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PROCESSES FOR ADVANCED TREATMENT OF WATER

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ABSTRACT

A case-oriented approach is used to illustrate developments and applications of biological and physicochemical technologies, either alone or in sequenced arrays, for advanced treatment to facilitate reuse of water in a range of environmental scenarios. Examples cited include chemical oxidation processes for removal of color and oxygen demand from textile mill effluents, sequential anaerobic/aerobic biological treatment of recalcitrant and inhibitory organic compounds in ammunition plant wastewaters, separation and recovery of organic solvents from mixed industrial waste streams, remediation of contaminated subsurface waters, membrane treatment of effluents from secondary biological wastewater treatment plants, and integrated bio-membrane treatment of industrial and municipal wastewaters. The paper provides an overview of methods and applications for source waters of various qualities. It concludes with a generalized guide to technology selection based upon specific water characteristics. © 1999 IAWQ Published by Elsevier Science Ltd. All rights reserved

KEYWORDS

Adsorption; anaerobic/aerobic sequenced bio-processes; bio-membrane processes; chemical oxidation; membrane processes; water treatment for reuse.

INTRODUCTION

Our use of the term water in an unqualified sense in the title of this plenary lecture at an international conference on wastewater treatment is purposeful. Its use is consistent with the senior author's long-held view (Weber, 1972) that the ever increasing frequency with which Earth's normal hydrologic cycle is short-circuited by expanding anthropogenic activities blurs the boundaries between natural waters, water supplies, and wastewaters. It is consistent as well with the related conclusion (Weber, 1972) that we must address ourselves to a virtual continuum of water quality, and to the development and implementation of means for transforming water of any quality along this continuum to any other quality required for a particular use.

The water quality continuum does have practical bounds, but these are widely separated. At the lower bound we have specific use-based standards. These range from contaminant concentration levels of analytically non-detectable to as high as several thousand milligrams per litre. The higher bound, which is based on available source water quality, generally ranges from a few milligrams per litre to tens of thousands of milligrams per litre.

While there are several unit treatment operations that can reduce the highest source levels of some contaminants to values approaching their most stringent use-standard levels, these generally involve prohibitive costs for stand-alone use. Rather, we are generally faced in any given situation with selecting some combination of unit operations that provides required levels of treatment at costs that are less per unit volume of water treated than the equivalent value(s) of water produced. Figure 1 is an attempt to capture the three-dimensional aspects of this situation for a specific combination of unit processes. The term M/V on the "x" axis of the figure represents the contaminant mass per unit volume of the source water, $\Delta M/V$ on the "y" axis the increment of mass reduction required to meet a given use standard, C on the "z" axis the cost per unit M/V , and C_T in three-dimensional space the total cost per unit volume of water treated. Each combination of technologies yields a different total cost per unit volume of water produced, which must then be compared to the use-related value of the product water to assess treatment train feasibility. This comparison should include considerations of water use, and the costs and benefits of measures that can be invoked to reduce water demands.

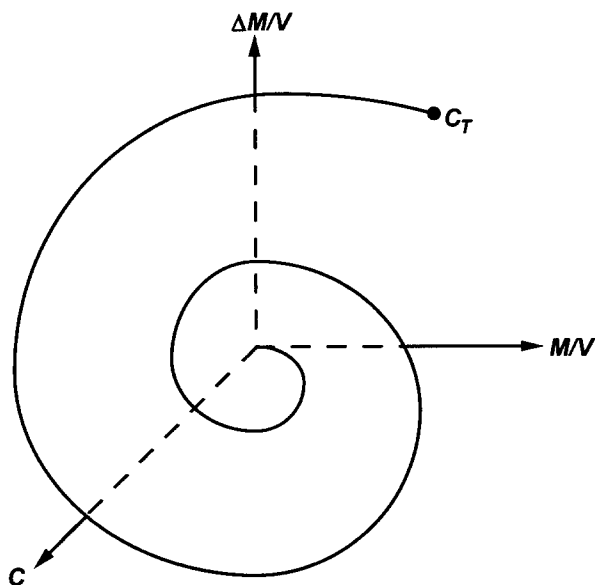


Figure 1. The Cost Spiral.

It is clear that the value placed on a particular measure of water quality remarkably influences the feasibility of using a given treatment train to produce a specific product from a specific source water. There is, for example, no question of treatment feasibility associated with converting a relatively low quality source water to a pristine drinking water product if the consumer values that product at the one dollar (U.S.A.) or so per gallon (i.e., ~470 Italian lire per liter) level associated with its purchase in plastic bottles at the food market. Symptomatic of public sector value systems, however, the usual consumer would typically object if he or she had to pay a similar cost for a similar quality water delivered at the household tap. A two-dimensional representation of the treatment feasibility domain resulting from the quality and cost element representation given in Figure 1 is shown schematically in Figure 2.

The obvious need for increased reuse of water significantly impacts both source-water quality and product water value. Water reuse can be of two primary forms. The first of these, *cascading* reuse, is a form practiced to one degree or another since humankind first used water. The practice involves sequential use of water of deteriorating quality for purposes having decreasingly stringent requirements. Fundamentally, a product water having the most stringent use standard commands the highest quality source water available. After its designated use it becomes the source for product water having the next most stringent requirement,

etc. In many applications, particularly industry and agriculture, such cascading reuse may require only minimal intermediate treatments, and the lowest quality water is then disposed of as an unusable waste.

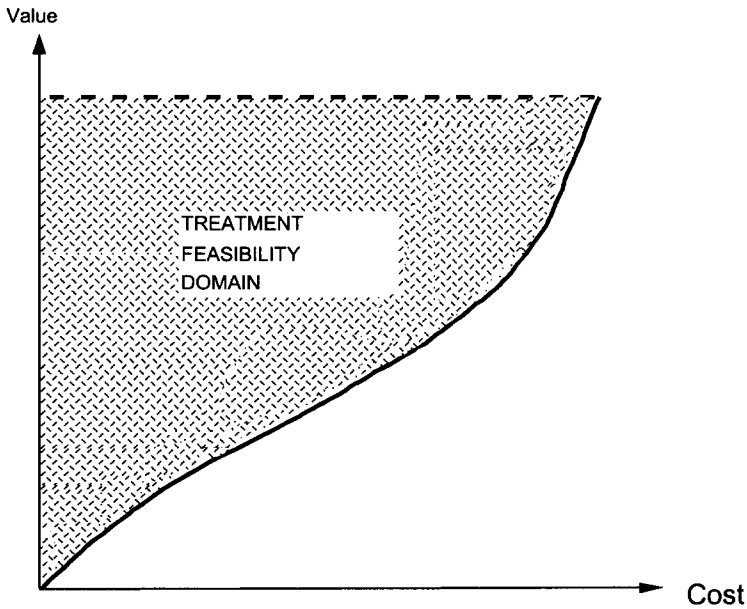


Figure 2. The Treatment Feasibility Domain.

The second type of water reuse involves repeated *recycling* for the same use. The ultimate configuration of a recycling scheme might be envisaged as closing the loop on the cascading reuse scheme; i.e., the lowest quality product becomes the source for the use having the highest quality requirement. The recycling reuse scenario is relatively new compared to simple cascading reuse with ultimate wastage of the lowest quality water. It is the scenario that presents the greatest technology challenge, and it is the scenario that is rapidly becoming most prevalent as the sizes and per capita water demands of human populations escalate simultaneously in the face of a fixed global water resource.

Our use of the term advanced *treatment vis à vis* advanced *processes* in the title of this paper is also purposeful. As the qualities of available water sources steadily decline under the influence of increasing anthropogenic activity, and as use-related standards increase as concentration levels of contaminant detectability and regulatory concern are lowered, it is likely that we will be required to provide increasingly advanced levels of treatment using more or less traditional technologies that can be advanced only marginally in unit effectiveness and efficiency as a function of research and development. In other words, no "silver-bullet" technology is evident on the horizon.

PROCESSES FOR ADVANCED TREATMENT

Process coupling

The ultimate reuse scenario described above requires the most complex array of coupled processes to ensure a safe water supply at an affordable cost. In municipal water reuse schemes this typically means coupling the most technically and cost effective "wastewater" treatment technologies with the most technically and cost effective "water" treatment technologies in complimentary treatment trains. In cases of reuse for potable purposes, treatment trains often employ redundant systems in efforts to assure safe water. Table 1 provides a summary of treatment technologies employed as redundant barriers for specific classes of

contaminants in a five-year potable water reuse evaluation scheme implemented by the city of Denver, Colorado (USA) in 1985.

Table 1. Redundant treatment barriers in the Denver potable water reuse evaluation scheme (after Lauer and Rogers, 1996)

Contaminant Category	Treatment Barriers						
	AS	Cl ₂ /NHCl ₂	GAC	HpLC	O ₃	RO/UF	UV
Viruses and Bacteria		⊕		⊕	⊕	⊕	⊕
Protozoa		⊕		⊕	⊕	⊕	
Metals and Inorganics			⊕	⊕		⊕	
Organics	⊕		⊕	⊕		⊕	

AS = Air Stripping; Cl₂/NHCl₂ = Chlorination/Chloramination; GAC = Granular Activated Carbon Adsorption; HpLC = High pH Lime Clarification; O₃ = Ozonation; RO/UF = Reverse Osmosis/Ultrafiltration; and, UV = Ultraviolet Irradiation.

Large-scale municipal reuse schemes provide good illustrations of the complementary and often redundant capabilities of process technologies for advanced treatment of water. To highlight technological advances in specific unit processes, however, it is useful to examine several different case-oriented examples. Included in the following sections are brief descriptions of several types of processes for enhanced treatment of water, including chemical and biological transformation processes, separation processes, and combined separation-transformation processes.

Chemical oxidation

Routine treatment of organic contaminants by chemical oxidation is frequently less economic than biological treatment. Systems involving the latter have inherent advantages in that treatment is fueled by energy derived from the target substrates, while chemical oxidation systems rely on inputs of energy in the form of chemical oxidants, leading to increased operating costs. Under certain circumstances, however, chemical oxidation systems can be used cost effectively to treat organic chemicals that are normally toxic or refractory to microorganisms, in effect serving either as detoxification or refractory organic "softening" pretreatment steps for biological processes or as self-contained processes for transformation of organic or inorganic contaminants to environmentally acceptable oxidation/reduction products. Large quantities of suspended or dissolved solids quickly consume chemical oxidants, so waters targeted for such treatment should have relatively low concentrations of oxidizable background material, in the range of no more than a few hundred mg/L of total organic carbon.

Recent advances in oxidation processes have focused primarily on reaction rates, since it is this aspect of such processes that generally governs the performance of otherwise thermodynamically favorable reactions of strong oxidants with target contaminants. Common oxidants include hydrogen peroxide, ozone, potassium permanganate, and chlorine dioxide. Each of these oxidants has unique properties and reactivity characteristics which influence its suitability and mode of use for different applications.

Hydrogen peroxide (H₂O₂), for example, is most reactive in the pH range from 3.0 to 4.0 (Merz and Waters, 1949). Adjustments in pH to enhance the oxidation effectiveness of this reagent are normally accomplished through use of one or more catalysts, the most common being ferrous sulfate (FeSO₄), in so-called Fenton reagent reactions in the pH region of ~3.5 (Bowers, 1996).

Recent uses of Fenton reagent reactions have focused on color removal from biologically recalcitrant textile mill dyes, and the destruction of toxic or refractory organics occurring in low concentrations in contaminated groundwater systems. Kuo (1992) examined the oxidation of five different types of simulated dye wastewaters containing major commercial dyes (reactive, direct, acidic, basic, and disperse dyes) and five wastewaters randomly collected from textile dyeing mills. Optimal combinations of H₂O₂ and FeSO₄

for each of the five simulated wastewaters studied were determined. The five selected dye mill wastewaters were then each treated with these optimal dosage combinations, resulting in average COD removals of 90%, and decolorizations greater than 97%.

Sequenced anaerobic/aerobic bio-processes

Numerous advances in biological treatment have occurred over the past ten years, fueled largely by increasingly strict regulations on discharges of BOD, organic phosphorus, nitrogen, and toxic organic chemicals from both industrial and municipal facilities. Aerobic treatment processes have traditionally been employed for reduction of BOD, but concurrent reductions of other contaminants often proves infeasible without coupling aerobic treatment with anaerobic or anoxic pretreatments. Anoxic and anaerobic treatments have been found to provide opportunities for transformation of biological phosphorus, nitrogen, and many organic chemicals that are toxic to aerobic systems (Zitomer and Speece, 1993). Although complete mineralization of toxic organics is not normally accomplished by anaerobic treatment, the ability of these systems to biologically transform toxic organic chemicals to forms more easily degraded in aerobic environments greatly expands the capabilities of biological systems for treatment of organic wastes.

While anaerobic systems are often beneficial in reducing the toxicity of refractory organics as a pretreatment step for subsequent aerobic treatment, a number of difficulties can be encountered with this process. Problems include sensitivity to shock or variable substrate loadings, and the need to retain naturally slow-growing anaerobic cell populations for longer periods of time; i.e., slow-growing anaerobic systems cannot afford losses of cell mass common to fast-growing aerobic systems. Shock loading problems in large systems can usually be overcome by using mass equalization basins. More innovative methods may be required for low flow systems that experience widely varying substrate loadings. One such method is the use of granular activated carbon (GAC) adsorption columns in the recycle loops of recycling anaerobic bioreactors to control concentrations of inhibitory compounds in the reaction chambers (Fox and Suidan, 1996).

Enhancing the residence time of sludge biomass can be accomplished by use of reactors providing appropriate means for cell attachment, such as sand, GAC, or polymeric membranes. GAC is particularly advantageous for treatment of refractory and/or toxic organic wastes because of its ability to attenuate large concentration swings in chemical feed streams, thereby allowing for enhanced process performance.

Berchtold *et al.* (1995) described the use of this technology for treatment of wastewater containing 2,4-dinitrotoluene (2,4-DNT) from a propellant production facility. Except for a specific *Pseudomonas* species that is able to use 2,4-DNT as a sole carbon source, 2,4-DNT is normally resistant to aerobic biotransformation (Spanggard *et al.*, 1991). A continuous-flow anaerobic fluidized-bed GAC (An-GAC) reactor was used as the first step of the sequential treatment process. The 2,4-DNT was found under methanogenic conditions (with ethanol as the primary substrate), to degrade predominantly to 2,4-diaminotoluene (2,4-DAT). The second sequential step employed the use of aerobic batch activated sludge (AS) reactors to fully mineralize the 2,4-DAT within a nine-hour treatment period.

Adsorption

Applications of sorption technologies in water treatment have grown from the original, and still widely-employed, use of GAC as a broad-spectrum sorbent for removal of assortments of organic chemicals and heavy metals over wide concentration ranges (Weber, 1972; Weber and Smith, 1986), to the development and application of more specialized polymeric adsorbents having chemical compositions and forms engineered specifically for targeted solute removal and/or recovery.

Polymeric adsorbents take one of two general forms; carbonaceous and non-carbonaceous. The carbonaceous forms perform in a manner similar to GAC, except that they are often more selective for specific organic chemicals at lower solute concentrations, and less easily fouled by dissolved organic matter. Non-carbonaceous polymeric sorbents, which have enjoyed long and widespread use as ion exchange resins, are more recently being evaluated for selective removal and concentration (for subsequent reuse) of organic solutes in industrial operations (Gusler *et al.*, 1993). Industrial process waste streams often contain a

number of components of varying chemical characteristics, some of which may be reusable if recovered in relatively pure form. The broad-spectrum properties of activated carbon that provide such an advantage for water recovery and reuse in many mixed contaminant situations may not be desirable for targeted solute recovery and reuse.

Interesting applications of this technology in work focused on the use of engineered adsorbents (polymeric and carbonaceous) to target removal of specific solutes in mixed-solute industrial waste streams, and allow for subsequent high-purity solvent recovery have been described by Weber and vanVliet (1981), Gusler *et al.* (1993), and Hand *et al.* (1994). In the Gusler *et al.* (1993) study, an industrial wastewater containing three organic components (phenol, toluene, and chlorobenzene), each at one-half of its aqueous solubility, was examined with respect to optimizing recovery and reuse of phenol. Evaluation of the equilibrium sorption isotherms in Figure 3 for XAD-12 polymeric resin and F-400 granular activated carbon (GAC) reveals a significantly different pattern of phenol adsorption by the polymeric adsorbent relative to its adsorption patterns for of toluene and chlorobenzene. The patterns of F-400 for adsorption of those solutes are more singular to each other. This suggests two possible approaches to recovery of the phenol.

First, one may use the polymeric resin (XAD-12) alone in a fixed bed reactor configuration. This results in selective separation of phenol from toluene and chlorobenzene on the XAD-12 resin, which has 71% higher capacity for phenol than for the other sorbates. A second approach would be to use a short fixed-bed GAC reactor, followed in series by an adsorber containing XAD-12 resin. This configuration would allow preferential removal of chlorobenzene on the GAC prior to its entrance into the XAD-12 reactor. This approach results in recovery of 99.3% of phenol at nearly 100% purity (Gusler *et al.*, 1993).

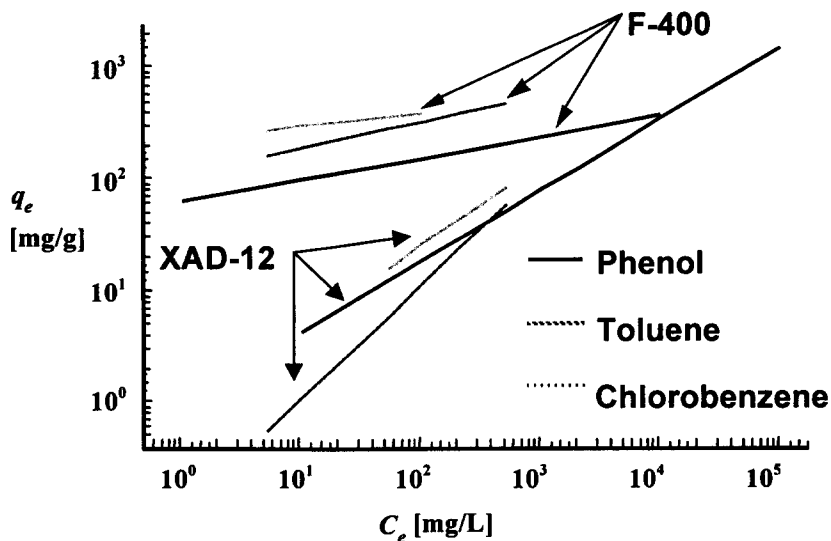


Figure 3. Equilibrium isotherms for a mixed industrial waste (after Weber and vanVliet, 1981; Gusler *et al.*, 1993; and Hand *et al.*, 1994).

A second example deals with a problem commonly encountered in the use of GAC to remove specific organic chemicals from polluted natural surface and subsurface waters. Such treatments are frequently complicated by the presence of dissolved natural organic matter (NOM), which can significantly reduce the sorption capacity of GAC for organic pollutants (Carter *et al.*, 1992; Carter and Weber, 1994; Hand *et al.*, 1994). One treatment strategy for moderate levels of dissolved NOM involves its pre-adsorption by selected high affinity sorbents followed by removal of targeted contaminants by GAC. A study by Boening *et al.* (1980) reveals that several polymeric resins have significantly greater affinities than GAC for adsorption of dissolved NOM. Selectively removing NOM, therefore, can improve solute adsorption on more selective resins such as GAC.

Another strategy noted by Hand *et al.* (1994) involves the use of sorbents that are selective for the target compounds and that do not significantly sorb dissolved NOM. Pilot-plant studies by this group showed that the synthetic carbonaceous adsorbent Ambersorb 563 is more efficient in removing trichloroethylene (TCE) from water having a low NOM background concentration.

Membrane processes

A number of pressure-driven membrane processes are finding increased use for advanced water treatment, including microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO). Table 2 summarizes the increasingly limiting membrane pore sizes (and thus increasing permselectivity) and associated target contaminants for these respective pressure-driven separation processes. Other membrane processes driven by forces other than mechanical pressure (e.g. osmosis and electrodialysis) also have environmental applications, but these are more limited and will not be discussed here.

While fouling and member life continue to be problematic in many applications, recent advances in polymer technology have led to membranes having improved contaminant rejection capabilities, lower pressure requirements, and greater durability. In reverse osmosis, for example, these developments have led to two-fold increases in membrane performance over that of conventional cellulose acetate membranes (Matsuura, 1993). Such achievements have been realized primarily by the use of chemically resistive layers of poly(amide)-based or poly(ether)-based materials placed on top of cellulose acetate membranes; i.e., composite membranes. For RO applications, these polymer composites provide a very thin layer having either a positive or negative surface charge, resulting in higher water fluxes due to higher salt rejections originating from ion-ion repulsion.

Table 2. Membrane pore size characteristics and target contaminants

Membrane Process	Average Membrane Pore Size [nm]	Target Contaminants
MF	200	Suspended solids, turbidity, and pathogens (<i>Cryptosporidium</i> , <i>Giardia</i> , bacteria).
UF	2-50	Macromolecules, viruses, colloids, and proteins.
NF	< 2	Color, organics, pesticides, and metals.
RO	< 1	Dissolved solids, nitrates, and radionuclides.

MF = Microfiltration; UF = Ultrafiltration; NF = Nanofiltration; and, RO = Reverse Osmosis

The use of RO systems for production of potable water from saline and brackish sources has become increasingly feasible because of the increasing cost effectiveness of such systems relative to thermal distillation systems. Some of the more innovative applications of membranes have come about in the conversion of wastewaters for reuse applications. Large-scale applications include a number of municipal treatment facilities designed for water reuse, including the plant operated by the city of Denver, Colorado from 1985-90 (Lauer and Rogers, 1996), Water Factory 21 (McCarty *et al.*, 1981), and the proposed San Diego Water Repurification Plant (McEwen and Richardson, 1996).

Another interesting area of development in membrane treatment technology is that of so-called bio-membrane processes. Such processes may take one of three general system forms: extraction systems (Livingston, 1994); fixed-film systems (Illias and Schimmel, 1995; Aziz *et al.*, 1995; Ganapathi *et al.*, 1995); and filtration systems (Urbain *et al.*, 1996). In each case, the membrane serves as a barrier to facilitate selective solute transport, enhance biofilm attachment, and/or restrict microorganism access to downstream permeate.

Illustrative case-oriented examples of bio-membrane process applications have been given by Livingston (1994), and by Ilias and Schimmel (1995). For industrial waste streams originating from an organic synthesis process, the presence of high concentrations of inorganic materials such as acids, bases, salts, and left-over catalysts can adversely affect the ability of microorganisms to effectively degrade toxic organic compounds. The use of extraction bio-membrane systems can circumvent this problem by specifically targeting facilitated transport of organics through the membrane. Livingston (1994) describes the use of extractive membrane bioreactors (EMB) which employ silicone rubber and other organophilic polymers known to be permeable to volatile chlorinated hydrocarbons, but are impermeable to water and other polar and ionic compounds. For EMB systems similar to that shown in Figure 4, this allows for better control of the composition medium to be biodegraded, effectively replicating similar conditions under which the biomedium culture was developed (Livingston, 1994).

Industrial or municipal wastewaters may also contain trace amounts of organic compounds at concentrations too low to sustain biological populations in normal reactor systems. This problem may be resolved through the use of fixed-film bio-membranes permeable to organics on one side while supporting biofilm growth on the other side. This allows the growth of cell populations an order of magnitude greater than those achieved in suspended growth reactors (Ilias and Schimmel, 1995).

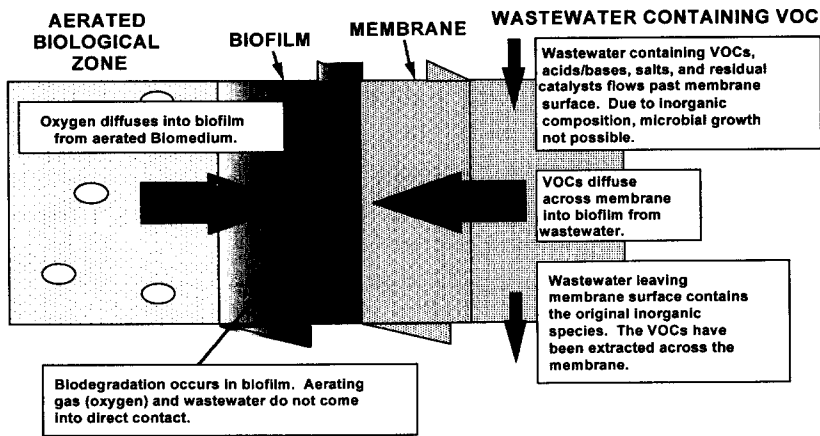


Figure 4. Detoxifying chemical process wastewaters (after Livingston, 1994).

Table 3. Biological and physicochemical treatment processes for advanced water treatment

Target Contaminants	An/A Bio	Bio-M	COX	GAC	POLY	MF-UF	RO
Diss. Solids			⊕				⊕
Susp. Solids	⊕	⊕				⊕	
BOD	⊕	⊕		⊕		⊕	⊕
Pathogens			⊕			⊕	⊕
Non-VOCs	⊕	⊕		⊕	⊕		⊕
VOCs	⊕	⊕	⊕	⊕	⊕		⊕
Metals	⊕						⊕

An/A Bio = Anaerobic/Aerobic Bio-Processes; Bio-M = Bio-Membranes; COX = Chemical Oxidation; GAC = Granular Activated Carbon Adsorption; POLY = Synthetic Polymer Adsorption; MF-UF = Microfiltration and Ultrafiltration; RO = Reverse Osmosis

SUMMARY

Table 3 provides a brief summary of the technologies for advanced water treatment discussed in this paper, along with target contaminant categories that these technologies are respectively best suited to address.

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