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MODELING AND SIMULATION OF WASTEWATER REUSE SYSTEMS(U)  
LOUISIANA STATE UNIV BATON ROUGE DEPT OF CHEMICAL  
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MODELING AND SIMULATION OF WASTEWATER REUSE SYSTEMS

Annual Report

August 1978

(1 June 1977 to 30 June 1978)

Cecil L. Smith

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Abstract

This report summarizes the work completed to date. Copies of all papers presented to date are included in the appendix.

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## 1.0 Summary of Work

The work accomplished to date can be divided into two categories: (1) component models and (2) model integration. The latter is accomplished with a general purpose process simulator program for water reuse processes.

### 1.1 Component Models

The component modules include the following: (1) ultrafiltration, (2) reverse osmosis, (3) ozonation, and (4) hypochlorination. The simulator structure enables a model for the equalization tank to be created using the general features of the simulator.

#### 1.1.1 Ultrafiltration

The present model for the tubular ultrafiltration module is based on the film theory for mass transfer through the boundary layer at the inner wall of the UF tube. Permeability coefficients are included for water, dissolved solids, and TOC. For a given module configuration, inlet flow, inlet suspended solids concentration, inlet dissolved solids concentration, inlet TOC concentration, and pressure drop across the module, the model predicts the permeate flow, permeate dissolved solids concentration, and permeate TOC concentration. The model assumes that no suspended solids pass through the membrane. This model is described in detail in the interim report "A Mathematical Model of a Tubular Ultrafiltration Unit for Water Re-Use Systems".

#### 1.1.2 Reverse Osmosis

The model for the hollow fiber reverse osmosis module is also based on the film theory for mass transfer through a boundary layer formed at the outer surface of the hollow fibers. For a given module configuration, inlet flow rate, inlet dissolved solids concentration, inlet TOC concentration, and pressure drop across the module, the permeate flow, permeate dissolved solids concentration, and permeate TOC concentration are predicted. Analogous values for the concentrate stream are calculated by material balance. This model is described in detail in the interim report "A Mathematical Model of a Hollow Fiber Reverse Osmosis Unit for Water Re-Use Systems".

#### 1.1.3 Ozonation

The model for ozonation in the presence of UV light is based on basic kinetic mechanisms for the reaction and decomposition of ozone. The ozone enters as a gas, is dissolved into the aqueous phase and then either reacts with the organic material in the water or decomposes. For a given reactor configuration and inlet water conditions, the model predicts the effluent water TOC concentration. The model is described in detail in the interim report "A Mathematical Model of an Ozonation Contacting Unit for Water Re-Use Systems".

#### 1.1.4 Hypochlorination

The model for the hypochlorination module is based on the dissociation mechanism for hypochlorite. The model predicts the free and available chlorine concentration from inlet stream conditions. The model is described in detail in the interim report "A Mathematical Model of a Hypochlorination Unit for a Water Re-Use System".

#### 1.2 Model Integration

The component modules were incorporated into the process simulator to facilitate the simulation of a variety of configurations for water reuse processes.

##### 1.2.1 Dynamic Process Simulator

In addition to the modules specific to water re-use processes, the simulator includes modules for general process elements such as mixed tanks, pumps, stream splitters, etc. The simulator permits the configuration of the water reuse process to be specified via data cards, along with specifications for the input streams. The simulator calculates the flow and concentration of each stream in the process as a function of time. The results may be presented in tabular form or may be plotted. To provide some assurance that the models and process configuration is consistent, total and component material balances are computed for the run. The dynamic process simulator is described in detail in the interim report "A Dynamic Model for Application in Water Re-Use Systems".

##### 1.2.2 Integrated Model Example

Using the dynamic process simulator, a simulation run was made using the configuration proposed for the MUST WPE. The run demonstrated that the component models behaved as expected in an integrated configuration, and that the dynamic process simulator could successfully simulate a realistic water re-use process. Simulation of a 20-hour cycle required 24 minutes of computer time on an IBM 370/3033.

Publications and Presentations

The following publications and presentations have resulted from this work:

1. Starks, D. M., and C. L. Smith, "A Dynamic Model of a Water Treatment Unit", Proceedings of the 1977 IEEE Conference on Decision and Control, New Orleans, December 7-9, 1977.
2. Starks, D.M., and C. L. Smith, "A Mathematical Model of a Reverse Osmosis System for Water Purification Use", Proceedings of the Ninth Annual Pittsburgh Conference on Modeling and Simulation, Pittsburgh, April 27-28, 1978.
3. Starks, D.M., and C. L. Smith, "A Mathematical Model of an Ultrafiltration System for Water Purification Use", Proceedings of the 1978 Summer Computer Simulation Conference, Newport Beach, July 24-26, 1978.

Copies of all of these papers are included in the appendix.

3.0

Students

The following student received a graduate degree based on work on this project:

- David M. Starkes, Ph.D., Chemical Engineering  
Louisiana State University  
August, 1977. Dissertation Title: Mathematical Model of a  
Water Processing Unit

Appendix

Copies of papers presented to date

# TA3 - 11:00

## A DYNAMIC MODEL OF A WATER TREATMENT UNIT

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### Abstract

For support of field medical units, the U.S. Army is developing a self-contained waste water treatment system to produce potable water for use within the unit. To aid in developing the control strategy and the fault detection/fault isolation logic, a dynamic model of the system is being developed.

The first phase of the effort is to develop models of the system's components, such as ultrafiltration, reverse osmosis, ozonation, etc. These models are then combined to obtain a model for the complete system. To the greatest extent possible, fundamental equations form the basis of the models.

In determining coefficients for the models, the data was obtained from the experiments that formed the basis of the design of a pilot plant version of the unit. To refine the model, this data is then supplemented by operational data from the pilot plant.

### 1. Introduction

The U.S. Army has a requirement to provide a mission-oriented medical treatment system which is designed and equipped to facilitate rapid establishment and disestablishment. The flexibility permits immediate response for a medical support unit to any tactical, environmental or geographical change. This system will provide a contamination-free and controlled environment in which medical surgical and ancillary procedures and other supporting functions can be performed. The mobile medical treatment system is termed the MUST Medical Complex. Associated with the MUST Medical Complex is a Water and Waste Management Subsystem (WWMS). This subsystem is required to treat and dispose of, without degradation of the environment or danger to personal health, all toxic and contaminated waste materials generated within the functional areas of the Medical Complex. In addition to the waste treatment and disposal, the WPE within the WWMS must be capable of producing potable water from a fresh or brackish water source and nonconsumptive reuse water from the MUST Medical Complex waste water effluent.

The objective of the present program is to:

1. Develop an integrated dynamic model describing the operational characteristics of the water processing element. Emphasis will be placed on the reuse mode of operation utilizing the MUST hospital composite waste or the x-ray, laboratory, and OR

composite waste. The methodology to apply the model to other configurations and other wastes will be developed and documented.

2. Using the dynamic model as the basis, develop a control/monitoring system for the operation of the WPE.
3. Using the dynamic model as the basis, develop a fault detection/fault isolation package for the WPE.

This presentation will only consider the first of these objectives.

### 2. System Description

When in the re-use mode of operation, the WPE utilizes the following components in series:

1. Equalization (mixing) tank
2. Ultrafiltration
3. Microfiltration
4. Reverse osmosis
5. Ozonation (accompanied by exposure to ultraviolet light)
6. Chlorination

Dynamic models are required for each of these components.

The sources of the recycle water include showers, operating room, kitchen, laboratory, and x-ray. Due to the large number of sources of contaminants, it is impractical to identify every molecular compound. In such cases, the usual practice is to introduce pseudo-compounds, which will consist of several molecular compounds of similar characteristics. Currently, only two pseudo-compounds are being used, with all contaminants being treated as either dissolved or suspended solids.

In developing the design for the WPE, the first step was to develop and operate a 1/4-scale breadboard system. The next step was to construct a full scale pilot plant. Data from both serve as the experimental basis for the dynamic model.

### 3. Model Formulation

The dynamic models for each component of the WPE are based on fundamental concepts, such as heat balances, material balances, mass transfer equations, etc. For example, the model for the ultrafiltration unit involves the following equations:

1. Material balance on total contaminant
2. Total material balance
3. Diffusion of the contaminant into and out of the boundary layer at the tube wall.

Models of other elements of the WPE involve relationships such as heat transfer, reaction kinetics, etc.

Although the basic theoretical equations are relatively easy to develop, numerical values for the various coefficients are more difficult to obtain. Empirical correlations are also often necessary. In the ultrafiltration model, the following are used:

1. The mass transfer coefficient is related to the diffusivity.
2. The osmotic pressure is related to the concentration of contaminant.

Furthermore, some physical properties (such as the diffusivity) may be functions of operating conditions (such as contaminant concentrations).

#### 4. Model Calibration

In order for the model's behavior to match that of the physical system, the unknown parameters in the various relationships are adjusted until the model results agree with the experimental results to within an acceptable tolerance. In the ultrafiltration model, five parameters must be determined in this manner. In this case, experimental data is currently available only for steady operation, although future plans call for data to be collected during periods of non-steady state operation.

After the parameters have been fit to the experimental data, the model behavior must be observed under several different conditions to verify that its behavior appears to be "reasonable". Further tests of this type are necessary after the component models have been integrated into a model of the complete system.

The presentation will describe the experiences of the authors in developing component models for the WPE, with emphasis being on the process of establishing a valid model from the available experimental data.

# A MATHEMATICAL MODEL OF A REVERSE OSMOSIS SYSTEM FOR WATER PURIFICATION USE

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## ABSTRACT

This paper describes the development of a mathematical model depicting the reverse osmosis unit employed in the water processing element under development by the U.S. Army Medical Research and Development Command for support of field medical units. Methodology for solving the model equations and fitting the model parameters to experimental operating data has been developed and programs written for a digital computer. This article presents in detail the model equations, the parameter fitting procedures, and some typical results.

## DEVELOPMENT OF BASIC EQUATIONS AND METHODS USED

The model equations developed characterize the operation of the duPont B-10 Reverse Osmosis Separator as illustrated in Figure 1. Contaminated water enters via the high pressure feed tube. Flowing in the radial direction, contaminated water becomes progressively more concentrated as water preferentially permeates through the membrane wall into the inner bore of the fibers. Concentrate is collected at the flow screen and moves axially towards the concentrate outlet. Purified water collected inside the fibers exits the module as permeate.

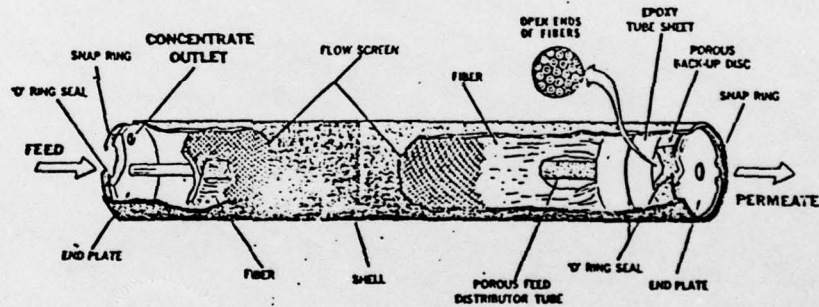


Figure 1. Reverse Osmosis Separator

## FORMULATION OF EQUATIONS

To develop the area available for the flux of water and contaminants into the fiber bore, let  $N_f$  be the number of perfectly straight, evenly distributed fibers parallel to the axial direction per unit of cross sectional area. Collectively, these idealized fibers would have the same fluxes of water and contaminants as the actual fiber configuration.

For the differential element in Figure 2, the area available for flux becomes:

$$(N_f)(\pi D_f L)[\pi(r + \Delta r)^2 - \pi(r)^2]$$

where:  $D_f$  is the outside diameter of the fiber (m)  
 $L$  is the length of the fibers (m)  
 $r$  is the radial coordinate (m)

A steady state material balance on the dissolved solids, component 'A', for the slice of module between  $r$  and  $r + \Delta r$  in Figure 2 yields:

Input rate of A = Output rate of A

$$(2\pi r L)V(r)C_A(r) = 2\pi(r + \Delta r)LV(r + \Delta r)C_A(r + \Delta r) + J_A(r)(N_f \pi D_f L)[\pi(r + \Delta r)^2 - \pi r^2]$$

where:  $V(r)$  = radial velocity of fluid (m/hr)

$C_A(r)$  = concentration of A (gm/m<sup>3</sup>)

$J_A(r)$  = flux of A into inner channel of hollow fibers (gm/hr-m<sup>2</sup>)

A similar steady state material balance on the TOC, component 'C', for the differential element in Figure 2 yields:

$$(2\pi r L)V(r)C_C(r) = 2\pi(r + \Delta r)LV(r + \Delta r)C_C(r + \Delta r) + J_C(r)(N_f \pi D_f L)[\pi(r + \Delta r)^2 - \pi r^2]$$

where:  $C_C(r)$  = Total Organic Carbon (TOC) concentration (gm/m<sup>3</sup>)

$J_C(r)$  = flux of C into inner channel of hollow fibers (gm/hr-m<sup>2</sup>)

A steady state total material balance yields:

$$(2\pi r L)V(r)\rho(r) = [2\pi(r + \Delta r)L]V(r + \Delta r)\rho(r + \Delta r) + [J_A(r) + J_B(r) + J_C(r)](N_f \pi D_f L)[\pi(r + \Delta r)^2 - \pi r^2]$$

where:  $\rho(r)$  = total mass density (gm/m<sup>3</sup>)

$J_B(r)$  = flux of water, component B, into the fiber bore (gm/m<sup>2</sup>-hr)

In addition, a steady state material balance on component 'B' can be written to obtain an equation analogous to equation (1). However, this equation is not necessary as it becomes a dependent equation when equations (1), (2), and (3) are used in the model.

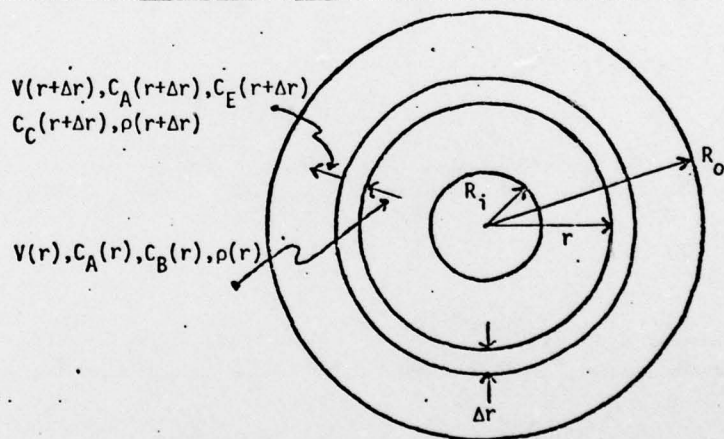


Figure 2. Radial Differential Element

#### ANALYTICAL EQUATIONS.

Equations (1), (2) and (3) are readily converted to differential equations by taking the limit as  $\Delta r$  approaches zero. The results are:

$$\frac{d}{dr} [rV(r)C_A(r)] = -N_f \pi D_F r J_A(r)$$

$$\frac{d}{dr} [rV(r)C_C(r)] = -N_f \pi D_F r J_C(r)$$

$$\frac{d}{dr} [rV(r)\rho(r)] = -N_f \pi D_F r [J_A(r) + J_B(r) + J_C(r)]$$

The boundary conditions are as follows:

$$V(R_i) = F_0 / (2\pi R_i L)$$

$$C_A(R_i) = C_{A0}$$

$$C_C(R_i) = C_{C0}$$

$$\rho(R_i) = \text{calculated from } C_A(R_i) \text{ and } C_C(R_i)$$

where:  $F_0$  = feed rate to module,  $m^3/hr$

$C_{A0}$  = inlet concentration of A,  $gm/m^3$

$C_{C0}$  = inlet TOC concentration,  $gm/m^3$

#### FLUXES AT THE BOUNDARY LAYER

To obtain the fluxes  $J_A$ ,  $J_B$ , and  $J_C$ , a stagnant boundary layer is imagined to exist near outer surface of the hollow fiber as shown in Figure 3. The following fluxes into and out of the boundary layer are defined:

$$J_A = B(C_{A2} - C_{A3}) \quad (7)$$

$$J_{DA} = k_A(C_{A2} - C_{A1}) \quad (10)$$

$$J_B = \gamma(\Delta P - \Delta \pi) \quad (8)$$

$$J_{DB} = k_B(C_{B1} - C_{B2}) \quad (11)$$

$$J_C = C(C_{C2} - C_{C3}) \quad (9)$$

$$J_{DC} = k_C(C_{C2} - C_{C1}) \quad (12)$$

where:  $J_{DA}$  = diffusional flux of contaminant out of the boundary layer,  $gm/m^2-hr$

$J_{DB}$  = diffusional flux of water into the boundary layer,  $gm/m^2-hr$

$J_{DC}$  = diffusional flux of TOC out of the boundary layer,  $gm/m^2-hr$

$C_{A1}$  = concentration of A in bulk fluid,  $gm/m^3$

$C_{A2}$  = concentration of A in boundary layer,  $gm/m^3$

$C_{A3}$  = concentration of A in permeate,  $gm/m^3$

$C_{B1}$  = concentration of B in bulk fluid,  $gm/m^3$

$C_{B2}$  = concentration of B in boundary layer,  $gm/m^3$

$C_{B3}$  = concentration of B in permeate,  $gm/m^3$

$C_{C1}$  = concentration of C in bulk fluid,  $gm/m^3$

$C_{C2}$  = concentration of C in boundary layer,  $gm/m^3$

$C_{C3}$  = concentration of C in permeate,  $gm/m^3$

$\gamma$  = permeability coefficient for pure water,  $gm/m^2-hr-atm$

$B$  = permeability coefficient for dissolved solids,  $m/hr$

$C$  = permeability coefficient for TOC,  $m/hr$

$\Delta P$  = pressure drop across membrane,  $atm$

$\Delta \pi$  = osmotic pressure difference across membrane =  $\pi_2 - \pi_3$ ,  $atm$

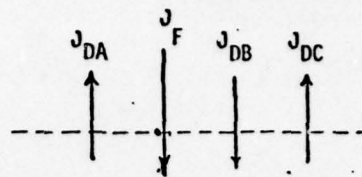
$\pi_2$  = osmotic pressure at boundary layer,  $atm$

$\pi_3$  = osmotic pressure of permeate,  $atm$

$k_A, k_C$  = mass transfer coefficients for solids and TOC, respectively,  $m/hr$

Bulk Fluid

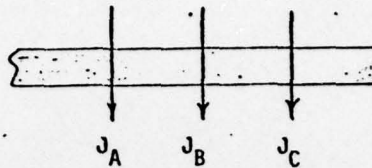
$C_{A1}, C_{B1}, C_{C1}$



Boundary Layer

$C_{A2}, C_{B2}, C_{C2}$

Fiber Membrane



Permeate

$C_{A3}, C_{B3}, C_{C3}$

Figure 3. Fluxes into and out of boundary layer

The osmotic pressure is related to the concentration of contaminant,  $C_E$ , and the absolute temperature,  $T$ , by the empirical relationship

$$\pi = f(C_E, T) = \alpha T C_E (1 + \beta C_E)^2 \quad (13)$$

where:  $\alpha$  = coefficient,  $\text{atm}\cdot\text{m}^3/\text{gm}\cdot^\circ\text{K}$

$\beta$  = coefficient,  $\text{m}^3/\text{gm}$

$T$  = absolute temperature, degrees Kelvin

$C_E$  = effective contaminant concentration =  $C_A + k_E C_C$ ,  $\text{gm}/\text{m}^3$

$k_E$  = coefficient expressing the concentration of A equivalent to a unit concentration of C for purposes of osmotic pressure, dimensionless

To obtain  $\Delta\pi = \pi_2 - \pi_3$ , it is necessary to evaluate  $\pi_2$  using  $C_{A2}$  and  $C_{C2}$ ;  $\pi_3$  using  $C_{A3}$  and  $C_{C3}$ .

The flux  $J_F$  due to the bulk flow toward the boundary layer can be obtained by writing a material balance around the boundary layer:

$$J_F = J_A + J_B + J_C + J_{DA} - J_{DB} + J_{DC} \quad (14)$$

In addition, component balances for component A and for component C can be written:

$$J_F X_{A1} = J_A + J_{DA} \quad (15)$$

$$J_F X_{C1} = J_C + J_{DC} \quad (16)$$

where  $X_{A1}$ ,  $X_{C1}$  = mass fractions of dissolved solids and TOC in bulk stream, respectively.

A component balance can also be written for component B, but it will not be an independent equation.

In addition, the mass fractions  $X_{A3}$  and  $X_{C3}$  are related to  $J_A$ ,  $J_B$ , and  $J_C$  as follows:

$$X_{A3} = \frac{J_A}{J_A + J_B + J_C} \quad (17)$$

$$X_{C3} = \frac{J_C}{J_A + J_B + J_C} \quad (18)$$

Correlations for the mass transfer coefficients,  $k_A$  and  $k_C$ , have been developed analogous to those available for flow across heat exchanger tube bundles. The final result is:

$$k = C_1 (N_{Re})^{e_1} \quad (19)$$

$$N_{Re} = \frac{D_0 V}{\nu} \quad (20)$$

where:  $N_{Re}$  = Reynolds number  
 $D_o$  = outer diameter of fiber  
 $v$  = radial velocity  
 $\nu$  = kinematic viscosity  
 $k$  = mass transfer coefficient

$C_1$  = coefficient which is a function of various physical constants  
 $e_1$  = exponent of the Reynolds number

The same values for the exponent  $e_1$  will be used for both the solids and TOC but different values will be used for  $C_1$ , owing to different physical properties of the two components.

#### APPROXIMATIONS IN THE FLUX EQUATIONS

In the reverse osmosis unit, the concentrations of dissolved solids and TOC will be relatively small. Furthermore, the RO membrane will tend to reject these components. For these reasons the following assumptions will be made:

1. The membrane permeability fluxes  $J_A$  and  $J_C$  are small compared to  $J_B$ .
2. The diffusional fluxes  $J_{DA}$ ,  $J_{DB}$ , and  $J_{DC}$  are small compared to  $J_B$ .
3. The density is that of water namely  $\rho$ .

Table I gives the flux equations with these approximations included.

Table I. Simplified Equations for Fluxes at the Boundary Layer

Transport Equations	Osmotic Pressure Relationships
$J_A = B(C_{A2} - C_{A3})$	$\Delta\pi = \pi_2 - \pi_3$
$J_B = \gamma(\Delta P - \Delta\pi)$	$\pi_2 = \alpha TC_{E2}(1 + BC_{E2})^2$
$J_C = C(C_{C2} - C_{C3})$	$\pi_3 = \alpha TC_{E3}(1 + BC_{E3})^2$
$J_{DA} = k_A(C_{A2} - C_{A1})$	$C_{E2} = C_{A2} + k_E C_{C2}$
$J_{DC} = k_C(C_{C2} - C_{C1})$	$C_{E3} = C_{A3} + k_E C_{C3}$

#### Material Balance at Boundary Layer

$$J_F = J_B$$

$$J_F X_{A1} = J_A + J_{DA}$$

$$J_F X_{C1} = J_C + J_{DC}$$

$$X_{A3} = \frac{J_A}{J_B} \quad X_{C3} = \frac{J_C}{J_B}$$

#### Mass Fraction/Density/Concentration Relationships

$$X_{B2} = 1 - X_{A2} - X_{C2} \quad X_{B3} = 1 - X_{A3} - X_{C3}$$

$$C_{A2} = \rho X_{A2} \quad C_{A3} = \rho X_{A3}$$

$$C_{B2} = \rho X_{B2} \quad C_{B3} = \rho X_{B3}$$

$$C_{C2} = \rho X_{C2} \quad C_{C3} = \rho X_{C3}$$

#### SOLUTION METHOD

The solution method basically consists of numerically integrating the differential equations resulting from the total and component material balances. However, this is somewhat complicated by the fact that the fluxes must be determined at each increment by solving the set of nonlinear equations summarized in Table I.

#### PARAMETER ESTIMATION CONSIDERATIONS

Values of several key variables, such as mass transfer coefficients and osmotic pressure relation coefficients, must be determined from experimental data. A Pattern Search strategy was used to find a set of values for these parameters which minimized a cost function expressive of the difference between model calculations and experimental results.

Table II lists the parameters whose values have been obtained by the parameter estimation procedure. The parameters  $N_f$ ,  $\gamma$ ,  $B$ , and  $C$  may only be obtained from the operation of the module itself.

Of the remaining parameters, three of these ( $C_{1A}$ ,  $C_{1C}$ , and  $e_1$ ) are required for the mass transfer relationships and three ( $\alpha$ ,  $\beta$ , and  $k_E$ ) are required for the osmotic pressure relationship. It should be possible to develop experimental procedures to evaluate these independent of the RO module test, but in the absence of these independent experiments, these six parameters are also evaluated from the RO test data.

Table II Model Parameters

$N_f$	Number of ideal fibers per unit cross-sectional area of the fiber bundle
$\gamma$	Permeability coefficient for pure water
$B$	Permeability coefficient for dissolved solids
$C$	Permeability coefficient for TOC
$e_1$	Exponent of Reynolds number in transfer correlation
$\alpha$	Coefficient in osmotic pressure correlation
$\beta$	Coefficient in osmotic pressure correlation
$k_E$	Mass of dissolved solids equivalent to a unit mass of TOC for use in the osmotic pressure correlation
$C_{1A}$	Coefficient in mass transfer correlation for A
$C_{1C}$	Coefficient in mass transfer correlation for C

## COST FUNCTION

The test data basically consists of two types:

1. Permeate flux data
2. Permeate concentration data (dissolved solids and TOC)

The above values are read directly or computed from the experimental data.

The cost function employed is the following:

$$\text{COST} = \sum_{i=1}^n Z_i \left[ W_{Fi} \left( \frac{F_{e,i} - F_{m,i}}{F_{e,i}} \right)^2 + W_{Di} \left( \frac{C_{Ae,i} - C_{Am,i}}{C_{Ae,i}} \right)^2 + W_{Ti} \left( \frac{C_{Ce,i} - C_{Cm,i}}{C_{Ce,i}} \right)^2 \right]$$

where:  $Z_i$ ,  $W_{Fi}$ ,  $W_{Di}$ , and  $W_{Ti}$  are weighting factors for, respectively, the point as a whole, permeate flux, permeate solids concentration, and permeate TOC concentration.

$C_A$ ,  $C_C$  represent permeate dissolved solids and TOC concentrations, respectively. Subscripts  $e$  and  $m$  indicate experimental data and model predictions, respectively.

Table III provides a comparison between a typical data point and model predictions for the same feed and operating conditions after having determined values of the model parameters as outlined above. (Module pressure = 54.37 atm, temperature = 302.78°K)

Table III. Comparison of Experimental Data and Model Predictions

	Feed	Permeate	Concentrate	Permeate	Concentrate
Flow rate (m <sup>3</sup> /hr)	1.02	0.36	0.66	0.335	0.66
Solids concentration (gm/m <sup>3</sup> )	12100.0	104.0	18600.0	105.0	18000.0
TOC concentration (gm/m <sup>3</sup> )	581.0	31.0	881.0	30.0	850.0

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A Mathematical Model of an Ultrafiltration System for Water Purification Use

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**Abstract**

This paper describes the development of a mathematical model depicting the ultrafiltration unit employed in the water processing element under development by the U.S. Army Medical Research and Development Command for support of field medical units. Methodology for solving the model equations and fitting the model parameters to experimental operating data has been developed and programs written for a digital computer. This article presents in detail the model equations, the solution techniques employed, the parameter fitting procedures, and some typical results.

**Development of Basic Equations**

The model equations developed characterize the operation of the Abcor HFD ultrafiltration unit shown in Fig. 1. Contaminated water enters the tube at  $z=0$  and flows in the  $z$  direction, exiting at  $z=L$ , and is collected as concentrate. While in the tube, some of the water and contaminants pass through the membrane attached to the tube's inner surface and is collected as permeate.

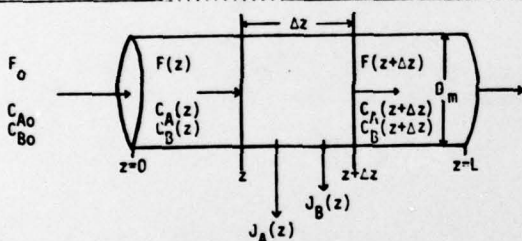


Fig. 1. Abcor HFD Tubular Ultrafiltration Unit

- $C_{A0}$  - inlet concentration of contaminant;  $gm/m^3$
- $C_{B0}$  - inlet concentration of water;  $gm/m^3$
- $D_m$  - diameter of module; m
- $F_0$  - inlet flow rate;  $m^3/hr$
- $J_A$  - flux of contaminant across membrane;  $gm/m^2-hr$
- $J_B$  - flux of water across membrane;  $gm/m^2-hr$
- $L$  - length of module; m
- $z$  - axial spacial coordinate; m

Each ultrafiltration tube is modeled as a long hollow cylinder with a semi-permeable membrane adhering to the inner surface of the tube. The membrane acts as an extra fine mesh, retaining suspended solids and some very large organics, but passing water and much of the dissolved solids. During operation, a thin boundary layer is assumed to form at the membrane through which water must pass before reaching the tube wall. Detailed equations derived from writing component and overall material balances for the bulk and boundary layer sections of the tube form the basis of the model.

For the present work, the model is only concerned with the suspended solids and the dissolved solids in the feed. Experimental data indicates that both have an effect on the flux of water and dissolved solids through the membrane, the permeate being essentially free of any suspended solids. As presently written, the model assumes that the effect of suspended solids and dissolved solids on the flux is the same, which permits the dissolved and suspended solids to be lumped into a single contaminant, 'A', for the feed and concentrate streams. As more experimental data becomes available, it may be possible to dis-

tinguish between the effects of dissolved solids and suspended solids on the fluxes.

A steady state material balance on the total solids, component 'A', for the differential element in Fig. 1 yields:

Input rate of A = Output rate of A

$$F(z) C_A(z) = F(z + \Delta z) C_A(z + \Delta z) + \pi D_m J_A(z) \Delta z \quad (1)$$

where:  $F(z)$  = fluid volumetric flow rate ( $m^3/hr$ )  
 $C_A(z)$  = concentration of A ( $gm/m^3$ )  
 $J_A(z)$  = flux of A through membrane ( $gm/hr-m^2$ )

As the permeate contains no suspended solids,  $J_A$  must consist entirely of dissolved solids.

Similarly, a steady state total material balance on the differential element in Fig. 1 yields:

Input rate = Output rate

$$F(z) \rho = F(z + \Delta z) \rho + \pi D_m [J_A(z) + J_B(z)] \Delta z \quad (2)$$

where:  $\rho$  = total mass density ( $gm/m^3$ )  
 $J_B(z)$  = flux of water, component B, through the membrane ( $gm/m^2-hr$ )

In addition, a steady state material balance on component 'B' can be written to obtain an equation analogous to equation (1). However, this equation is not necessary as it becomes a dependent equation when equations (1) and (2) are used in the model.

Equations (1) and (2) are readily converted to differential equations by taking the limit as  $\Delta z$  approaches zero. For equation (1), the result is:

$$\frac{d}{dz} [F(z) C_A(z)] = - \pi D_m J_A(z) \quad (3)$$

Equation (2) becomes:

$$\frac{dF(z)}{dz} = - \frac{\pi D_m}{\rho} [J_A(z) + J_B(z)] \quad (4)$$

The boundary conditions are as follows:

$$F_0(0) = F_0 \quad C_A(0) = C_{A0}$$

where:  $F_0$  = feed rate to module,  $m^3/hr$   
 $C_{A0}$  = inlet concentration of A,  $gm/m^3$

**Fluxes at the Boundary Layer**

To obtain the fluxes  $J_A$  and  $J_B$ , a stagnant boundary layer is imagined to exist near the inner wall of the tube as shown in Fig. 2. The following fluxes into and out of the boundary layer are defined:

$$J_A = B(C_{A2} - C_{A3}) \quad (5)$$

$$J_B = \gamma(\Delta P - \Delta\pi) \quad (6)$$

$$J_{DA} = k_A(C_{A2} - C_{A1}) \quad (7)$$

$$J_{DB} = k_B(C_{B1} - C_{B2}) \quad (8)$$

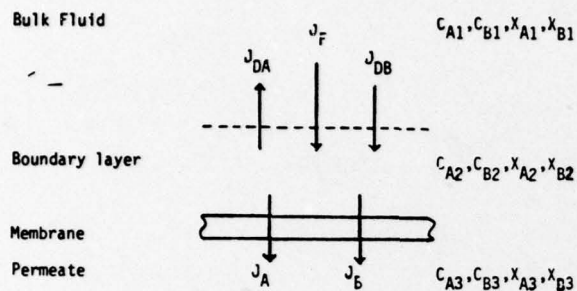


Fig. 2. Fluxes into and out of Boundary Layer

where:  $J_{DA}$  = diffusional flux of contaminant out of the boundary layer, gm/m<sup>2</sup>-hr  
 $J_{DB}$  = diffusional flux of water into the boundary layer, gm/m<sup>2</sup>-hr  
 $C_{A1}$  = concentration of A in bulk fluid, gm/m<sup>3</sup>  
 $C_{A2}$  = concentration of A in boundary layer, gm/m<sup>3</sup>  
 $C_{A3}$  = concentration of A in permeate, gm/m<sup>3</sup>  
 $C_{B1}$  = concentration of B in bulk fluid, gm/m<sup>3</sup>  
 $C_{B2}$  = concentration of B in boundary layer, gm/m<sup>3</sup>  
 $C_{B3}$  = concentration of B in permeate, gm/m<sup>3</sup>  
 $\gamma$  = permeability coefficient for pure water, gm/m<sup>2</sup>-hr-atm  
 $B$  = permeability coefficient for dissolved solids, m/hr  
 $\Delta P$  = pressure drop across membrane, atm  
 $\Delta \pi$  = osmotic pressure difference across membrane =  $\pi_2 - \pi_3$ , atm  
 $\pi_2$  = osmotic pressure at boundary layer fluid, atm  
 $\pi_3$  = osmotic pressure of permeate, atm  
 $k_A, k_B$  = mass transfer coefficients for solids and water, respectively, m/hr

The osmotic pressure is related to the concentration of contaminant,  $C_A$  and the absolute temperature,  $T$ , by the empirical relationship

$$\pi = f(C_A, T) = \alpha TC_A(1 + \beta C_A)^2 \quad (9)$$

where:  $\alpha$  = coefficient, atm-m<sup>3</sup>/gm<sup>0</sup>-K  
 $\beta$  = coefficient, m<sup>3</sup>/gm  
 $T$  = absolute temperature, deg Kelvin

To obtain  $\Delta \pi = \pi_2 - \pi_3$ , it is necessary to evaluate  $\pi_2$  using  $C_{A2}$  and  $\pi_3$  using  $C_{A3}$ .

The flux  $J_F$  due to the bulk flow toward the boundary layer can be obtained by writing a total material balance around the boundary layer:

$$J_F = J_A + J_B + J_{DA} - J_{DB} \quad (10)$$

In addition, a component balance for component A can be written:

$$J_F X_{A1} = J_A + J_{DA} \quad (11)$$

where  $X_{A1}$  = mass fraction of dissolved solids in the bulk stream.

A component balance can also be written for component B, but it will not be an independent equation.

In addition,  $X_{A3}$ , the mass fraction of dissolved solids in the permeate is related to  $J_A$  and  $J_B$  as follows:

$$X_{A3} = \frac{J_A}{J_A + J_B} \quad (12)$$

This completes the set of equations describing the fluxes at the boundary layer.

In the ultrafiltration unit, the concentration of dissolved solids will be relatively small. Furthermore, the membrane will tend to reject these components. For these reasons, the following assumptions will be made:

1. The membrane permeability flux  $J_A$  is small compared to  $J_B$ .
2. The diffusional fluxes  $J_{DA}$  and  $J_{DB}$  are small compared to  $J_B$ .
3. The density is that of water.

Table I presents the flux equations with these approximations included. Observe that it is no longer necessary to evaluate  $J_{DB}$ .

Table I

Simplified Equations for Fluxes at the Boundary Layer

Transport Equations	Osmotic Pressure Relationships
$J_A = B(C_{A2} - C_{A3})$	$\Delta \pi = \pi_2 - \pi_3$
$J_B = \gamma(\Delta P - \Delta \pi)$	$\pi_2 = \alpha TC_{A2}(1 + \beta C_{A2})^2$
$J_{DA} = k_A(C_{A2} - C_{A1})$	$\pi_3 = \alpha TC_{A3}(1 + \beta C_{A3})^2$

Material Balance at Boundary Layer

$$J_F = J_B$$

$$J_F X_{A1} = J_A + J_{DA}$$

$$X_{A3} = \frac{J_A}{J_B}$$

Mass Fraction/Density/Concentration Relationships

$$X_{B2} = 1 - X_{A2} \quad X_{B3} = 1 - X_{A3}$$

$$C_{A2} = \rho X_{A2} \quad C_{A3} = \rho X_{A3}$$

$$C_{B2} = \rho X_{B2} \quad C_{B3} = \rho X_{B3}$$

#### Solution Method

The solution method basically consists of numerically integrating the differential equations (3) and (4) resulting from the total and component material balances. However, this is somewhat complicated by the fact that the fluxes must be determined at each increment by solving the set of nonlinear equations summarized in Table I.

The computation of  $J_A$  and  $J_B$  at every point  $z$  requires the solution of the set of equations in Table I, which entails two nested iteration loops.

For both loops, the interval halving method was found to be necessary to achieve a reliable convergence of the loops. A similar situation exists in the reverse osmosis unit downstream of the ultrafiltration unit. The equations proved to be highly nonlinear, and with varying degrees of sensitivity. Consequently, the reliability of the interval halving method had to take precedence over computational efficiency.

For the first or outer iteration loop, a value of  $C_{A2}$  must

be estimated. It is known that  $C_{A1} \leq C_{A2} \leq \rho$ , which permits interval halving to be utilized.

However, knowing the value of  $C_{A2}$  only permits  $J_{DA}$  to be calculated. To proceed further, a value is assumed for  $C_{A3}$ , which must be in the interval  $0 \leq C_{A3} \leq C_{A1}$ . This permits  $J_B$  to be readily calculated as follows:

$$J_B = k_A \rho \left( \frac{C_{A2}}{C_{A3}} - 1.0 \right) \quad (13)$$

by combining the following three equations:

$$X_{A3} = J_A / J_B$$

$$C_{A3} = \rho X_{A3}$$

and  $J_A = k_A (C_{A2} - C_{A3})$ .

Harriott and Hamilton (2) give the following relationship for tubular ultrafiltration modules:

$$N_{Sh} = 0.0096 N_{Re}^{0.913} N_{Sc}^{0.346} \quad (14)$$

where:  $N_{Sh} = \frac{kD}{D_{AB}}$  = Sherwood number

$$N_{Re} = \frac{DV}{\nu} = \text{Reynolds number}$$

$$N_{Sc} = \frac{\nu}{D_{AB}} = \text{Schmidt number}$$

where:  $k$  = mass transfer coefficient, m/hr  
 $\nu$  = kinematic viscosity,  $m^2/hr$   
 $D_{AB}$  = diffusivity,  $m^2/hr$   
 $D$  = characteristic length, m

This relationship was incorporated directly into the model. However in attempting to improve the accuracy of the model, the relationship

$$D_{AB} = aX_{A1} + b \exp(-cX_{A1})$$

was used to make the diffusivity a function of concentration where  $a$ ,  $b$ , and  $c$  are coefficients.

Now that values are available for  $C_{A2}$  and  $C_{A3}$ , the osmotic pressures  $\pi_2$  and  $\pi_3$  can be computed, and then  $J_B^*$  can be evaluated using

$$\Delta\pi = \pi_2 - \pi_3$$

and

$$J_B^* = (\Delta P - \Delta\pi).$$

If the values of  $J_B$  and  $J_B^*$  do not agree, the value of  $C_{A3}$  must be changed. Using the interval halving technique, the interval of uncertainty for  $C_{A3}$  is reduced and the computations repeated.

Once a value of  $C_{A3}$  is obtained that gives the same value for  $J_B$  and  $J_B^*$ , the assumption for  $C_{A2}$  must be checked. Since values are available for  $J_B$ ,  $J_A$ , and  $J_{DA}$ , we can compute  $C_{A2}^*$  as follows:

$$J_B X_{A1} = k_A (C_{A2}^* - C_{A1}) + B (C_{A2}^* - C_{A3})$$

rewritten as

$$C_{A2}^* = \frac{J_B X_{A1} + k_A C_{A1} + B C_{A3}}{k_A + B}$$

If  $C_{A2}^*$  is not equal to  $C_{A2}$ , a new value for  $C_{A2}$  must be assumed. The interval halving logic permits the interval of uncertainty for  $C_{A2}$  to be reduced and the calculations repeated.

Using this procedure, the nonlinear flux equations are solved for each integration step.

#### Parameter Estimation Considerations

Values of several key variables, such as mass transfer coefficients and osmotic pressure correlation coefficients, must be determined from experimental data. A Pattern Search strategy (4) was used to find a set of values for these key variables which minimized the difference between model calculations and experimental results.

The Pattern Search technique was chosen for several reasons:

- Simplicity
- Ease of use
- Relative insensitivity to numerical error, i.e., no partial derivatives are evaluated
- Reasonably fast convergence to the solution
- The method is fairly well known

Table II lists the parameters whose values have been obtained by the parameter estimation procedure.

Table II

Fit Parameters

$\gamma$	Permeability coefficient for pure water
B	Permeability coefficient for A
$\alpha$	Coefficient in osmotic pressure correlation
$\beta$	Coefficient in osmotic pressure correlation
a,b,c	Coefficients in the diffusivity relationship

The experimental data on which the parameters were fit was essentially a "snapshot" of a steady-state condition for the UF unit. The following information was available:

1. Specifics of Module Configuration and Geometry
  - a. Length (m)
  - b. Inner diameter (m)
  - c. Inner surface area of module available for flux ( $m^2$ )
2. Operating Test Data
  - a. Inlet volumetric flow rate ( $m^3/hr$ )
  - b. Inlet contaminant concentration ( $gm/m^3$ )
  - c. Inlet pressure (atm)
  - d. Outlet pressure (atm)
  - e. Temperature (°K)
  - f. Average flux of contaminant ( $gm/m^2-hr$ )
  - g. Average flux of water ( $gm/m^2-hr$ )

Table III lists one set of the experimental data that was available.

Table III  
Experimental Data

Diameter:	0.0254 m
Inner Surface Area:	1.64 m <sup>2</sup>
Length:	24.4 m
Feed Flow Rate:	6.81 m <sup>3</sup> /hr

Inlet Concentration (gm/m <sup>3</sup> )	Permeate Flux (Kg/m <sup>2</sup> -hr)	Permeate Concentration (gm/m <sup>3</sup> )
1240	226.	517
2090	153.	869
7620	83.2	1560
13400	71.3	2120

In fitting the data, the cost function employed was the following:

$$\text{COST} = \sum_{i=1}^n z_i \left[ W_{Fi} \left( \frac{F_{e,i} - F_{m,i}}{F_{e,i}} \right)^2 + W_{Di} \left( \frac{C_{Ae,i} - C_{Am,i}}{C_{Ae,i}} \right)^2 \right]$$

where: COST = cost function to be minimized

- $i$  = data point identifier
- $n$  = number of data points
- $z_i$  = weighting factor of point  $i$  relative to other data points
- $W_{Fi}$  = weighting factor for flux for data point  $i$  ( $0 \leq W_{Fi} \leq 1$ )
- $W_{Di}$  = weighting factor for contaminant concentration for data point  $i$  ( $0 \leq W_{Di} \leq 1$ )
- $F_{e,i}$  = experimental value of permeate flux for data point  $i$
- $F_{m,i}$  = permeate flux predicted by model for data point  $i$
- $C_{Ae,i}$  = experimental permeate dissolved solids concentration for data point  $i$
- $C_{Am,i}$  = permeate contaminant concentration predicted by model for data point  $i$

In addition to the requirement that the weighting factors  $W_{Fi}$  and  $W_{Di}$  must each be less than 1.0, they should logically sum to 1.0 for each point  $i$ :

$$W_{Fi} + W_{Di} = 1.0$$

Table IV illustrates the quality of the fit to the data given in Table III. The weighting factor  $z_i$  was 1.0 for each point, with  $W_{Fi}$  and  $W_{Di}$  being 0.8 and 0.2, respectively, for each point.

Table IV  
Model Fit

Data	Permeate Flux (Kg/m <sup>2</sup> -hr)		Permeate Concentration (gm/m <sup>3</sup> )		Error
	Model	Error	Data	Model	
226.	216.	-4.4%	517	476	-7.9%
153.	161.	+5.2%	869	766	-11.8%
83.2	89.3	+7.3%	1560	1637	+4.9%
71.2	65.6	-8.0%	2120	2336	+10.2%

#### Continuing Work

Present efforts are being directed toward incorporating TOC (Total Organic Carbon) relationships into the model, but work is awaiting the availability of experimental data on TOC. At that time, additional efforts will be directed toward improving the model fit beyond that illustrated in Table IV. The ultrafiltration model is also being combined with models for reverse osmosis, ozonation, and hypochlorination to provide a comprehensive model for the water processing unit.

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