day to 30,000 m<sup>3</sup>/day, supplying drinking water to municipalities in Canada using the DaguaFlo technology. An example of a modular unit of the DaguaFlo plant which can treat water at flow rates of up to 1000 m<sup>3</sup>/day is presented in Figure 4. The possible placement of all components of the treatment plant in a commercial container for treatment operations up to 1000 m<sup>3</sup>/d presents the possible use of this technology for supplying drinking water in remote areas.



**Figure 4.** A modular unit of the DaguaFlo drinking water treatment plant

## Conclusions

An innovative combination of ozonation and ultrafiltration membrane processes is used in the DaguaFlo technology for effective removal of contaminants and production of high-quality drinking water. This technology does not use any chemical coagulants while benefitting from the generation of microbubbles, ensuring minimal membrane fouling and chemical-free cleaning of membranes. The absence of sludge or toxic waste generation enables the discharge of reject water into local water streams. The use of ozone before the ultrafiltration membranes provides primary disinfection, reducing the potential production of disinfection by-products that are commonly produced by the use of chlorine. The use of ozone is also beneficial for the removal or considerable reductions in the concentrations of micro-pollutants that have recently become common concerns in the treatment of drinking water.

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