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Potable Water Quality and Membrane Technology

Requirements for potable water quality have changed dramatically from the late 1970s due to enhanced regulation of pathogens and disinfection by-products. Consequently, new treatment technology and analytic techniques are required to meet this challenge, affecting clinics, laboratories, universities, and other organizations involved in the analysis and treatment of potable water. The changing health effects and regulations affecting potable water treatment and associated applications of membrane technology are discussed.

ABSTRACT *Outbreaks of waterborne disease and chronic adverse health effects of disinfection by-products (DBPs) have generated potable water regulatory changes that have affected consumers, laboratories, and clinics nationwide. Existing and future regulations require numerous and better techniques for analysis of pathogens and DBPs, as well as improved water treatment. Membranes are the most versatile and promising technology for achieving higher-quality potable water. Clinical personnel will develop and provide analytic techniques to meet these challenges.*

Health Effects

Pathogens

The search for a method to purify drinking water was spurred by a cholera epidemic in London in 1834. Through painstaking research, John Snow, MD, discovered that the source of the outbreak was a contaminated well. But it was not until the 1890s, after Schwann, Pasteur, Koch, and others made invaluable contributions to bacteriology, however, that chlorine was accredited as a disinfectant.[1] Chlorination remained the primary method of controlling waterborne disease until 1970.

Water treatment texts have correlated the declining rate of reported waterborne diseases to the number of water treatment plants using chlorine for disinfection from 1900 through 1950 in the United States.[2,3] Until *Cryptosporidium* species were identified in the 1990s, *Giardia lamblia* was the most important waterborne pathogen. Between 1971 and 1985, there were 92 reported outbreaks of giardiasis involving 24,365 individual cases.[2]

Outbreaks of waterborne disease are common to surface or ground water supplies. Fox[4] categorized the causes of waterborne disease outbreaks from 1971 to 1985 and found that 49% were due to inadequately treated groundwater (well supplies), 24% to inadequately treated surface water (lakes, rivers), 16% to storage or distribution contamination, and 11% to miscellaneous causes.

The transmission of waterborne diseases via potable water was considered well controlled until a 1993 outbreak of cryptosporidiosis in Milwaukee. This outbreak resulted in more than 100 fatalities and 400,000 illnesses among the local population.[2] Most of the fatalities and serious illnesses were among individuals with deficient immune systems. The cause of the infection was discovered by a Milwaukee physician who recognized symptoms, required clinical testing, and confirmed cryptosporidiosis. This occurrence has been attributed to contamination of Lake Michigan and the inability of the water treatment process to adequately eliminate the *Cryptosporidium* organisms. Moreover, this occurrence and the efforts of medical and clinical professionals allowed the potable water community to recognize that waterborne disease was not as well controlled as believed and that new regulations,

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clinical techniques, and treatment technologies were required.

Unfortunately, many occurrences of waterborne disease go unrecognized and unreported. Fox[4] summarized the questions that must be asked regarding outbreaks once an individual has been reported as infected:

- Did illness occur?
- Was medical care sought?
- Was the appropriate clinical test (blood, stool) conducted?
- Was the laboratory proficient?
- Was the clinical test result positive?
- Did the patient comply with treatment?
- Were the results reported to the appropriate health agency?
- Was the report timely?
- Did the health agency act?

Each of these questions provides an opportunity for breaking the sequence and not reporting an outbreak.

Clinical personnel are directly involved with the reporting sequence and have reliable techniques for detecting pathogens in stool and blood samples. Aqueous samples present a greater challenge than biological specimens due to the lower concentration of pathogens. Existing and emerging pathogens that are currently most significant in potable water have been summarized recently by Fox[4] (Table 1). The effects of these pathogens include gastrointestinal disease, ulcers, and death. The need to reassess microbiologic treatment and monitoring of potable water systems has been demonstrated by the recent outbreaks of *Escherichia coli* and resulting deaths in Canada and the reporting of inaccurate reporting of *Cryptosporidium* oocysts and *G lamblia* cysts in Sydney, Australia's potable water.[5] Although techniques exist for the detection and monitoring of microorganisms, new techniques with increased sensitivity to lower concentrations of pathogens in aqueous samples and ready availability to the potable water community must be developed.

The American Water Works Association Research Foundation has begun projects involving spectroscopy for the detection of protozoa, evaluation of antibodies to *Cryptosporidium* species and *G lamblia* using flow cytometry, and vital dye staining of *G lamblia* and *Cryptosporidium* species.[6-8] These projects offer funded research opportunities for university, clinic, and laboratory personnel who want to improve detection of pathogens that affect potable water. Measurement of *Cryptosporidium* species in aqueous samples is costly and time-consuming; for this reason, Sylvania et al[9] have suggested the use of beads as surrogates for *Cryptosporidium* species.

Disinfection By-Products

Disinfection by-products (DBPs) were first detected in potable water by Rook.[10] Organic matter is ubiquitous to natural water, as is DBP formation during disinfection. Only DBPs formed during chlorination or ozonation are regulated. Consequently, consumers of disinfected potable water are at risk of ingesting DBPs. Regulated DBPs formed during chlorination include trihalomethanes (THMs; chloroform, dichlorobromomethane, dibromochloromethane, and tetrachloromethane) and haloacetic acids (HAAs; trichloroacetic;

AU: micrometer ok?

AU: credit line for table? or cite [4] at title?

Table 1. Major Potable Water Pathogens

Protozoa (3-25 µM)	Algae (1-500 µM)	Bacteria (1 µM)	Virus (0.02-0.03 µM)
Existing			
Cryptosporidium species		*Escherichia coli*	Hepatitis A
Giardia lamblia		*Salmonella* species	Rotavirus
Amoeba		*Shigella* species	Polio
		Vibrio cholera	Norwalk
		Legionella species	
Emerging			
Cyclospora species	Cyanobacterial toxins	*Helicobacter pylori*	Unnamed
Microsporidia		*Mycobacterium avium* complex	
		Pseudomonas aeruginosa	

dichloroacetic; monochloroacetic; dibromoacetic; and monobromoacedtic). Bromate is a regulated DBP formed during ozonation.

DBPs such as chloroform are alleged carcinogens and represent a threat of chronic toxicity, as opposed to pathogens, which represent a threat of acute toxicity. Federal regulations[11] are developed from health effect models that are designed to limit fatalities based on DBP consumption to no more than 1 per 100,000 based on the consumption of 2 L of water per day at the maximum allowable DBP consumption. Studies have correlated the consumption of more than 5 glasses of water per day containing more than 75 µg/L THMs or 18 µg/L dibromochloromethane to an excessive rate of spontaneous abortions among women.[12]

Accurate measurement of DBPs and DBP formation potential, performed by gas chromatography, requires strict control of the DBP reaction with regard to disinfectant concentration, organic concentration, pH, temperature, bromide concentration, and time. Laboratories conducting chemical and biologic analysis of potable water as required by regulation must be registered with state and federal governments. They have a very large market, which is expected to grow substantially as regulation of potable water is increased.

Regulations

The development of potable water regulations is complex, requiring the efforts of the medical, analytic, and engineering communities. Improved analytic techniques have allowed detection of low levels of contaminants and have benefited health effects research. Once the presence of contaminants is demonstrated, the cost and benefits of proposed regulations are evaluated. Regulations that have driven technologic change and had the greatest recent impact on potable water treatment are the Surface Water Treatment Rule[13, 14] and the Disinfection By-Product Rule.[11]

The Surface Water Treatment Rule requires a 3-log reduction (LR) of *G lamblia* and allows 2.5 LR for coagulation, sedimentation, and filtration (CSF) and 0.5 LR for disinfection. Conventional treatment of surface waters involves the addition of iron or aluminum salt, which forms a solid floc and settles, removing contaminants. CSF processes operating with conventional engineering guidelines are given 2.5 log reduction; however, the 0.5 log reduction for disinfection requires a specific CT (the product of disinfectant concentration and time) and varies by water quality and organism. The CT concept, well known to microbiologists for years, is a simplification of the Chick-Watson law.[15]

Initial DBP regulation began in the late 1970s with THMs. Presently, the maximum contamination limits (MCLs) for THMs and HAAs is 80 µg/L and 60 µg/L, respectively, which will be fully enforced beginning in December 2001. The federal government will further reduce the THM and HAA MCL in the near future, which will significantly affect the water community. An MCL of 40 µg/L THMs and 30 µg/L HAAs has been proposed.

As shown in the 3 following formulas, the control of DBPs and pathogens poses a conflict for traditional potable water treatment technologies. DBP formation and pathogen inactivation increase with increasing disinfectant dose and contact time. One means of reducing pathogens without increasing DBPs is to remove the organic precursors prior to disinfection. Both coagulation and membranes can achieve that, but membranes are significantly more effective.

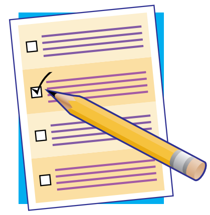
1. DBP Reaction
Organics + Chlorine → DBPs + Chlorine

2. Disinfection Reaction
Viable Pathogens + Chlorine → Nonviable Pathogens + Chlorine

3. Regulatory Conflict: Pathogens vs DBPs
Pathogens ↓ Chlorine↑ : DBPs↑ Clorine ↑

Membrane Technology

Membranes, UV radiation, ozonation, and carbon adsorption are processes that have been newly applied to potable water treatment. The most significant of these is membranes, which can be broadly defined as a barrier to the flow of suspended, colloidal, or dissolved solids in any solvent. Membrane processes used for potable water treatment are reverse osmosis (RO), electrodialysis reversal (EDR), nanofiltration (NF), ultrafiltration (UF), and microfiltration (MF).[16] The lower limits of EDR-RO, NF, UF, and MF particle rejection in potable water are 0.0001 µm, 0.001 µm, 0.01 µm, and 0.1 µm, respectively. However, the size range for each process is broad (Fig 1). Membranes remove contaminants but do not form by-products and offer the broadest range of contaminant rejection. RO, sometimes called



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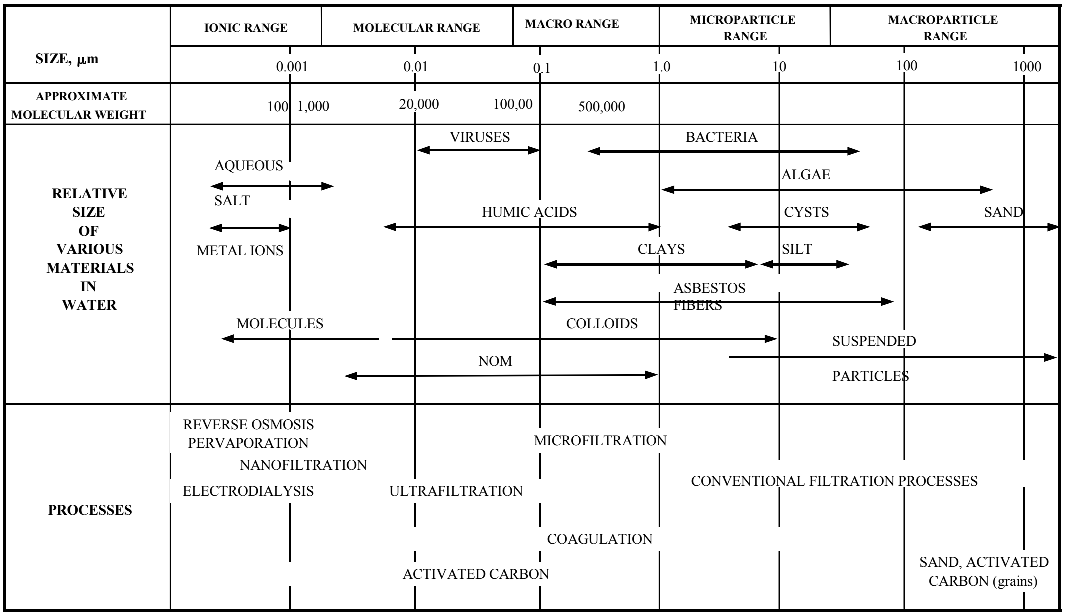

Fig 1. Size ranges of membrane processes and contaminants. NOM, natural organic matter.

desalination, removes salt from brackish or seawater, and can reject almost all solutes. RO membranes are also used in laboratories to produce high-purity water. EDR can remove ions or salts from brackish or seawater, but has no capability to remove uncharged species such as pathogens.[17] NF is used to remove hardness (due to calcium and magnesium) and DBP precursors. UF and MF are used to eliminate turbidity, pathogens, and particles from fresh waters.

The fundamental differences between diffusion controlled (RO/NF), size exclusion (UF/MF), and charge (EDR) processes are illustrated in Figures 2 to 4. Mass transfer through membranes or films is affected by factors such as contaminant size, charge, surface chemistry, pore size, and solubility.[16-21] UF/MF rejection is controlled primarily by size exclusion (Fig 2). Rejected particles are simply too large to pass through the pores. Osmosis and RO (Fig 3) are diffusion controlled. If a semipermeable membrane filters aqueous solutions of varying salinity, water will flow from the low- to the high-saline solution by osmosis; salts will pass from the high- to the low-saline solution by diffusion. RO results when a pressure greater than the osmotic pressure is applied to the high-saline solution and water is forced into the low-saline solution. Diffusion is not affected by pressure. Flow into the low-saline solution increases as pressure increases, and at high pressures, very pure water is produced. Charge significantly affects RO mass transfer. Duranceau and Taylor[22] demonstrated 50% rejection of sodium (molecular weight, 23; charge, –1) and no rejection of ethylene dibromide (molecular weight, 189; charge, 0) by NF in field studies.

EDR is an ion-exchange process (Fig 4). Charged ions move toward a countercharged pole through a cation- or an anion-permeable membrane. These membranes are staggered, which allows segregation of ions between the membranes. Cations move toward the negative pole, passing through a cation-permeable membrane, and are trapped when rejected by an anion-permeable membrane. The reverse occurs for anions. Consequently, only charged contaminants are removed by EDR, which typically limits the process to the removal of salts.[17]

Currently, *G lamblia* is the limiting microorganism for design of disinfection processes. The US Environmental Protection Agency allows 2.5 log rejection for CSF treatment.[13] Determination of LR is shown in (4) and illustrated in Table 2. LR is often reported as simply greater than the level of detection as shown in the LR 1 column of Table 2. Regulators often interpret such reporting as the upper limit of rejection. Reporting the LR as shown in the first row of the LR 2 column, which reads "infinite rejection for a 1-log challenge," avoids misinterpretation.

It is important to report feed concentration using the volume of the collected sample to correctly determine LR. For example, collection of a 1-mL sample in Table 2 shows infinite rejection of a 1-log challenge. This is determined by setting C_p = 0#/mL and C_f = 10#/mL. If the permeate and feed concentration are inadvertently reported in L, then the LR may be reported erroneously as infinite rejection for a 4-log challenge instead of a 1-log challenge. LR can be accurately reported only when detection is made in permeate and feed samples.

$$LR = -Log\left(\frac{C_s}{C_f}\right)$$

where: LR = log rejection
C_s = sample concentration
C_f = feed concentration

UF/MF rejects 6 logs and RO/NF rejects 5 logs or more of *cryptosporidium*, more than 1,000 times the rejection of conventional potable water treatment. However, UF/MF rejects fewer DBP precursors (natural organic matter) and more *Cryptosporidium* organisms than RO/NF (Fig 5).[23] It is surprising that UF/MF rejects an order of magnitude more *Cryptosporidium* organisms than RO/NF as UF/MF pores are larger than RO/NF pores. However, the largest membrane pores are an order of magnitude smaller than *Cryptosporidium* organisms. The difference in rejection is due to the difference in construction. Membranes for potable water are made in a spiral-wound or a hollow-fiber configuration. Spiral-wound membranes are assembled by gluing flat sheets together. The glue lines separate the raw water from the treated water but pass organisms at high concentrations. Hollow fiber membranes are assembled by potting the ends of hollow fibers in a rigid polymer without glue lines.

Before the advent of log-reduction regulations, RO/NF was used for rejection of dissolved solids. The most challenging water was seawater with a total dissolved solids concentration of 36,000 mg/L. A 2 log reduction was all that was needed. UF/MF has been used only for turbidity rejection. Recently, engineers conducting 6-log challenges using *Cryptosporidium* spp and aerobic spores found that some organisms always pass membranes. However, 20 log rejection or more is attainable using membranes in a multiple barrier approach.[18]

Summary

Changing potable water regulations have created opportunities for improving analytic techniques and the treatment of potable water. Analysis of microbiologic species and chemical contaminants is and will continue to be required of all suppliers of public potable water. These challenges must be met by the development of new analytic techniques and treatment techniques for potable water. Membranes are the most promising tech-

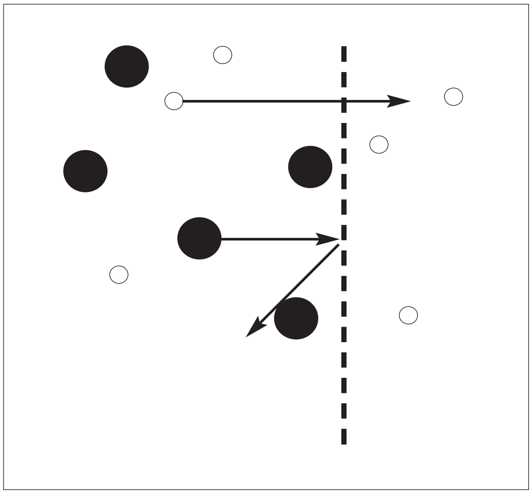

Fig 2. Particle rejection by size exclusion.

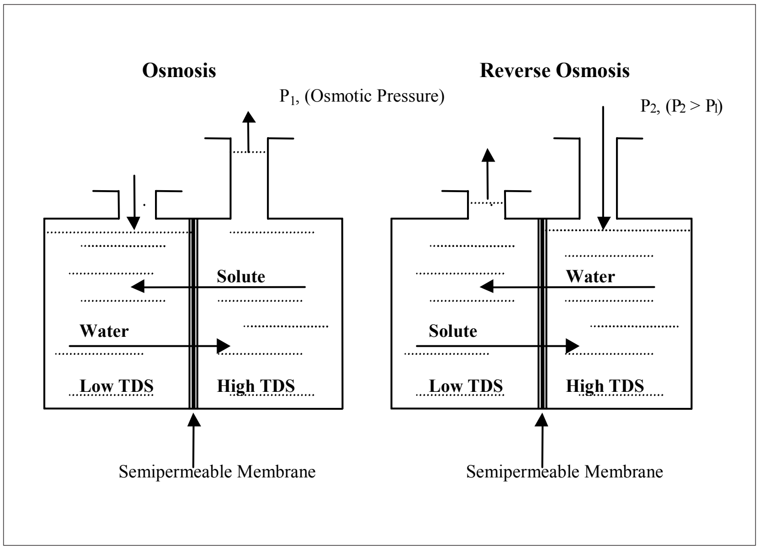

Fig 3. Osmosis and reverse osmosis. TDS, total dissolved solids.

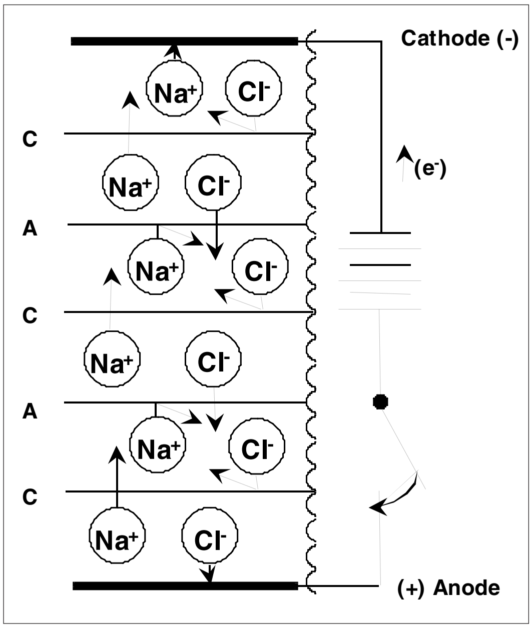

Fig 4. Electrodialysis cell. A, anion membrane. C, cation membrane.

Table 2. Illustration of Log Rejection of Microorganisms

Feed Concentration	Sample Volume (L)	Permeate Concentration (No./L)	LR 1	LR 2
10,000	0.001	0	>1	∞1
10,000	0.01	0	>2	∞2
10,000	0.1	0	>3	∞3
10,000	1	0	>4	∞4
10,000	10	1	5	5
10,000	100	10	5	5
10,000	1,000	100	5	5

nology for potable water treatment for meeting these challenges. Based on the recent regulatory trends, the demand for better microbiologic and chemical analysis and treatment of potable water will increase significantly.[24,25]

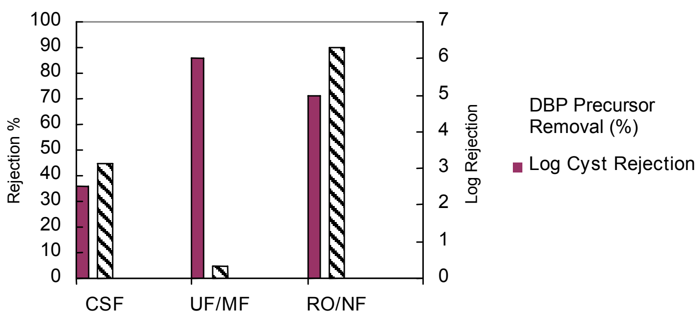

Fig 5. Comparison of disinfection by-product (DBP) and cyst rejection. CSF, coagulation, sedimentation, and filtration; MF, microfiltration; NF, nanofiltration; RO, reverse osmosis; UF, ultrafiltration.

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