

# Combined ozonation and ultrafiltration membrane processes for the production of drinking water

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**Abstract:** The combination of ozonation and ultrafiltration membrane processes is used in a patented technology called Dagua for effective, chemical-free water purification and disinfection. Drinking water is produced from groundwater or polluted surface waters according to stringent quality standards without using coagulation processes, with a reject water stream that can be reintroduced to the environment without additional treatment. Ozone disinfects, promotes coagulation and removes turbidity, color, odor, colloidal matter and organic compounds. Additionally, the generation of microbubbles provides a unique continuous cleaning mechanism for the downstream ultrafiltration membranes, preventing fouling without the use of chemical cleansers or the production of chemical waste sludge. The Dagua technology is a modular, highly scalable solution which is also suitable as a pretreatment process for nano-filtration or reverse osmosis. The efficient and sustainable treatment of water by the Dagua technology is directly linked to the efficient use of energy and chemicals, reduced generation of sludge and greenhouse gases, and use of smaller space. There are currently eight full-scale municipal installations of this technology in Canada, the largest being 30,000 m<sup>3</sup>/d.

**Keywords:** Ozonation, ultrafiltration membranes, drinking water, microbubbles

## INTRODUCTION

The combined use of ozonation and membrane filtration produces an efficient means for the removal of water contaminants and production of drinking water from lakes, rivers and groundwater, while reducing the potential formation of chlorinated disinfection by-products (DBPs). Traditional water treatment technologies that use a combination of membrane processes and ozonation commonly use ozone after the membranes in order to prevent damage to the composition, structure and function of the membranes that may result from the highly reactive nature of ozone and possible degradation of polymeric material in the membranes. However, recent studies have used pre-ozonation in an effort to reduce membrane fouling and maintain stable permeate flux using ceramic membranes (Stüber et al., 2013; Lehman and Liu, 2009; Chen and Masten, 2005).

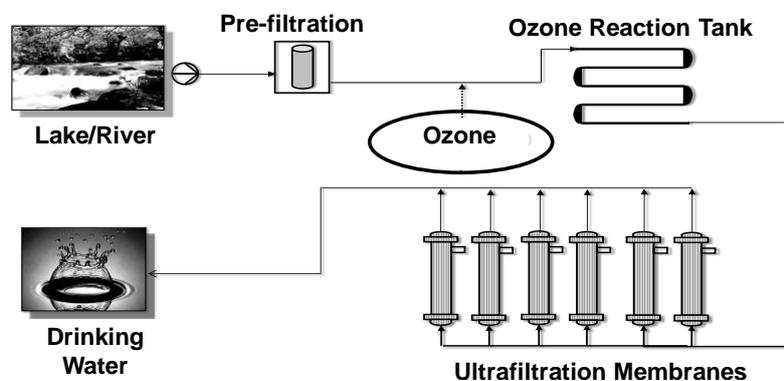
An innovative combination of pre-ozonation and ultrafiltration membrane processes for drinking water production has been used in a technology called Dagua. This technology benefits from the generation and use of microbubbles to continuously clean the membranes and prevent their fouling without using chemicals. Consequently, the membrane permeate is ready for use while the reject stream can be directly discharged into the environment since there is no chemical coagulation or frequent backwashing of membranes by toxic chemicals.

The performance of the Dagua technology was demonstrated in the treatment of highly loaded raw water with maximum turbidity of 130 NTU; maximum total organic carbon level of 7.2 mg/L, and heavily contaminated with pathogenic microorganisms (up to 20,000 CFU/100 mL fecal coliforms) in meeting the log removals required for viruses, *Giardia* cysts and *Cryptosporidium* oocysts (Niquette et al., 2007). The concentrations of all pollutants in the treated water were below the regulatory measures for drinking water standards.

## MATERIALS AND METHODS

### Design and Operation of the Dagua Technology

During the operation of the Dagua technology, the raw water initially passes through a pre-filtration unit that uses a rotary sieve with a pore size of 200-400  $\mu\text{m}$ . The water is then injected with ozone by a venturi injector to ensure high mass transfer and efficient dissolution of ozone in water. The ozonated water passes through a reaction chamber with a retention time of a few minutes (commonly 4-6 minutes) for proper disinfection and oxidation of contaminants. The water emerging from the reaction chamber passes through a degassing unit such as a centrifuge for gas-liquid separation and removal of the non-dissolved gases from the water. The gas-saturated water will pass through the ultrafiltration membranes for the removal of colloidal and suspended particles and dissolved organic materials and further improvements to the turbidity and color of water, resulting in the production of drinking water.



**Figure 1** Schematic diagram of the Dagua technology

Ozonation and ultrafiltration membrane processes are the major contaminant removal processes in the Dagua technology. Ozonation causes the disinfection and chemical oxidation of contaminants. As a powerful disinfectant gas, ozone removes pathogenic substances including viruses, bacteria and *Giardia* and *Cryptosporidium* protozoa. It is also a potent oxidant and oxidizes organic and inorganic contaminants including the emerging water pollutants that appear in water as a result of industrial and pharmaceutical activities, as well as iron and manganese, often found in surface waters and groundwater. Ozone improves water clarification by promoting coagulation of suspended and colloidal particles (Jekel, 1994; Mysore and Garyl, 1996), and by removing taste, color and smell that are referred to as organoleptic properties of water. The follow-up ultrafiltration membrane process removes suspended particles and organic materials and further improves the turbidity and color of water.

The Dagua technology does not use chemical coagulation processes. In addition, acid/base cleaning of membranes in this technology, using citric acid (2-2.5%) and sodium hydroxide (0.5-1%) is carried out on an infrequent basis, once every 12 to 18 months, unlike conventional membrane-based treatment technologies that need frequent use of acid/base for membrane cleaning and de-clogging.

The Dagua technology benefits from the generation of microbubbles and their use to prevent membrane fouling, contributing to the continuous cleaning of membranes. Microbubbles are generated due to the transmembrane pressure gradient that transforms the dissolved gases into microbubbles when the gas-saturated water passes through the membranes. The microbubbles contribute to the continuous cleaning of membranes and prevention of their clogging by several physical-chemical mechanisms including scrubbing and self-collapse that creates pressure waves, contributing to the detachment of deposits (Agarwal et al., 2011; Takahashi et al., 2003). Furthermore, collapsing microbubbles have been reported to generate free radicals (Agrawal et al., 2011, 2012; Takahashi et al., 2003, 2007), which react rapidly and non-selectively with the chemical substances in the water and cause their rapid disintegration. Agrawal et al. (2011) reported that OH radicals and shock waves can be generated at the gas-liquid interface due to pyrolytic decomposition within the collapsing bubbles. In addition to oxygen and nitrogen (if air is used for ozone generation), the microbubbles also contain ozone that will be decomposed and produce OH radicals, promoting the oxidation of organic matter and enhancing destabilization of colloidal particles, further preventing the attachment of fouling material to the membrane capillaries.

The effect of continuous membrane cleaning and fouling prevention mechanism has been demonstrated by the maintenance of transmembrane pressure (TMP) below the critical membrane pressure of 35 psi during all full-scale operations of the Dagua technology (Yerushalmi, 2014).

## **Components of the Full-Scale Dagua Technology**

The full-scale Dagua plants use Microza filtration modules containing polyvinylidene difluoride (PVDF) hollow fiber membranes (Asahi Kasei Chemicals, 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 (206.8-377.1 kPa). 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 by venturi-type ozone injectors (Mazzei, California).

The analyses of water samples withdrawn from the full-scale Dagua plants 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) ([www.ceaeq.gouv.qc.ca/methodes/methode\\_para.htm](http://www.ceaeq.gouv.qc.ca/methodes/methode_para.htm)).

## **RESULTS AND DISCUSSIONS**

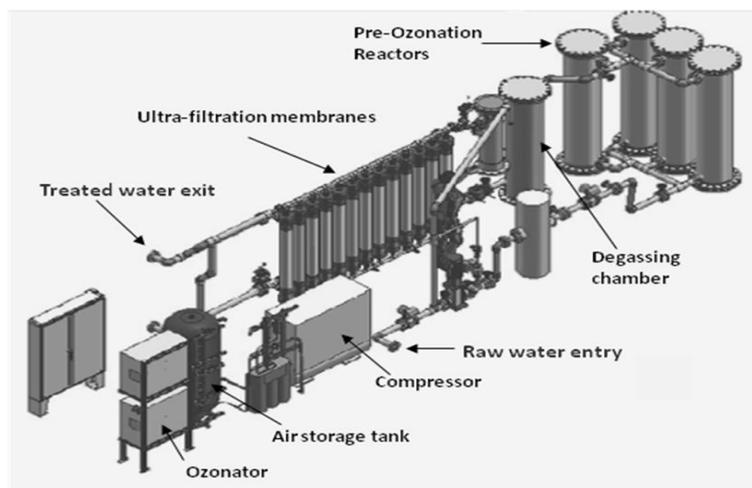
### **Full-scale Dagua Plants**

The operation of Dagua technology has been demonstrated in eight full-scale plants with the capacities of 640 m<sup>3</sup>/day to 30,000 m<sup>3</sup>/day, providing drinking water to municipalities in Canada. The oldest Dagua plant is nine years old while the most recent plant is three years old.

An example of a modular unit of the Dagua plant which can treat water at flow rates of up to 1000 m<sup>3</sup>/day is presented in Figure 2 while Figure 3 presents the components of the treatment plant. The possible placement of all components of the treatment plant in a mobile container for treatment operations up to 1000 m<sup>3</sup>/d presents the possible use of this technology for water purification and supply of drinking water in remote areas.



**Figure 2** A modular unit of the Dagua water treatment plant



**Figure 3** Components of a modular unit of the Dagua water treatment plant

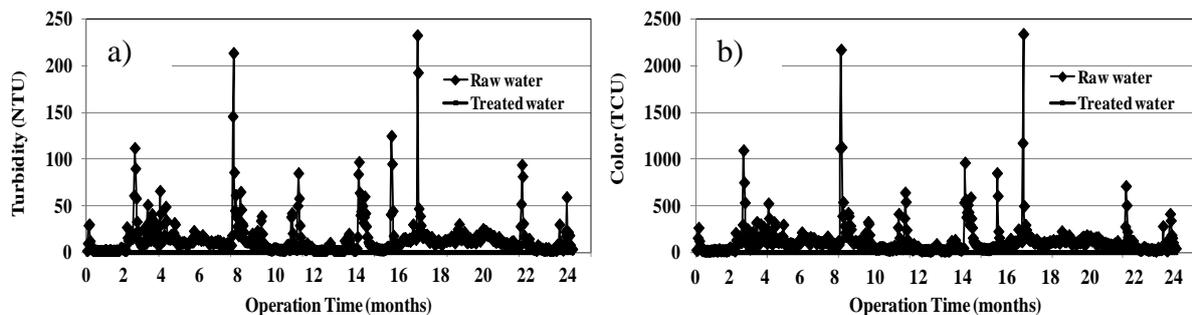
The operating Dagua plants in Canada are presented in Table 1.

**Table 1** Operating Dagua plants in Canada

Plant name	Water flow rate (m <sup>3</sup> /d)	Contamination issues
Radisson	740	Turbidity, colour, organic matter, pH
Montebello	2,000	Coliforms, iron, manganese, colour, pH
Gracefield	720	Turbidity, organic matter, colour, coliforms
Fort Coulonge	2,000	Organic matter, colour, coliforms, pH
Pontiac-Quyon	2,000	Organic matter, colour, coliforms, pH
Bryson	640	Organic matter, colour, coliforms, pH
Les Méchins	2,000	Organic matter, colour, coliforms
St-Denis (AIBR)	30,000	Organic matter, colour, coliforms

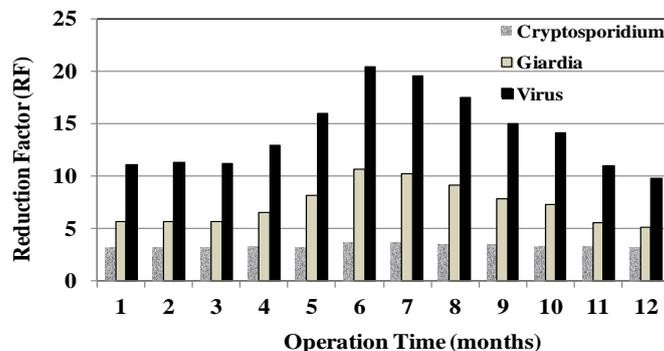
The full-scale Dagua plants have encountered occasional mechanical and electrical problems commonly occurring during long-term operations of full-scale plants. They included leaks in the piping, deterioration in the function of pumps and valves, reduced generation of ozone due to the low capacity of ozone generator and air compressor requiring adjustment of their capacities, reduced mass transfer of ozone during cold winter months requiring modifications to the ozone injection assembly, increased concentration of tannin in raw water requiring increased rate of ozone injection, and minor adjustments to the control and communication system. All encountered problems were rapidly corrected, ensuring uninterrupted and continuous operation of the treatment plants. The membranes were changed once in plants operating for more than seven years.

Figure 4 (a and b) presents the removal of turbidity and color during 24 months operation of a Dagua plant, treating water at 20,000 m<sup>3</sup>/day. Turbidity and color were effectively removed from the raw water, producing treated water with the turbidity of < 0.5 NTU and color of <1 TCU during the operation period. These contaminants showed similar patterns in their respective appearance, increasing in the summer, reaching peak turbidity values of 214 and 233 NTU (Fig. 4a) and peak color values of 2175 and 2344 TCU (Fig. 4b) during the two consecutive years.



**Figure 4** Removal of turbidity (a) and color (b) in a full-scale Dagua plant treating water at 20,000 m<sup>3</sup>/d

The removal of *Cryptosporidium* and *Giardia* protozoa and viruses are shown in Figure 5, demonstrating reduction factors (RF) which represent logarithmic removal of these pathogenic substances. Reduction factors were in the range of 9.8-20.4 for viruses, 5.1-10.6 for *Giardia* and 3.2-3.6 for *Cryptosporidium*. The concentrations of these pathogens as well as cyanobacteria, faecal and total coliforms in the treated water were below strict regulatory measures for drinking water standards.



**Figure 5** Removal of pathogenic substances in a full-scale Dagua plant treating water at 20,000 m<sup>3</sup>/d

## DISCUSSION

The long-term operation of the Dagua technology without the need for coagulation chemicals and without the generation of waste sludge makes it highly desirable for the production of drinking water in urban environments or remote areas that have limited facilities for the supply of chemicals and the disposal or treatment of wastewater and chemical sludge. Moreover, the full-scale Dagua plants are scalable, require minimum maintenance and can be fabricated as modular units.

The continuous cleaning of membranes in the Dagua technology 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 of every 20 minutes using air and filtered water in the Dagua 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). Membranes are commonly replaced once every seven to eight years.

The reject water from the Dagua process can be safely discharged into a river, local sewer system, storm drain or a pond depending on the local facilities and regulations. The reject water from the Dagua process does not contain pathogenic substances since they have been killed or inactivated by the ozonation process and retained by the membranes. In addition, the reject water does not contain toxic chemicals since the Dagua technology does not use any chemical processes such as coagulation and flocculation. Moreover, the salt concentration in the reject water from the Dagua process is equal to that in the raw water. The discharge of Dagua reject water into the local rivers has been safely exercised during the last nine years of full-scale operation of this technology.

In addition to the production of drinking water from surface waters or groundwater, the Dagua technology is also suitable as a pre-treatment process for nano-filtration or reverse osmosis. The pre-treatment of water will reduce the clogging and the frequency of backwashing of nano-filtration and reverse osmosis membranes, thus increasing the uninterrupted operation of treatment plants and prolonging the active life of membranes. Moreover, the Dagua technology has been strategically designed for integration in a water reuse system composed of a treatment framework with advanced processes for cost effective transformation of grey water or wastewater into highly purified drinking water. The possibility of full-cycle water remediation and reclamation by using high-efficiency membrane filtration and adequate disinfection will respond to the demand for improved water efficiency and environmental protection in water-scarce regions.

The specific advantages of the Dagua technology are listed below:

- Lack of coagulant or flocculant use
- No generation of toxic waste: No need for the handling and treatment of toxic waste
- Possible discharge of the reject water into the environment
- Entirely automated operation with the possibility of remote control
- Limited fouling of membranes: low frequency of cleaning, extended longevity of membranes
- High efficiency of treatment: using only 10%-15% of treated water for membrane cleaning

## REFERENCES

1. Agarwal A., Ng W. J. and Liu Y. (2011). Principle and applications of microbubble and nanobubble technology for water treatment. *Chemosphere*, **84**, 1175-1180.
2. Agarwal A., Xu H., Ng W. J. and Liu Y. (2012). Biofilm detachment by self-collapsing air microbubbles: a potential chemical-free cleaning technology for membrane biofouling. *Journal of Materials Chemistry*, **22**, 2203-2207.
3. Chen K. C. and Masten S. J. (2005). Effect of combined ozonation-ultrafiltration on membrane fouling and water quality. *Journal of the Chinese Institute of Environmental Engineering*, **15**(4), 263-268.
4. Jekel M. R. (1994). Flocculation effects of ozone. *Ozone Science and Engineering*, **16**, 55-66.
5. Lehman S. G. and Liu L. (2009). Application of ceramic membranes with pre-ozonation for treatment of secondary wastewater effluent. *Water Research*, **43**(7), 2020-2028.
6. Mysore C. and Garyl A. (1996). Effects of ozone on the colloidal stability and aggregation of particles coated with natural organic matter. *Environmental Science and Technology*, **30**, 431-443.
7. Niquette P., Hausler R., Lahaye P. and Lacasse M. (2007). An innovative process for the treatment of high loaded surface waters for small communities. *Environmental and Engineering Science*, **6**, 139-145.
8. Stüber J., Miehe U., Stein R., Köhler M. and Lesjean B. (2013). Combining ozonation and ceramic membrane filtration for tertiary treatment. *Chemie Ingenieur Technik*, **85**(8), 1237-1242.
9. Takahashi M., Kawamura T., Yamamoto Y., Ohnari H., Himuro S. and Shakutsui H. (2003). Effect of shrinking micro-bubble on gas hydrate formation. *Journal of Physical Chemistry B.*, **107**, 2171-2173.
10. Takahashi M., Chiba K. and Li P. (2007). Free-Radical generation from collapsing microbubbles in the absence of a dynamic stimulus. *Journal of Physical Chemistry B.*, **111**, 1343-1347.
11. Yerushalmi L. (2014). Drinking water treatment using the combined ozonation and membrane ultrafiltration processes. International Ozone Association, Pan America Group (IOA-PAG), Montréal, Canada, August 23-27.