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# **MODELING ULTRAFILTRATION AND FILTRATION PHENOMENA APPLIED IN CHEMICAL PULPING PROCESSES**

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**Kari Kovasin**

Dissertation for the degree of Doctor of Science in Technology to be presented with due permission for public examination and debate in Auditorium Ke 2 at Helsinki University of Technology (Espoo, Finland) on the 7th of February, 2003, at 12 o'clock.

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## ABSTRACT

This thesis is based on seven publications dealing with separation processes applied in the manufacturing of chemical pulp. The first part discusses the correlation of mass transfer phenomena in the ultrafiltration of spent cooking liquor. The second part focuses on the fiber mat formation that takes place in pulp washers and thickeners.

The main interest throughout the thesis is to develop useful and scientifically justified methods for correlating mass transfer and filtration mechanisms in order to interpret the phenomena taking place in the studied unit operations. The methods presented can be used in the designing and dimensioning of industrial-scale equipment and systems.

Two types of basic mass transfer models, the film model and a more complete approach utilizing an integral solution for two-dimensional diffusion, have been introduced for the calculation of the macrosolute concentration at the surface of the ultrafiltration membrane. A pilot-scale test rig was used to measure ultrafiltration of spent cooking liquor. Mass transfer correlations to estimate the permeate flux have been developed based on both models to be used for plant design calculations. The modeling studies indicated that the integral solution method gave a physically more realistic behavior for the macrosolute concentration at the membrane wall than the widely used film model.

It was further shown that dynamic programming may be effectively used to optimize a multistage industrial-scale ultrafiltration-diafiltration process. This method required a shorter calculation time and resulted in slightly better optimal conditions than the direct search algorithm.

The studies on mat formation during pulp washing showed that the washing tester is a useful tool to obtain the required information for the washer. A calculation procedure has been developed to obtain design parameters for the dimensioning of industrial pulp washers. The suggested theory was based on Darcy's law under the conditions of constant pressure filtration of incompressible beds and reasonable accuracy was obtained for purposes of washer design. A more accurate approach for the modeling of the mat formation stage was formulated on the fundamental filtration theory of compressible fiber beds. Two such methods were discussed. In the more general model of mat formation, the equation of continuity during filtration was also taken into account. This model was then used for evaluating the effect of various operational parameters on drum washer performance. This model was also able to predict earlier generated experimental test data rather well. A new method for considering the additional filtration resistance due to suspended gases (e.g. air) was also suggested.

*“For what is a man, what has he got  
If not himself, then he has naught  
To say the things he truly feels  
And not the words of one who kneels  
The record shows I took the blows  
And did it my way”*

“I did it may way” – A song by Frank Sinatra

## Preface

The publications that this thesis refers to cover a rather long time period, starting from 1984 and ending in the current year. In fact, a significant part of the work was carried out during the time I was working for Rauma-Repola Oy in Rauma and Pori. Such a long time span may raise questions about the relevancy of scientific efforts dating back nearly 20 years. Considering the focus of the thesis, namely discussing a few chemical engineering applications in the connection of pulp manufacturing processes, the studied topics may be even more interesting today than they were when the work started. The utilization of membrane separation processes is increasing in the pulp industry. Furthermore, the correct dimensioning of pulp washers calls for accurate methods since the maximum capacities of pulping lines have grown nearly 200 % compared with the time when my work in this area began.

Over the years, many people have contributed to my efforts to focus on the chemical engineering applications to be used in pulping technology. First of all, I wish to express my deepest gratitude to Professor Harry V. Nordén, who was my teacher of Chemical Engineering at HUT. He was a demanding tutor but he also opened my eyes to the significance of pulping applications. I am also very grateful to Ismo Reilama who has been advising me since the beginning of my career in the industry. He has also encouraged me to use my own brain and think in an unconventional way in order to solve technical problems – both of them most valuable pieces of advice!

I would like to express my warm thanks to all my colleagues and co-workers from the days of the lignosulfonate pilot plant up to the present in SciTech-Service. There is a long list of people who have assisted in various phases of this thesis to a larger or smaller extent. Some of them are sadly no longer with us.

My special thanks are due to a group of professors who have positively contributed to my scientific work and thinking over the years. Professors Charles G. Hill Jr. and Richard Hughes tutored me during my Asla-Fulbright scholarship year at the University of Wisconsin. I am also very grateful to Professor Juhani Aittamaa who encouraged me to finalize the thesis. The example of Professor Panu Tikka is also worth acknowledging because he has shown that endurance is also a great virtue when completing a doctoral thesis. Finally, I wish to express my special thanks to Professor Lars Nyström and Dr. Kari Ala-Kaila who performed the pre-examination of this thesis and could greatly improve it by their suggestions.

The support and patience of my family has given me a lot of encouragement and the strength that has helped me to survive. Without such understanding these lines would never have been written.

Rauma, November 19<sup>th</sup>, 2002

Kari Kovasin

## List of publications

This thesis comprises the following publications:

1. Kovasin, K., Nordén, H.V., **Determination of liginosulphonate rejection from test results in the ultrafiltration of spent sulphite liquor**, *Svensk Papperstidning*, **87** (1984) No. 6, R44 – R47.
2. Kovasin, K.K., Hill, C.G., Jr., **A method for correlating permeate flux in the ultrafiltration of macrosolutes**, *Journal of Pulp & Paper Science*, **12** (1986) No. 4, J119-J125.
3. Kovasin, K.K., Hughes, R.R., Hill, C.G., Jr., **Optimization of an ultrafiltration-diafiltration process using dynamic programming**, *Computers and Chemical Engineering*, **10** (1986) No. 2, 107 – 114.
4. Hakamäki, H., Kovasin, K., **The effect of some parameters on brown stock washing: A study made with a pulp tester**, *Pulp & Paper Canada*, **86** (1985) No. 9, T243 – T249.
5. Kovasin, K., Tuomi, A., **Entwicklung der Holzschliffeindickung bei hohen Temperaturen**, *Wochenblatt für Papierfabrikation*, **114** (1986) No. 7, 230 – 234.
6. Kovasin, K., Nordén, H.V., Ahonen, L., **Modelling the filtration stage of a pulp washer**, *Filtration & Separation*, **25** (1988) No 1, 42 – 45.
7. Kovasin, K., Aittamaa, J., **Factors defining the capacity of pulp washers**, *Paperi ja Puu*, accepted for publication on 21.08.2002. To be published in 2003.

## The author's contribution

Publication 1: The idea to start preparing mass transfer related publications based on the experimental ultrafiltration data obtained at the ultrafiltration pilot plant of the Rauma pulp mill came from the author. The author also individually prepared this first article in the series. Prof. Nordén acted as supervisor and advisor during the preparation of the publication.

Publication 2: This article was prepared during the author's visit to the University of Wisconsin – Madison. The idea for the paper was the author's who also individually performed the theoretical formulation and the calculations. Prof. Charles G. Hill Jr., who was the author's tutor during his stay at UW, gave instructions and support for the work and also performed the final proofreading of the manuscript.

Publication 3: This article was also prepared during the year in Wisconsin. The idea to apply dynamic programming for the optimization problem of an ultrafiltration system occurred to the author while participating in a course given by the late Prof. Richard Hughes. The author developed the optimization model and performed the required computations and also prepared the manuscript of the article. Professors Hughes and Hill contributed to the article with valuable instructions and advice.

Publication 4: The idea behind this article was generated at Rauma-Repola Pori Works in order to demonstrate the benefits of the pulp tester. The author was then responsible for planning and supervising the experimental work on which the article is based. The author did the preparatory work for the article. The article is based on a paper prepared and given by Mr. Hannu Hakamäki and the author at the Pulp Washing'83 symposium in Mont Gabriel, PQ, Canada.

Publication 5: This article is based on a paper given by the author. All theoretical considerations on how to interpret the pulp tester results were provided by the author. The experimental work for generating the filtration characteristics of pressure groundwood pulp was also conducted by the author. The author prepared the manuscript with the assistance of the co-author Mr. Alpo Tuomi.

Publication 6: The idea for applying the filtration model by Prof. Nordén for the modeling of the Pro-Feed pressure washer was generated by the author and the author also acted as supervisor of the subsequent M.Sc. thesis of Ms. Leena Ahonen. The author prepared a paper to be first given at the Pulp Washing'87 Symposium. The author finally prepared the manuscript for the article.

Publication 7: The idea for preparing a general filtration model for the mat formation stage of pulp washers came from the author. The author performed all the work required for the publication. Prof. Aittamaa assisted in the final preparation of the article text.



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**NOTATION**

$a$	The volume fraction of gases in the pulp mat, (dimensionless)
$A$	The cross-sectional area of the pulp mat during the mat formation, ( $m^2$ )
$c$	Fiber consistency in the mat, ( $kg/m^3$ )
$c_o$	Sedimentation consistency, ( $kg/m^3$ )
$c_s$	Fiber consistency in the inlet fiber suspension, ( $kg/m^3$ )
$\bar{c}_{mat}$	Average fiber consistency in the formed mat, ( $kg/m^3$ )
$C_b$	Macrosolute concentration in the bulk fluid, ( $kg/m^3$ )
$C_g$	Macrosolute concentration in the gel on membrane surface, ( $kg/m^3$ )
$C_p$	Macrosolute concentration in permeate, ( $kg/m^3$ )
$C_w$	Macrosolute concentration at the membrane surface, ( $kg/m^3$ )
$d_i$	Decision variable of optimization
$g$	Fiber mass per unit area (basis weight), ( $kg/m^2$ )
$g_i^*$	Objective function of the optimization at $i$ th stage, (\$)
$G$	Surface loading (square meter load) of washer, ( $kg/(m^2s)$ , tonne/( $m^2d$ ))
$h$	Length of the piston movement in the pulp tester, (m)
$J_v$	Permeate flux, ( $m^3/(m^2s)$ )
$k_v$	Mass transfer coefficient, (m/s)
$k$	Kozeny factor (dimensionless)
$K$	Permeability, ( $m^2$ )
$K_f$	"Filtration coefficient", ( $m/s^{0.5}$ )
$L$	Thickness of the fiber bed, (m)
$M$	Compressibility parameter, ( $kg/m^3$ )
$n$	Drum speed, (1/s or 1/min)
$N$	Compressibility parameter, (dimensionless)
$p$	Compacting pressure, (kPa)
$\Delta p$	Applied pressure drop of filtration, (kPa)
$Q$	Filtrate flow rate, ( $m^3/s$ )
$R$	Macrosolute rejection, (dimensionless)
$R_{OBS}$	Observed macrosolute rejection, (dimensionless)
$s_i$	Information flow vector at $i$ th stage
$S_o$	Specific surface of fibers, (1/m)
$t$	Time, (s)
$t_m$	Time of mat formation, (s)
$T$	Temperature, ( $^{\circ}C$ )

$u$	Superficial filtrate velocity through the mat, (m/s)
$U_i$	Objective function for optimization of stages 1 to $i$ , (\$)
$v$	Specific volume of fibers, (m <sup>3</sup> /kg)
$V$	Volume of the filtrate, (m <sup>3</sup> )
$y_{air}$	Fraction of air in the pulp suspension, (vol-%)

Greek letters:

$\alpha$	Specific cake resistance, (m/kg)
$\varepsilon$	Porosity of fiber bed, (dimensionless)
$\mu$	Viscosity of the filtrate, (kg/(ms))

Abbreviations:

BDT	Bone dry tonne of pulp
vol-%	Volume percentage of air (gas) in pulp suspension

## INTRODUCTION

Chemical pulping processes comprise a multitude of separation processes and other unit operations for various purposes. In this respect this field of the process industry does not differ from the processes of the chemical industry, which are commonly focused on as the main application area for separation technology and other chemical engineering operations. However, the process- and equipment-related problems of pulping processes (using the principles of chemical engineering) are often considered to be much more difficult than typical chemical engineering problems. There are various reasons for this. One is the fact that many unit processes of chemical pulping are performed in multiphase conditions where especially the liquor phases contain tens or even hundreds of chemical constituents that form highly non-ideal mixtures. In addition, many physical properties of the systems are unknown and difficult to measure or estimate by means of the standard procedures of chemical engineering. For that reason and particularly due to the multiphase – multi-component systems, mathematical modeling becomes complicated with no clever simplifications. This general perception of “impossible chemical engineering problems” may have contributed to the strange notion that pulping technology should not even belong under the umbrella of the science of chemical engineering.

If we take a closer look, it is possible to find lots of examples of chemical engineering applications in connection to pulping processes. Many of them have been recently reviewed by Gullichsen [8]. A great number of them deal with separation processes. For example, the treatment of fiber suspensions involves filtration operations through the entire pulping and papermaking process, after the fibers have first been released from the wood matrix. In this respect, the filtration of fiber suspensions represents a separation process of primary importance for the entire pulp and paper industry. Such filtration phenomena take place in the brownstock and post-oxygen washing systems, in the bleaching area after each bleaching stage, in various thickening operations as well as in the wet end of pulp drying and paper machines. Filtration systems, where the liquid medium is black liquor, are especially challenging due to the varying nature of black liquor and the fact that dispersed air is often present, thus affecting the filtration phenomena. Brownstock washing is a typical unit process of this kind.

Another field of increasing significance from the separation technology point of view can be found in the treatment of pulping process effluents. The pulping process comprises a series of chemical reaction stages where most of the organic material of the wood is dissolved in order to release and further modify the fibers into products of desired physical and chemical properties. During these reaction stages, liquid effluents are formed. The effluents contain a plethora of chemical constituents, most of which can be categorized as macromolecules originating from the lignin, carbohydrate and other organic material in the wood. An overwhelmingly large portion of the dissolved material is separated in the black liquor and further processed into energy and fresh cooking chemicals in the recovery system of the pulp mill. The rest of the dissolved material ends up in the various outgoing effluents of the pulping process and often represents a serious environmental burden.

Recently, a lot of effort has been put into developing separation technologies to purify the effluent streams of pulp mills. The main driving force has been the reduction of the environmental impact. Today the re-use of process waters, i.e. closing the water circulation, is one of the main principles of the sustained development of pulping and papermaking processes.

Another area of interest regarding the separation technology utilized in the pulping processes has been the focus on the manufacturing of various by-products. It is a common practice in the kraft process to separate extractives for by-products, such as tall oil and turpentine. Over the years, serious attempts have been made to generate profitable lignin-based by-products (lignosulfonates and kraft lignin) from the spent

cooking liquors of the various pulping methods. Membrane separation technology (ultrafiltration) has proved to be technically viable. However, uncertain market demand and rather poor economic expectations for such by-products have prevented any real breakthrough for lignin or liginosulfonate by-products. The gradual disappearance of sulfite mills has also meant that the raw material sources for liginosulfonates are nowadays very limited. A vast potential of dissolved lignin in kraft black liquor certainly exists but the current prospects for a profitable by-products business are not promising.

Regarding membrane separation technology, a dramatic development has taken place in the manufacture of new types of synthetic membranes, which are better able to tolerate the harsh chemical and physical conditions in the effluent streams of the pulping process, [9]. As for pulp and paper technology, new ideas for applying ultrafiltration and other membrane separation processes have emerged. The need to reduce water consumption and re-use the effluents has particularly catalyzed efforts to find new targets for membrane separation technology. Such applications as the ultrafiltration of debarking and bleaching effluents, either in order to render the effluents less harmful from the environmental point of view or to reduce the demand for fresh water, have already become mill-scale. In addition, there have been similar projects for treating certain wastewater fractions from mechanical pulping production lines and paper mills, [9].

This thesis comprises two sections covering the modeling of separation processes utilized in the chemical pulping processes. The first part discusses ultrafiltration of lignin-based macromolecules out of the spent liquor of the cooking process. The mass transfer phenomena during the ultrafiltration process are first studied based on two publications. After that, one of the mass transfer models is applied for the design and optimization of an industrial ultrafiltration–diafiltration process plant. The solution of the optimization problem is based on utilizing dynamic programming for stage-wise process configuration.

The second part deals with the study, simulation and modeling of the filtration phenomena of pulp fiber suspensions. First, the laboratory simulation and interpretation of the test results of dewatering pulp suspensions are discussed both from the experimental and theoretical point of view. Finally, fundamental theories of filtration and fluid flow are utilized in developing mathematical models to be applied for the calculation of the mat formation stage in pulp washing equipment.

#### **THE TARGETS OF THE WORK:**

- The modeling of the mass transfer in the ultrafiltration of lignin-based macromolecules is aimed at obtaining reliable correlations for analyzing experimental ultrafiltration data. The purpose of such correlations is the dimensioning of industrial ultrafiltration systems.
- The process design of industrial ultrafiltration systems implies the finding of a set of economically viable design parameters. An industrial ultrafiltration-diafiltration system of the kind discussed here is a multistage process with a large number of variables to be optimized. The aim is to show that dynamic programming is a powerful tool for optimizing such multistage ultrafiltration systems.
- The modeling of the filtration of fiber suspensions is aimed at producing an accurate dimensioning method for selecting the filtration area of industrial pulp washing equipment. For this reason, an experimental filtration testing method is first discussed based on simplified filtration theories. The fundamental approach, which takes the compressible nature of the fiber beds into account, is mainly intended for accurate washer dimensioning purposes but also various operational parameters can be evaluated by means of the model.

# 1 MODELING THE MASS TRANSFER IN THE ULTRAFILTRATION OF SPENT COOKING LIQUOR

## 1.1 BACKGROUND

In the late 1970's and early 1980's, considerable interest and experimental activities were focused on the developing of industrial processes to separate lignosulfonates out of the spent cooking liquor of sulfite pulping processes, see for instance [10]. One of the test fields was the sodium-based sulfite pulp mill of Rauma-Repola Oy located in Rauma, Finland. A semi-industrial ultrafiltration pilot plant was built there and operated for a period of a few years in order to study the separation of high molecular lignosulfonates. The production capacity of the pilot plant was big enough to manufacture sufficiently large quantities of lignosulfonate products to be tested for various applications (e.g. as constituents of oil drilling mud, concrete etc.).

Since the experimental practices were of the appropriate discipline, relevant measurements were also gathered in order to study the mass transfer phenomena taking place during the macromolecule separation in the ultrafiltration modules. These measurements comprise the experimental data used in this study.

### Mass transfer phenomena in the ultrafiltration of macromolecular solutions:

In the analysis of the mass transfer phenomena occurring in ultrafiltration or reverse osmosis, one can consider several alternative approaches. One concept is to utilize pore models and transport equations stemming from irreversible thermodynamics. Such an approach is warranted for modeling the ultrafiltration of solutions containing small molecular solutes, since the main mass transfer resistance in such systems is in the separating membrane. However, in the case of the ultrafiltration of macromolecules, such as lignosulfonates, the mass transfer is controlled by the accumulation of macromolecules at the surface of the membrane on the high-pressure or concentrate side of the membrane. This phenomenon is called concentration polarization and it was first thoroughly discussed by Michaels, [11].

Figure 1 describes the development of the concentration profile of the macrosolute in the boundary layer close to the membrane surface. Convective transport takes the solute from the bulk stream towards the membrane surface. In steady state, this transport is counterbalanced by the diffusion of the solute backwards and away from the membrane surface (because the membrane is mainly impermeable to the solute). This condition can be achieved only if the solute concentration at the membrane wall is higher than that in the bulk solution.

Michaels further presented that in the case of high-molecular solutes, the wall concentration finally reaches the point where a solid gel is formed at the membrane surface. This situation thus represents the upper limit for the permeate flux since the gel concentration ( $C_g$ ) means the highest available solute concentration. By forming the steady state mass balance of the solute in the boundary layer and assuming complete rejection of the solute, it was possible to obtain a simple correlation for the limiting permeate flux, [11]:

$$J_v = k_v \ln \left( \frac{C_g}{C_b} \right) \quad (1)$$

In equation (1),  $k_v$  stands for the mass transfer coefficient and  $C_b$  is the solute concentration in the bulk concentrate. This famous formula by Michaels has also been called the gel polarization model and it has been extensively used in the analysis of experimental macromolecule ultrafiltration data.

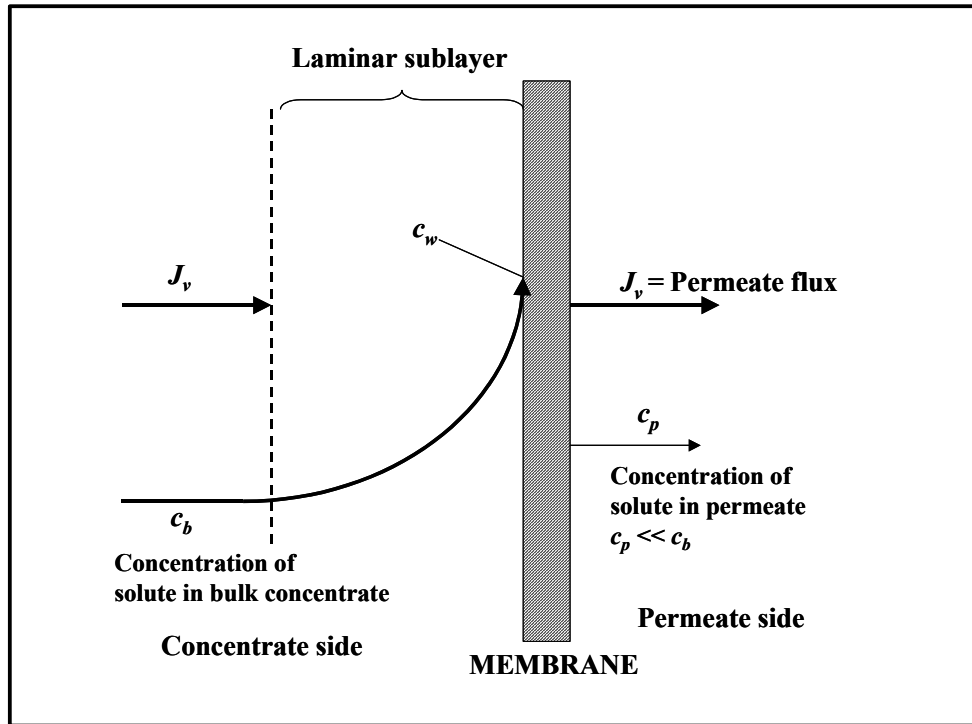


Figure 1: Concentration polarization in ultrafiltration, [1]

The solute concentration does not necessarily always reach the gelling concentration. For this reason, it is more appropriate to express the concentration polarization by means of variable wall concentration ( $C_w < C_g$ ). In addition, the complete rejection of the solute does not always take place either. These statements lead to the film model, presented for example by Goldsmith [12]. The film model, as described by equation (2), can be obtained by solving the one-dimensional diffusion equation in the boundary layer close to the membrane surface.

$$J_v = k_v \ln \left[ \frac{C_w - C_p}{C_b - C_p} \right] \quad (2)$$

Besides the gel polarization model of Michaels, the film model has been one of the most widely utilized mass transfer models for the ultrafiltration of macromolecules.

The gel polarization model of equation (1) was found to predict correctly the qualitative behavior of macromolecular ultrafiltration. However, the quantitative comparison with experimental data was less satisfactory, as pointed out by Shen and Probstein [13]. The reason for this was the use of the constant diffusivity of the macromolecular solute as well as estimating the diffusivity without sufficient accuracy. In order to overcome this shortcoming, several studies emerged in the 1970's. For example, Kozinski and Lightfoot, [14] introduced a mass transfer model based on the fundamental equations of motion, species continuity and species flux expressions for a particular set of boundary conditions. They found that it is important to consider the diffusivity of the solute in the boundary layer as dependent on the solute concentration. The concentration-dependent diffusion coefficient was also considered by Shen and Probstein, [13]. In addition to that, they extended the governing diffusion equation to consider the axial convection of the solute also (parallel to the membrane surface in the direction of the concentrate flow). The solution of the diffusion equation was facilitated by means of the transformation to an

ordinary differential equation by introducing suitable dimensionless parameters – a powerful method very much discussed e.g. by Bird et al. [15].

Trettin and Doshi, [16], also suggested that including the axial convection term would considerably improve the accuracy of ultrafiltration mass transfer modeling. Especially in cases where the concentration of the macrosolute at the membrane surface reaches its solubility limit, more elaborate modeling is necessary than for instance the one-dimensional film model. They described an integral solution of the diffusion equation. The integral solution was solved by approximating the solute concentration in the boundary layer near the membrane surface as a polynomial in distance measured from the membrane wall. The order of the polynomial was determined by process parameters. A similar approach was also suggested by Leung and Probstein, [17] using a quadratic polynomial for the approximation of solute concentration.

One of the main purposes for such mass transfer models as the ones described above, was to obtain suitable means for interpreting experimental ultrafiltration results with macromolecule solutions. Often the aim was to obtain permeate flux correlations for industrial ultrafiltration plant design. Traditionally, experimental ultrafiltration studies have been performed by using well-defined model compounds, such as a protein called bovine serum albumin (BSA) or dextran. The molecular weight of such macromolecules ranges from a few tens of thousands up to 100 000. It is worth mentioning that lignosulfonates in spent sulfite liquor have close to the same magnitude of molecular weight.

The gel polarization model of Michaels as described by equation (1) leads to a simple procedure for expressing experimental ultrafiltration data. If the permeate flux is plotted against the logarithm of the bulk solute concentration ( $C_b$ ), a straight line should be obtained. The slope of the line is the mass transfer coefficient ( $k_v$ ). Furthermore, the intercept of the line with the abscissa axis will give the value of the gel concentration ( $C_g$ ). This procedure has been extensively utilized in the published ultrafiltration literature up to the present time. However, as earlier mentioned, the accuracy of the method has been criticized e.g. by Shen and Probstein, [13]. Therefore, more representative correlation methods were called for. For example, Nakao and Kimura, [18], have presented a method for correlation experimental ultrafiltration results in concentration polarization determined ultrafiltration systems. Their method is based on the film model (Equation (2)).

#### Optimization of multistage ultrafiltration systems:

In the design of industrial process plants, an economical optimization of the system is often required due to the fact that there are numerous ways to perform the desired technical task. Especially when a process comprises several stages in series, it is important to perform optimization. A multistage ultrafiltration or reverse osmosis process is no exception to this principle. The process optimization of multistage membrane separation processes has not been much discussed in the literature. Baker *et al.* [19] have discussed the optimization of reverse osmosis systems. Heikonen *et al.* [20] and Kreula *et al.* [21] have studied the design of multistage ultrafiltration systems for whey and skim milk. In their analysis, the target of optimization was to minimize the total membrane area of the system where the total number of separation stages was fixed.

The optimization problems encountered in the design of multistage ultrafiltration and reverse osmosis systems are typically multivariable and non-linear. While normal methods of non-linear programming can be used to solve these problems, the associated solutions can become complicated because of the large number of parameters to be optimized and the non-linearity of the design equations. This situation is particularly characteristic of systems where the number of stages in series becomes greater than two or three.



If the structure of the process flowsheet is serial with respect to process stages and does not include branches or recycle loops, dynamic programming is a powerful technique for process optimization. Aris *et al.* [22] have applied dynamic programming for the optimization of multistage extraction systems. Aris has also used dynamic programming for optimization of chemical reactor systems, [23]. Mitten and Nemhauser [24] have discussed the usage of dynamic programming in general for the optimization of multistage processes found in the process industries. A major advantage of dynamic programming is that the task of optimization can be reduced to a number of subtasks requiring the optimization of a single task at a time. This approach can considerably shorten the total computation time necessary to find the overall optimal conditions.

Even though there are obvious benefits for using dynamic programming in the optimization of multistage membrane separation processes, no references to such applications have been found in the literature

## 1.2 DETERMINATION OF LIGNOSULFONATE REJECTION FROM TEST RESULTS IN THE ULTRAFILTRATION OF SPENT SULFITE LIQUOR, [1]

The first paper of the thesis, [1], presents a method for the correlation of experimental data from the ultrafiltration of spent sulfite liquor. The target of the study was to generate a mass transfer model to be used for the design of an industrial-scale ultrafiltration system for the manufacturing of purified lignosulfonate products.

In membrane separation technology, the quantity that expresses the separation efficiency of the membrane is called the rejection of the solute. Due to the concentration polarization as depicted earlier in Figure 1, the actual solute rejection of the membrane ( $R$ ) depends on the solute concentration at the proximity of the membrane surface according to equation (3):

$$R = 1 - \frac{C_p}{C_w} \quad (3)$$

In equation (3),  $C_p$  is the solute concentration on the permeate side and  $C_w$  is the solute concentration on the membrane surface on the concentrate side (also called wall concentration).

In case of concentration polarization, the observed membrane rejection is smaller than that of the true membrane rejection according to equation (3). The observed rejection can be defined by equation (4):

$$R_{OBS} = 1 - \frac{C_p}{C_b} \quad (4)$$

If the macromolecule solute has a wide molecular size distribution, as is the case with lignosulfonates in spent sulfite liquor (the molecular weight varies between a few thousand up to about 40000), the membrane rejection varies accordingly and the separation efficiency of the membrane cannot be described by just one figure, [1]. Since the membrane rejection behavior is needed in order to design and describe the ultrafiltration system properly, it is of great significance to have a method for measuring or estimating the solute concentration close to the membrane surface on the concentrate side, i.e.  $C_w$ . The reliable measurement of  $C_w$  is practically impossible. Therefore, it has to be calculated from the available measurements. The behavior of the wall concentration  $C_w$  during the course of the ultrafiltration process is needed especially in order to be able to utilize such mass transfer models as the film model (equation (2))

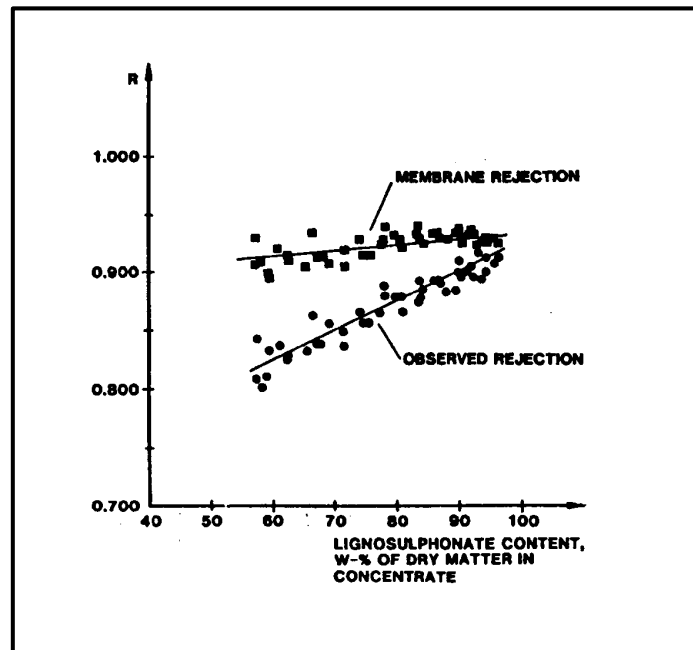
The study [1] presents a method for evaluating the wall concentration from experimental ultrafiltration results. The procedure is based on the correlation of the permeate flux obtained by the film model. Equation (2), is solved for  $C_w$  and the following equation is obtained:

$$C_w = C_p + (C_b - C_p) \exp(J_v / k_v) \quad (5)$$

In equation (5),  $C_p$ ,  $C_b$  and  $J_v$  can be obtained from test results. The mass transfer coefficient ( $k_v$ ) can be calculated with a suitable correlation, which depends on the flow regime valid for the conditions in the ultrafiltration equipment in question. In the laminar flow regime, which is the case in the applied ultrafiltration system, the appropriate and accepted correlation for the mass transfer coefficient is the Leveque correlation, suggested e.g. by [25].

In the study [1], a method is also presented for considering the increase in the average molecular weight of the solute in the course of the purification of the lignosulfonate solution during the ultrafiltration of spent sulfite liquor. This aspect has a special significance since the diffusion coefficient and consequently the mass transfer coefficient of the solute transport are strongly affected by the molecular mass of the solute.

When the wall concentration was calculated for the set of the ultrafiltration test results with spent sulfite liquor, it is possible to compare the observed and true membrane rejection values by using equations (3) and (4). Figure 2 shows the results as a function of the lignosulfonate purity.



**Figure 2:** Observed and membrane rejection of lignosulfonates during the ultrafiltration of spent sulfite liquor, [1]

The behavior of the observed and true membrane rejection can be further utilized in the mass transfer model suitable e.g. for design purposes. When applying the definitions of the observed and true rejection, i.e. equations (3) and (4), in the film model mass transfer equation for permeate flux, the following expression can be obtained for permeate flux:

$$J_v = k_v \ln \left[ \frac{R(1 - R_{OBS})}{R_{OBS}(1 - R)} \right] \quad (6)$$

The application of equation (6) is simple in the present case, since the dependency of the rejection values is linear in relation to the lignosulfonate purity in the concentrate, as depicted in Figure 2. This correlation was further utilized in the optimization study [3].

### 1.3 A METHOD FOR CORRELATING PERMEATE FLUX IN THE ULTRAFILTRATION OF MACROSOLUTES, [2]

As mentioned above, the assumption of negligible axial transport, on which the film model is based, may not be accurate enough in the ultrafiltration of such macromolecule solutions as is the case with spent sulfite liquors. The results of the previous study [1] already showed inconsistencies such as a local maximum for the solute concentration at the membrane wall in a lower concentration than the available maximum level for the bulk concentration of the solute. Based on the concentration polarization theory, the wall concentration should finally reach the gel concentration and stay there. For this reason, a more accurate model ought to be postulated for the mass transfer rate in the case of ultrafiltration in the specified conditions of the current problem.

The development of the improved mass transfer model starts by formulating the material balance (diffusion equation) and the equation of continuity describing the two dimensional laminar and steady-state flow of a Newtonian liquid. Such a flow can be assumed to take place in the parallel plate configuration of the ultrafiltration system in question. First it was shown that the analytical solution of the model reduces to the above presented film model if the axial transport is ignored. After that, a more complete solution taking axial transport into account was generated.

Typically, the solution of the diffusion equations can be facilitated by changing the partial differential equation to an ordinary differential equation by utilizing a transformation of the spatial coordinates. Such a classical approach has been presented with numerous examples e.g. by [15]. Another solution, applied for the present problem, involves assuming a polynomial form for the transverse concentration profile in the boundary layer close to the membrane wall. Upon integration of the diffusion equation by combining the assumed polynomial concentration profile, an analytical expression could be obtained for permeate flux. This solution can be called the "integral solution" due to the solution procedure.

By means of further manipulations, a useful correlation for the average permeate flux could be obtained. Furthermore, an expression for the exponent of the polynomial concentration profile could be developed. The integral solution thus generated is more general than the one presented earlier by Trettin and Doshi [16] since the incomplete rejection of the solute by the membrane has been taken into account. In this respect the resulting formulae are more applicable e.g. for the ultrafiltration of lignin-based material, since by observation in such cases there is measurable and not insignificant concentration of the macrosolute on the permeate side too.

The model was tested for the same experimental data as used in [1]. It could be shown that less internal scattering than in the case of the earlier used film model was obtained for the calculated wall concentrations. In addition to that, the more accurate model indicated a more consistent behavior for the wall concentration, i.e. a steady growth as a function of the bulk concentration until a constant level is reached, apparently corresponding to the gel concentration of the lignosulfonates. The comparison of the behavior of the wall concentration  $C_w$  calculated by means of the integral model and the film model of [1] is illustrated in Figure 3.

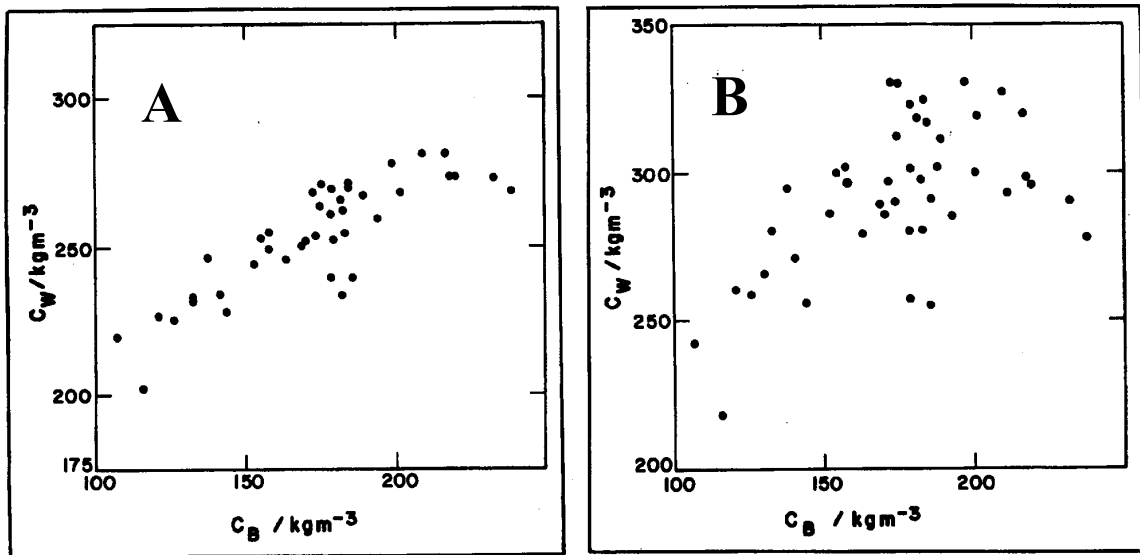


Figure 3: Lignosulfonate concentration at the membrane wall predicted by the integral method (A) and film model (B), [2]

At the end of the article [2], a semi-empirical correlation method for the evaluation of experimental ultrafiltration data was suggested. The principle was based on the apparent form of the wall concentration as well as on the correlation of the results by means of the dimensionless driving force for the permeate flux. The regression model obtained turned out to describe the test data excellently statistically speaking. Such a regression model would be more reliable for design and optimization purposes than the method based on the film model that was suggested by [1].

#### 1.4 OPTIMIZATION OF AN ULTRAFILTRATION-DIAFILTRATION PROCESS USING DYNAMIC PROGRAMMING, [3]

The third article discusses a practical design problem, i.e. the optimization of an industrial ultrafiltration system, [3]. The idea of applying dynamic programming for the optimization of multistage membrane separation processes seems to have been unstudied prior to the present work.

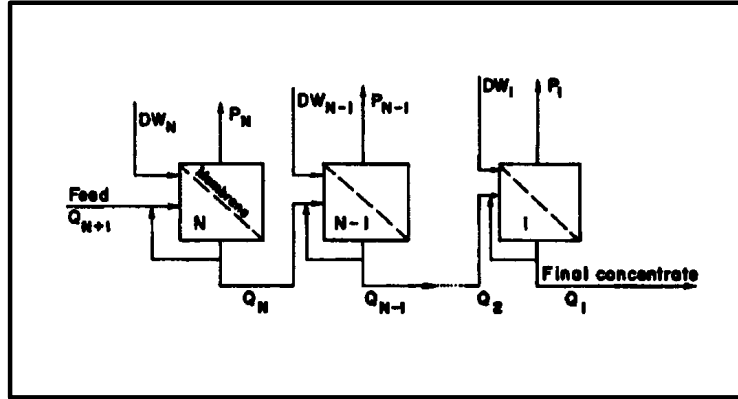


Figure 4: Structure of a multistage ultrafiltration-diafiltration system, [3]

It is evident that the structure and the process configuration of a multistage ultrafiltration system (see Figure 4) are suitable for the application of dynamic programming, since there are no branches and recycle loops between the separation stages. When dynamic programming is employed in the design of such stage-wise serial processes as ultrafiltration, sub-optimization is carried out in a sequence starting with the final stage and ending with the initial stage. Each sub-optimization comprises the maximization of an objective function with two contributions, one due to the current stage ( $g_i^*$ ) and the other the maximum of the cumulative objective function obtained over the preceding optimization steps 1 to  $i-1$ .

The objective function of the net value of the investment can be expressed by means of equation (7):

$$U_i = g_i^*(d_i, s_{i+1}) + \max[U_{i-1}(s_i)] \quad (7)$$

where  $d_i$  is the decision variable to be sub-optimized for the stage in question and  $s_{i+1}$  is the information vector.

The information vector contains the data necessary to characterize the concentrate stream leaving the preceding upstream stage. In the present case there are three components in the vector: the volumetric flow rate ( $Q_{i+1}$ ), the concentration of dissolved solids ( $w_{i+1}$ ) and the weight fraction of lignosulfonates in the dissolved solids ( $X_{i+1}$ ).

The cumulative objective function was approximated by means of a quadratic response surface. This is useful in saving the computation time and especially in the final reconstruction of the optimal policy. However, by introducing such an approximation there is a risk of causing a certain inaccuracy in the optimization. This matter was addressed when interpreting the results.

The design equations were based on the overall material balance and the macrosolute (lignosulfonate) and microsolute (non-lignosulfonates) balances over an ultrafiltration stage. One stage was defined to represent a set of connected ultrafiltration modules in parallel where there is an abundant recirculation of the concentrate in order to allow constant conditions for the solute concentration and flow rate inside the stage. This is one of the preconditions for the utilization of the film model. The film model correlations for the observed and membrane rejections based on the results of [1] were applied for the calculation of permeate flux. The remainder of the theoretical approach for describing the mass transfer phenomena was also based on [1].

The optimization of an ultrafiltration plant was performed by means of dynamic programming and for comparison, with a non-linear direct search algorithm (Complex

Search). The article [3] shows several graphical diagrams with which the behavior of the optimized parameters can be illustrated. It could be shown that both dynamic programming and the direct search algorithm could suggest the same value for the optimal number of stages in series. However, the dynamic programming could predict a "better" optimum, i.e. a bigger value for the total objective function. This is shown in Figure 5.

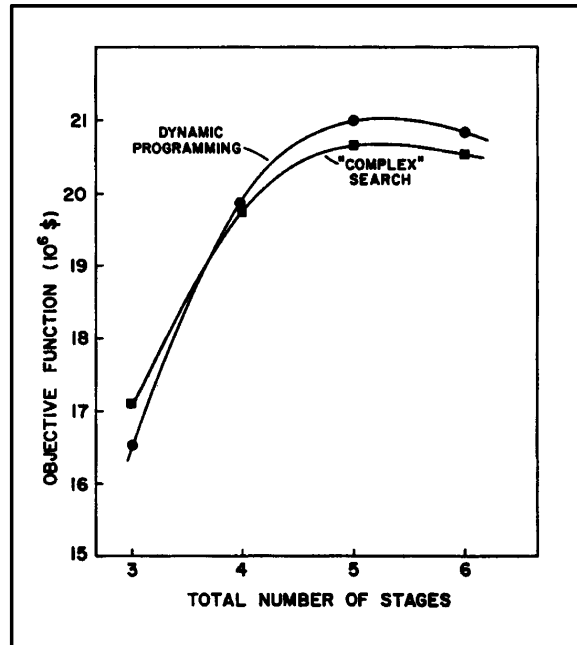


Figure 5: Overall objective function versus the total number of separation stages in series, [3]

According to the optimization results, the factors that had the greatest influence on the objective function were the total membrane area and the rate at which the lignosulfonates are produced. As an example, Figure 6 shows how the total membrane area is reduced when the number of separation stages in series is increased.

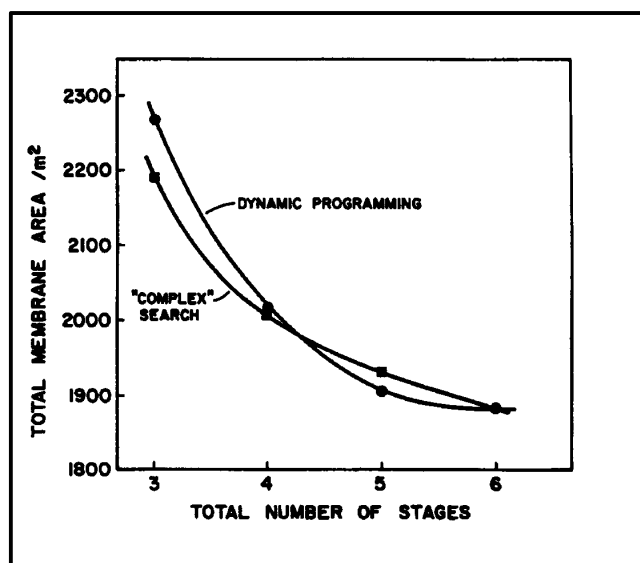


Figure 6: Total membrane area versus total number of stages, [3]

The effect of the approximation due to the quadratic response surfaces was a minor one especially in the cases where the total number of stages increased. This was beneficial since the optimum was set to a relatively large number of stages (five). The total computation time was only one third compared to that of the Complex Search alternative. This is due to the characteristics of dynamic programming. In fact, the reduction in computation time could be even greater in the case of dynamic programming since the Complex Search calculations were carried out after the results of the dynamic programming calculations were already available to be utilized for estimating the region where the optimum ought to be.

## 1.5 DISCUSSION OF THE MODELING OF ULTRAFILTRATION

### Modeling of mass transfer in the ultrafiltration of spent sulfite liquor:

The modeling of the mass transfer phenomena in the ultrafiltration of pulping liquors is more challenging than many other solute systems. The reason is the fact that the lignin-based macrosolute has a wide molecular weight distribution. In addition to that, the distribution changes during the enrichment of the solute in the course of the ultrafiltration process as described by [1]. Most of the reported mass transfer studies dealing with the ultrafiltration of macromolecular solutions have been performed in systems where the solute has basically only one size of macromolecule.

Since the molecular weight of the solute has a wide distribution, it is important to take this matter into account when evaluating the most important transport parameter, i.e. the diffusivity of the lignosulfonate macromolecules in the boundary layer close to the membrane wall. A method has been developed in [1] to take the varying distribution of the molecular weight into consideration based on experimental data. The obtained behavior of the average molecular mass was then utilized in the evaluation of the diffusivity of lignosulfonates and mass transfer coefficient in the analysis of mass transfer in the studies [1] and [2].

Regarding the mass transfer characteristics, a simpler approach (film model) was first attempted in order to evaluate the macrosolute concentration at the membrane wall for a set of experimental conditions in a pilot plant ultrafiltration system. This method turned out to produce such correlations for the membrane rejection that could be utilized for plant design purposes. However, from a more fundamental point of view, the assumption that axial solute transport can be ignored, which is the basis for the film model, brings about a rather large internal scattering of the calculated values of the solute wall concentration,  $C_w$ . In addition to that, the behavior of the solute wall concentration as a function of the solute concentration in the bulk liquid phase appears to behave in a non-realistic way in the highest solute concentrations (a local maximum is predicted).

When axial transport is taken into account, the modeling of the mass transfer phenomena becomes more complicated. However, it is possible to formulate an analytical solution for the permeate flux by means of the assumption of a polynomial equation for the concentration profile in the boundary layer at the membrane wall. When this mass transfer model was applied to the same experiments as earlier with the film model, significantly less internal scattering of the calculated wall concentrations and a physically more consistent behavior was obtained for the calculated wall concentration. The latter means a gradually increasing wall concentration, which approaches a constant maximum value apparently representing the gel concentration of lignosulfonates in the conditions of the experimental setup. Since wall concentration is difficult to measure, one has to rely on such an indirect calculation method in order to be able to express e.g. the true membrane rejection.

It was also possible to suggest a new method to correlate experimental permeate flux. This model is suitable, for example, for plant design purposes. The method implies the utilization of the observed mathematical form for the behavior of the wall concentration and then modifying the dimensionless driving force term accordingly. The parameters of the semi-empirical model can then be sought by means of regression analysis.

When comparing the results of the modeling studies with other published research, it should be noticed that the ultrafiltration of lignin-based solutions (kraft black liquor or spent sulfite liquor) has not been studied extensively from the mass transfer point of view. For example, Woerner and McCarthy [26] correlated experimental ultrafiltration data of kraft black liquor and spent sulfite liquor by means of the gel polarization model of Michaels (equation (1)). It is interesting to observe that the gel concentration of lignosulfonates obtained (265 g/l) was almost the same as that predicted by the present study (270 –280 g/l, see Figure 3). Pan *et al.* [27] have also used the gel polarization model in their ultrafiltration studies with non-wood pulping liquors. Bhattacharjee and Bhattacharya [28] correlated their ultrafiltration data obtained with kraft black liquor by means of the film model approach (the velocity variation method of Nakao and Kimura, [18]). Ramamurthy *et al.* [29] have used the same approach as in the present study, [1], for calculating the wall concentration by rearranging the film model permeate flux correlation.

The gel polarization model traditionally used by many researchers has been criticized because of the fact that the actual gelation rarely occurs in ultrafiltration. In addition, there have been large variations in the gel concentration in seemingly identical solutions, e.g. [30], [31] and [32]. For this reason, more accurate mass transfer models are needed - also for the analysis of the ultrafiltration data of pulping liquors. In fact, the integral model of [2] seems to be the most fundamental one so far applied in the correlation of the experimental ultrafiltration results of pulping liquors.

Regarding the recent progress in the analysis of mass transfer in macromolecular ultrafiltration systems, there has been a trend towards improving the accuracy of such conventional modeling as described above. Since the gel polarization model and also the film model have in many occasions turned out to give inaccurate predictions of experimental data, the accuracy has been significantly improved by expressing the mass transfer coefficient more precisely, e.g. [30] and [31]. One of the factors that has been improved, has been the correction of the mass transfer coefficient by means of accounting for the viscosity variation as a function of the solute concentration in the boundary layer, [31]. It has also been suggested that the variation of solute diffusivity as a function of solute concentration should be implicitly incorporated into the governing mass transfer equations, [33].

The incorporation of the axial convection term into the diffusion equation, as in the present study [2], has been the basis for a number of studies on mass transfer in macromolecular ultrafiltration. For example, Davis and Sherwood [34] used this approach and also took the effect of concentration-dependent viscosity into account. Perkins *et al.* [35] added the momentum equation into the model and solved a set of equations by using the approximative solution developed by Pohlhausen [15] to express the permeate flux as a function of the mass transfer coefficient and the concentration driving force.

Some of the most recent mass transfer studies have been based on CFD (Computer Fluid Dynamics), which allows the proposal of a complicated set of governing equations, e.g. Magueijo *et al.* [36].

Using more complicated mass transfer modeling than in the present study calls for carefully controlled experimental procedures in the laboratory in order to verify any improved accuracy for the predictions. In addition, the solute system needs to be well defined. Typically this means certain standard macromolecular solutions of the solute of



precisely known molecule weight. Against this background it seems correct to claim that the modeling principles used in the present study are relevant, considering the applied semi-industrial scale experimental arrangement and the solute system based on an industrial pulping liquor.

#### Optimization of a multistage ultrafiltration system:

It was shown that the complicated optimization task for a multistage industrial ultrafiltration-diafiltration system could be performed effectively by formulating the problem by means of dynamic programming. Dynamic programming seems to be able to produce even better optimal conditions in this specific case than the direct search of the optimal set of decision variables. The optimization example also featured the utilization of the earlier-developed mass transfer correlations (based on the film model).

The recently published literature covers very few examples of the optimization of membrane separation systems. This has been pointed out by Cross [37]. He states that process and system design is almost always carried out by specialized engineering or equipment suppliers and publications of such information are not available.

Shalan et al. [38] performed the process optimization for a combined one-stage ultrafiltration and nanofiltration system. For such a simple configuration the optimum conditions were sought by means of maximizing the annual profits using a Complex Search routine similar to that used for the comparisons of the dynamic programming solution in the present study [3].

Niemi [39] gives a comprehensive discussion of the modeling of membrane processes. He also presents a number of calculation examples for the optimization of various multistage ultrafiltration and reverse osmosis cases. Niemi found that the optimum number of separation stages in series was in most cases 2 or 3. The example processes were, however, different from the one used in this study. It is interesting to notice that some of the optimization strategies employed by Niemi were very slow to converge. Dynamic programming was not compared in Niemi's work.

Some of the examples given by Niemi [39] suggest that dynamic programming could be a viable optimization tool. The calculations of [3] were performed about 20 years ago when computer technology was far behind the level of today. Even then it could be shown that the computation time was significantly reduced if dynamic programming was chosen for the strategy of optimization compared to the standard search method. According to Niemi, [39], in some cases the convergence towards the optimal conditions could not be finished until a few hours of CPU time had been used. Dynamic programming may be a method for accelerating the finding of the optimum conditions in such slowly converging situations.

#### General comments on the application of ultrafiltration in pulp and paper mills:

As mentioned in the introduction to this work, sulfite pulping processes are gradually disappearing as a method for producing chemical pulp. Therefore, the possibilities for the separation of lignosulfonates by means of ultrafiltration, as described in this study, are practically non-existent today.

Nevertheless, the prospects for applying ultrafiltration in pulp and paper mills have become quite promising in the last few years. The applications are not in the manufacturing of purified lignin-based products, but in the treatment of wastewater effluents discharged from the process. Even back in the 1980's there was a lot of interest in reducing the environmental impact of pulp bleaching plants by means of ultrafiltration of the filtrates from the chlorination and alkali extraction stages. A few pilot- and industrial-scale ultrafiltration systems were built in Sweden and Japan, e.g. [40] and [41]. The

separation characteristics of the ultrafiltration of the effluents were extensively tested on a laboratory scale by Jönsson and her co-workers, [40], [42] and [43].

Ultrafiltration did not, however, have much success in spite of promising results and technically viable separation technology. The same environmental effects could be obtained by means of modifications inside the pulping and bleaching processes. In addition, the membranes suffered from fouling and their lifetime was not sufficient for continuous industrial work.

The development of membranes towards better resistance against fouling as well as the need for reducing water consumption in the pulp and paper mills have catalyzed new and more successful efforts for applying ultrafiltration for the treatment of effluents. Research on the ultrafiltration of bleaching effluents has been revived, e.g. [44] and [45]. A new industrial-scale ultrafiltration plant for treating bleaching effluents has been in use for several years at StoraEnso's Nymölla pulp mill in Sweden with promising results regarding a final breakthrough and acceptance within the pulp industry, [46].

Similar development has taken place in the application of ultrafiltration for reduction in the fresh water consumption of paper mills, e.g. [47] and [48]. Successful mill-scale applications have been introduced and ultrafiltration appears to have become recognized technology for closing water circulation in paper mills, too[49].

Even though ultrafiltration has been widely tested on laboratory and industrial scale using the above-described effluent systems of pulp and paper mills, the mass transfer characteristics have not been considered. The modeling principles of the present study should be directly applicable in these systems, provided that the macromolecule systems are properly characterized and the critical properties (e.g. diffusivities of the solute constituents) have been determined. The plant design and optimization of the effluent treatment ultrafiltration systems can be facilitated by means of such mass transfer correlations as those reported above.

## 2 FORMATION OF PULP MAT AND DETERMINING THE CAPACITY OF PULP WASHERS

### 2.1 BACKGROUND

The formation of a pulp mat on a moving filter medium (traditionally, a wire cloth but in modern pulp washing technology also a perforated plate) is the first phase in a series of operations taking place in a pulp washing or thickening apparatus. The mat formation stage is followed by the actual washing or displacement stages, the number of which can be one or more, depending on the type of washing apparatus. There is a final thickening phase before the washed pulp leaves the washing apparatus. In some applications this final thickening may be performed to a high consistency by means of pressing.

The role of mat formation is of primary importance with regard to the performance of the washing device. In such types of pulp washers, where the inlet pulp suspension is introduced at a rather low consistency (typically 1 – 3 %), the pulp production capacity of the device is determined at the mat formation stage. Typical washer applications of this kind are conventional vacuum and pressure drum washers and belt washers. Even in such modern pulp washers as drum displacers or displacement presses, the mat formation stage may become the limiting factor regarding the production capacity, if these machines are operating at low inlet consistency. However, increasing the inlet consistency to a level typical of diffuser washers and medium consistency drum displacers and presses (8 – 10 %), moves the capacity limitation to the wash liquid displacement stage or stages.

Furthermore, the uniformity of the fiber bed is created at the mat formation stage. In this respect, the conditions for the subsequent washing (displacement) stages are greatly influenced by mat formation. A pre-condition for high displacement efficiency is the fact that the pulp mat is as uniform as possible in terms of created fiber and void distributions.

The traditional approach for designing such rotary filters as conventional filter washers has been based on leaf tests, where the filtration characteristics have been determined by means of rather simple laboratory procedures, e.g. [50]. The interpretation of leaf test data is most often based on the assumption of incompressible cake filtration. In general, the assumption of incompressible pulp beds has been made quite often in the literature dealing with the dimensioning of pulp washers and thickeners, e.g. [51], [52] and [53].

Suppliers of pulping equipment have typically determined the effect of the critical variables on the washer capacity and thus obtained design data for the dimensioning of the washing equipment in varying conditions and situations. Even though there is only limited published information available, this type of design data has been based on pulp drainage testing with pulp testers capable of simulating industrial washing equipment, [53], [54], [55]. Drainage testing of mechanical pulps has also been conducted in conditions that aim at simulating industrial systems, e.g. [56].

The ways to interpret pulp drainage results from such test equipment may have relied on a purely empirical approach, e.g. [55]. More advanced semi-empirical methods utilize Darcy's law as well as the assumptions of constant pressure filtration and incompressible fiber beds. The filtration data from a test device has been fitted into obtained filtration models, e.g. [53] and [54].

Even though this latter kind of approach is practical and brings a quite acceptable theoretical basis to the interpretation of the results, there are still some drawbacks. One is the plain fact that the fiber mats are compressible. However, if the applied pressure difference is small (e.g. of the order of 0 – 20 kPa), the assumption leads to fairly minor

errors in consideration of the purpose of the testing. Another drawback is the fact that extensive experimental work may be needed in order to generate the design data.

Pulp drainage testing can also be performed in order to determine the fiber-specific property data (specific surface and volume of the fibers as well as the compressibility parameters) needed in filtration calculations by means of the Kozeny-Carman equation. The pioneering work of Robertson and Mason, [57] as well as Ingmanson and his coworkers, [58, 59, 60, 61], paved the way for the development of pulp drainage testing of basic filtration properties. The developed compressibility-permeability cell and the accompanying testing methods have become a standard procedure in determining filtration parameters for different paper-making fibers, e.g. [62], [63] and [64].

The problem of mat formation in pulp washers can be modeled based on the fundamental filtration theory of Kozeny-Carman by utilizing fiber-specific property data. Such an approach also takes into account the compressibility of the fiber mat and correctly addresses the dynamic behavior of the mat formation stage. The pioneering work of Ingmanson *et al.* in the 1950's [58, 59, 60, 61] greatly contributed to the understanding of filtration phenomena. Even though the emphasis of these studies was mainly on thinner fiber mats, as is the case with paper machines, the theory can be extended to the formation of the thicker pulp mats of pulp washing equipment, [65]. The general filtration model presented by Meyer, [66], is very useful from the technical point of view since the transient period of the mat formation stage has been expressed in such a way that can be applied for paper machines, pulp drying machines and also for pulp washers.

A few methods have been presented for modeling the filtration phenomena in pulp washers using the Kozeny-Carman equation and incorporating it with Darcy's law. Nordén *et al.*, [67] presented a graphical method for calculating the filtration phenomena of pulp mats. The method does not directly address the mat formation stage but presents integrated correlations useful in evaluating the pressure loss, liquid flow and average consistency in already formed fiber beds, e.g. during the wash liquid displacement in a pulp washer.

There are also a few recent studies concerning the mathematical modeling of compressible cake filtration, i.e. applicable for such systems where pulp fibers are treated. For example, the work by Stamatakis and Chi Chien [68] as well as Nordén and Kauppinen [69] should be mentioned. In the latter, the filtration equations have been formulated as partial differential equations in the same form as in diffusion. The model was further applied to the dewatering of a pulp suspension on a pulp washer. Jönsson and Jönsson have discussed both steady-state and dynamic phenomena in the fluid flow of compressible and porous media, [70], [71]. Niemenmaa [72] has also discussed the modeling of filtration phenomena taking place in a specific type of pulp washing apparatus.

In these studies, the modeling of time-dependent filtration phenomena is performed by formulating the continuity equations for the filtrate and solids flow, expressing the effect of the pressure gradient by means of Darcy's equation and then formulating the compressibility effects by means of a power function of the type first presented by Qviller [73]. Solving the model equations involves using rather complicated numerical methods.

In spite of the fact that the constitutive equation of the compressibility of a fiber bed is practically always expressed by means of the power function formula of Qviller, the compressibility behavior can also be put forward using an analogy to a system of loaded springs, as described by the Terzaghi-Voight model, [74], for example. This approach has been successfully applied at least for such compressible systems as kaolin and other clay suspensions and should be possible for fiber suspensions as well.

## 2.2 THE EFFECT OF SOME PARAMETERS ON BROWNSTOCK WASHING, [4]

The first study on pulp washer dimensioning deals with experimental work using a pulp tester. The tester in question was developed at the beginning of the 1980's by Pertti Haapamäki, [75]. Prior to [4] there had been only one published study using this tester, [76], discussing the effect of air on bleach plant washer capacity and washing efficiency.

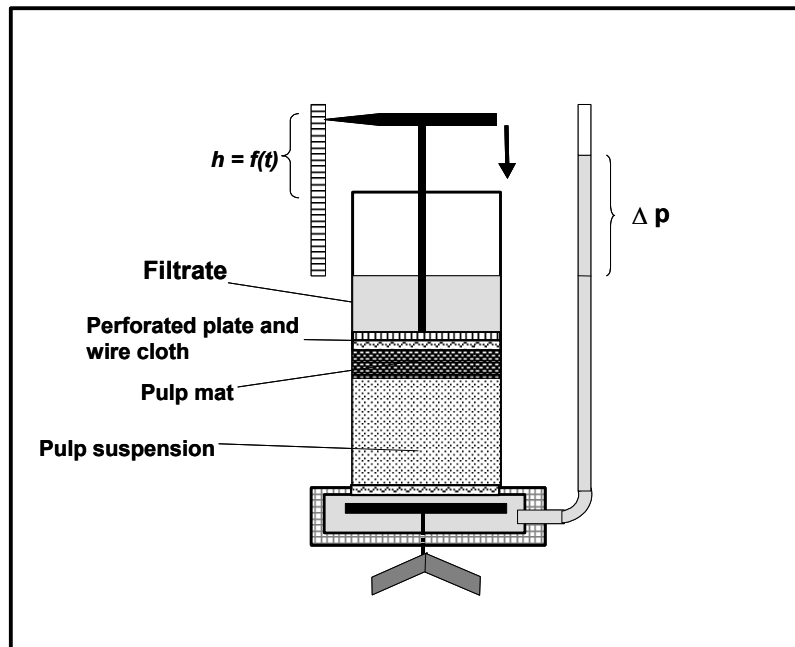


Figure 7: A schematic picture of the pulp-washing tester

The tester used in the studies of [4] is illustrated in Figure 7. When a mat formation test is conducted, the pulp suspension to be studied is first poured into the cylinder of the tester. After that, a piston is placed on the surface level of the suspension. The piston is equipped with a perforated plate with a wire cloth. The piston is then pushed down with a force that generates the desired constant filtration pressure. The pressure is observed by means of a manometer and the pushing force is controlled accordingly. During the piston movement (downwards into the slurry), a pulp mat is formed on the wire, while the filtrate goes through the mat and the wire.

The motion of the piston is observed (distance vs. time). The movement of the piston depends on the filtration resistance of the pulp suspension being studied. At each test point, the temperature and consistency of the slurry are also observed.

In the study of [4], a simplified mathematical approach was given for the mat formation stage. Basically this is the overall fiber balance during the mat formation stage. The average pulp load ( $\text{kg fiber}/(\text{m}^2\text{s})$ ) was expressed using the pulp tester parameters. Correspondingly, the average pulp load was formulated for a drum washer. By doing this, the available pulp load of a drum wash filter could then be calculated from the tester measurements.

The study also contains pulp-washing data. After each mat formation test, the trial was continued with a washing test. The Nordén E-number was calculated from the tester results using a modified E-number correlation, which is equivalent to the original definition of the E-number given for instance in [77].

An extensive test series was performed with the tester at a Finnish pulp mill. The studied pulp suspension was taken from the brownstock washing line of the mill. All the tests were performed over a period of a few days when the pulping line was producing softwood pulp (pine) of about 30 kappa number. Typical mat formation conditions and parameters were chosen to match the situation in brownstock washing with pressure washers. It is worth mentioning that the air content of the pulp suspensions was also chosen as a parameter.

Since the theory used in the interpretation of the tester results was formulated in a rather simple way, the number of studied test points was quite large: the entire test material comprised over 1100 test points. No fundamental relationships between the parameters were incorporated and the approach and treatment of the results was purely empirical. The test results were run through regression analysis and a power model was fitted into the results. The  $R^2$  coefficient of the model was 0.948, which was considered acceptable.

The regression model obtained for the washer square meter load is given in equation (7). The equation will be used for comparison to check the validity of the theoretical models to be presented later on.

$$\frac{G}{BDT/(m^2d)} = 0,675 \left( \frac{n}{1/\text{min}} \right)^{0,62} \left( \frac{c_s}{\%} \right)^{0,65} \left( \frac{T}{^\circ\text{C}} \right)^{0,26} \left( 1 + \frac{y_{air}}{\text{vol} - \%} \right)^{-0,24} \left( \frac{\Delta p}{\text{kPa}} \right)^{0,30} \quad (7)$$

In the study [4] a few graphical presentations are given in order to illustrate the relative effects of the operational parameters on the washer load. As an example, Figure 8 presents the effect of the inlet consistency and washer drum speed on the washer capacity.

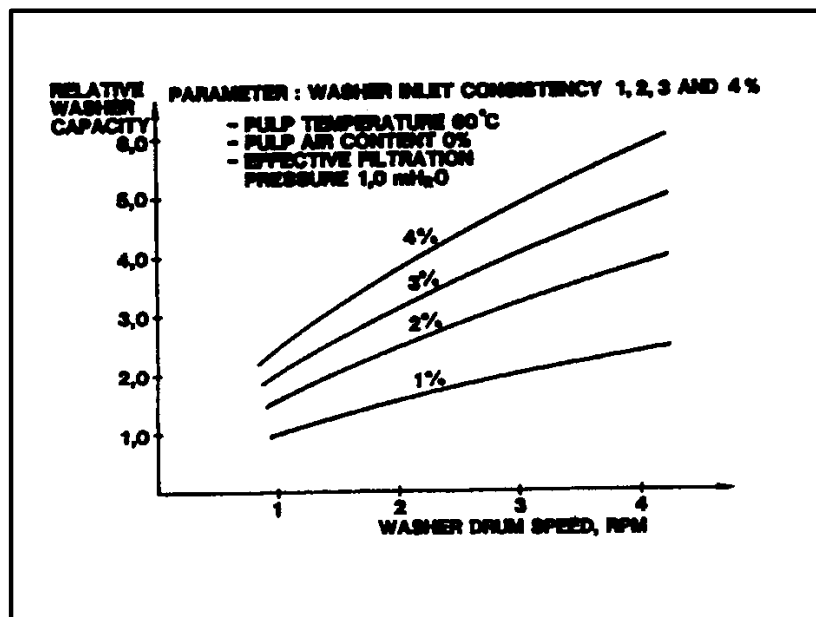


Figure 8: Effect of drum speed and inlet consistency on the washer capacity, [4].

It is interesting to notice the effect of the air content on the washer capacity. The qualitative knowledge about the harmful effect of the air present in the pulp suspension is familiar to anybody that has worked with brownstock washing in practice. The measurements performed give rise to a quantitative expression of the air effect: even a

small volume percentage of suspended air in the inlet pulp suspension to a washer can drastically reduce the available production capacity at the washer, as shown by Figure 9.

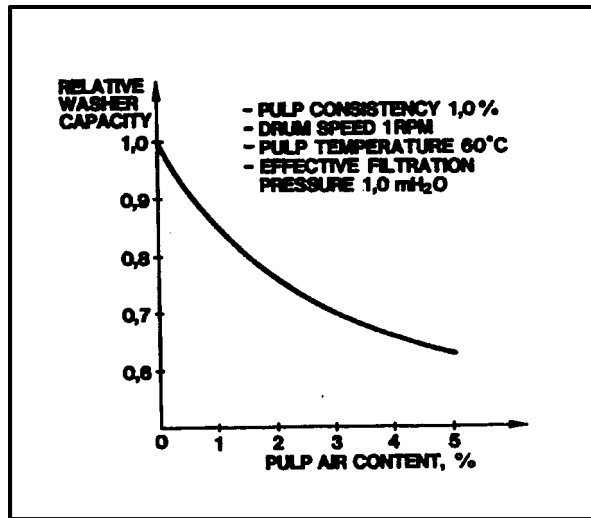


Figure 9: Effect of the air content in the pulp suspension on the washer capacity, [4].

An important aspect is the correlation of the tester results with the production rate of industrial washers. Such a comparison was made and the result is shown in Figure 10. The correlation between the tester results and corresponding actual washer production rate is good, which gives more assurance for applying equation (7) as a reference of the actual drum washer capacity.

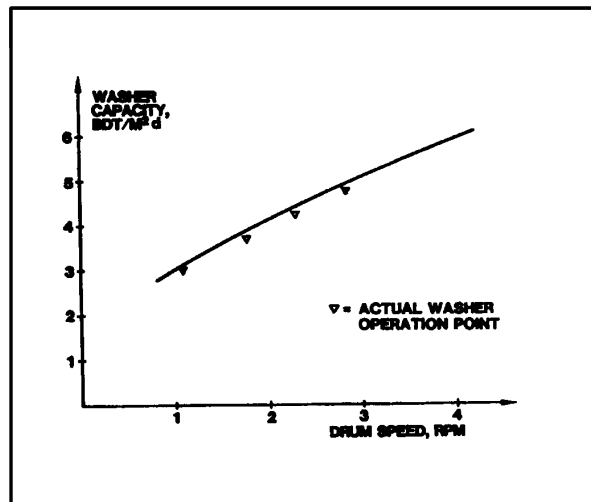


Figure 10: Comparison of the capacity curve of the tester and actual washer operation points, [4].

The second part of the study deals with measurements of washing efficiency. A few interesting observations were made. It was observed that the E-number descended quite linearly with an increased wash liquid loading as expressed by the washer dilution factor (see Figure 11). This observation is also familiar from industrial practice. When the E-factor was further plotted against the superficial liquid velocity through the pulp mat, a linear reducing tendency was obtained regarding the effect on the E-number (Figure 12).

It was pointed out that the liquid superficial velocity was one of the most important parameters in washing efficiency. This parameter is very much related to the conditions during the mat formation stage. The higher the washer-specific capacity is aimed to be, the thicker the mat needs to be formed with also a higher average consistency. Thus, with a given time available for wash liquid displacement, a higher superficial velocity needs to be arranged, thus affecting the available E-number negatively.

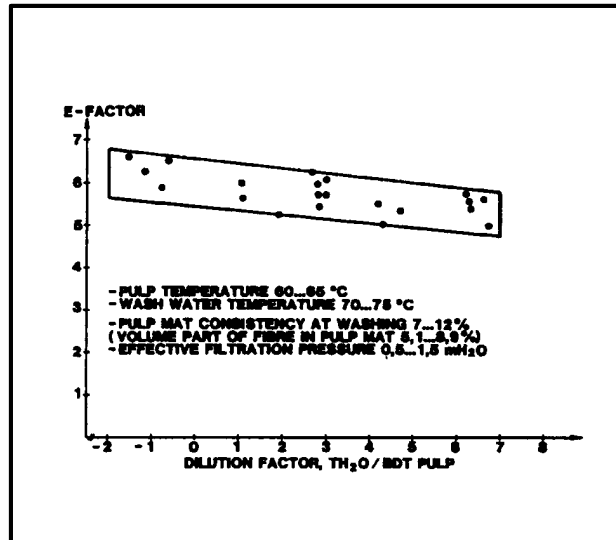


Figure 11: Effect of dilution factor on the washing efficiency, [4]

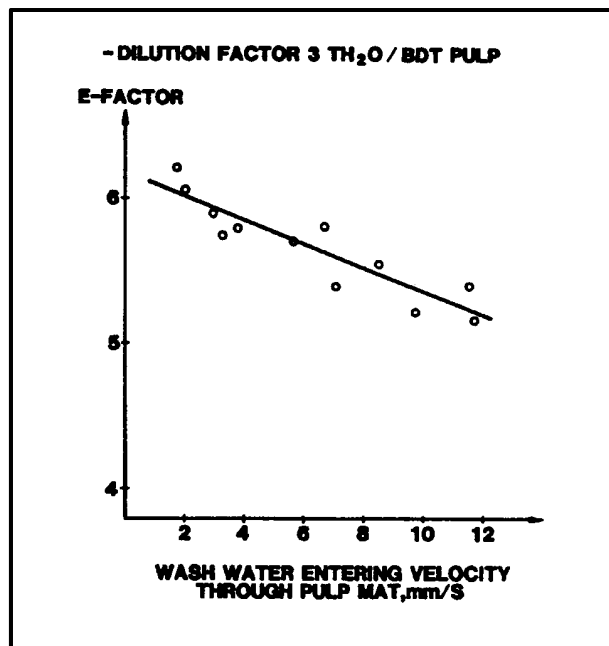


Figure 12: Effect of wash liquid superficial velocity through the pulp mat on the E-factor, [4].

The study of [4] is probably one of the most extensive experimental works regarding the effect of the parameters on mat formation in pulp washers. It showed the value of the pulp tester and also gave numerical data that can be used for checking a more theoretical analysis of mat formation. At the time of the study, the regression model obtained could also be utilized for washer dimensioning and related purposes.



### 2.3 DIMENSIONING OF THICKENERS, [5]

In the next article, [5], the dimensioning principles of drum thickeners and the application of the pulp tester mentioned above were discussed. Even though the emphasis was on a pressure thickener (type PWT), the approach is completely identical to that of drum-type pulp washers.

The simple dimensioning principle that was presented in [4] was considerably improved in this study. The derivation of the dimensioning equations was not presented in the article but it is worth showing it here considering the scope of the dissertation.

If a dynamic overall fiber balance is created for the mat formation stage and it is solved for the fiber mass per unit area of the filter medium, the following expression can be obtained:

$$g = \frac{c_s \bar{c}_{mat}}{\bar{c}_{mat} - c_s} \int_0^{t_m} u dt \quad (8)$$

The variables are defined as follows:

$g$	= fiber mass per unit area (basis weight), kg/m <sup>2</sup>
$c_s$	= fiber consistency in the feed suspension, kg/m <sup>3</sup>
$\bar{c}_{mat}$	= average fiber consistency in the formed mat, kg/m <sup>3</sup>
$t_m$	= time of mat formation, s
$u$	= superficial filtrate velocity through the mat, m/s

Equation (8) is the same as equation (1) in study [5].

Darcy's law states that the flow rate of the filtrate is directly proportional to the applied pressure gradient and inversely proportional to the viscosity of the filtrate. The proportionality constant  $K$  is called permeability or Darcy's constant. Darcy's law is presented by the following equation (9):

$$Q = -\frac{K}{\mu} \frac{A \Delta p}{L} \quad (9)$$

Stating that the total volume of the filtrate is an integral of the flow rate, i.e.  $Q = \frac{dV}{dt}$  and with the assumption that the fiber mat is incompressible and further assuming a negligible filtration resistance in the wire cloth, it is possible to modify equation (9) to the following form, [78]:

$$\frac{dV}{dt} = \frac{A^2 \Delta p}{\alpha \mu c_s V} \quad (10)$$

With the pulp tester, the cumulative filter volume can be expressed by means of the piston position:

$$V = Ah \quad (11)$$

Inserting equation (11) into equation (10), rearranging and integrating with time, an expression of the piston movement can be obtained:

$$h = K_f t^{0.5} \quad (12)$$

where  $K_f$  is a "filtration coefficient" defined as follows:

$$K_f = \left( \frac{2\Delta p}{\alpha\mu c_s} \right)^{0,5} \quad (13)$$

If equation (12) is differentiated with time, an equation for the superficial filtrate velocity can be obtained:

$$u = \frac{dh}{dt} = \frac{K_f}{2} t^{-0,5} \quad (14)$$

Equation (14) is inserted into equation (8) and the integration can thus be readily performed in order to obtain the expression for the basis weight of the pulp mat ( $g$ ). If the basis weight is multiplied by the drum speed, the expression for the pulp load of the washer drum can be obtained:

$$G = gn \quad (15)$$

After changing the units and also expressing the mat formation time in terms of the drum speed, the dimensioning equation (4) in the study [5] can be obtained. The equation (12) is significant from the tester point of view, since it provides a tool for interpreting pulp tester results in an improved way. The plots of the piston position vs. the square root of time will produce straight lines with slopes equal to the filtration coefficient  $K_f$ . The number of test points with the pulp tester can thus be reduced significantly compared with study [4], since the variables of the applied pressure difference and temperature can be taken into account with the definition of  $K_f$ , i.e. equation (13).

Figure 13 illustrates an example of the results obtained with the testing of pressure groundwood pulp of varying freeness number. The well-known effect of the reduced freeness number on the capacity of thickeners can thus be quantified.

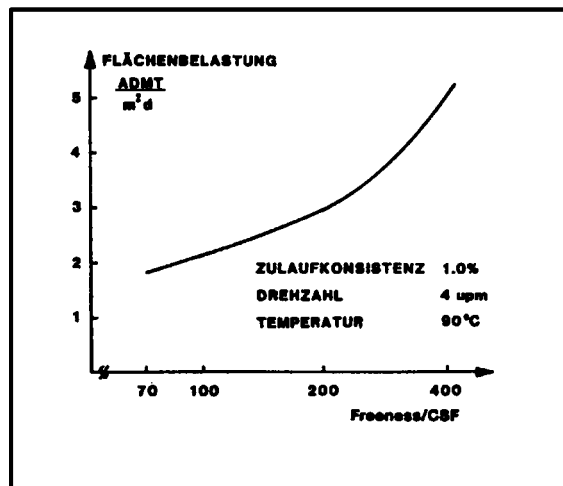


Figure 12: Effect of the freeness number of PGW pulp on the thickener capacity, [5].

The theoretical considerations presented by study [5] have been used to date for interpreting the results obtained with pulp testers of the type described by [4] and [5]. From the technical point of view, the accuracy of the simulations has been acceptable and the assumptions presented above (e.g. an incompressible pulp mat) may be justified since

most of the experimental work has been performed utilizing a fairly low pressure difference (max. about 20 kPa).

## 2.4 MATHEMATICAL MODELING OF FILTRATION PHENOMENA IN PULP WASHERS, [6]

Concerning theoretical filtration models, everything starts from the classic Darcy's law. One form of Darcy's law was shown earlier with equation (9). As regards modeling purposes, it is more appropriate to put Darcy's law in a slightly different way:

$$u = -\frac{K \Delta p}{\mu \Delta L} \quad (16)$$

Incorporating the Kozeny-Carman equation into Darcy's law further sets the basis for expressing the pressure drop gradient in a porous bed of particles. The Kozeny-Carman equation is formulated as follows:

$$K = \frac{1}{k S_o^2} \frac{\varepsilon^3}{(1 - \varepsilon)^2} \quad (17)$$

In equation (17),  $S_o$  is the specific surface of the particles and  $\varepsilon$  is the porosity of the particle bed.  $k$  is known as the Kozeny factor. For fibrous materials, it is appropriate to express the Kozeny factor according to the Davies formula, [79]. Ingmanson *et al.* [58] have determined the constants for the Davies formula by using careful water permeability experiments with glass and nylon fibers and thus the following form of the Davies equation was obtained:

$$k = 3,5 \frac{\varepsilon^3}{(1 - \varepsilon)^{0,5}} [1 + 57(1 - \varepsilon)^3] \quad (18)$$

Even though equation (18) has been the most common way to express the Kozeny factor in fiber systems, there is an alternative expression, which covers the whole porosity range slightly better, [80]:

$$k = 5,0 + \exp[14(\varepsilon - 0,8)] \quad (19)$$

The porosity of the fiber bed can be expressed by means of fiber consistency ( $c$ ) and the specific volume of the fibers ( $v$ ):

$$\varepsilon = 1 - vc \quad (20)$$

The last important basic equation, which is needed for defining the fiber bed system to be permeated, describes the compressibility characteristics of the fiber bed by relating mat consistency with the compacting pressure. As earlier mentioned, the commonly used compressibility correlation is the power function expression by Qviller [73]. The power function has been used basically in two forms:

$$c = Mp^N \quad (21)$$

$$c - c_o = Mp^N \quad (22)$$

In equation (22),  $c_o$  is the sedimentation consistency or basically the consistency of the suspension from where the fiber bed is formed.

The above-described equations comprise the fundamental formulation of the filtration phenomena in pulp fiber beds – during the formation of the pulp mat as well as the washing (displacement) stages.

One of the ways to utilize the filtration equations is to integrate equation (16) over the fiber bed depth and thus obtain a useful expression to describe the filtration characteristics of fiber beds, as presented by Ingmanson, [58]. Nordén et al [67] developed this idea further by introducing dimensionless forms of the integrated Ingmanson-type equations where the compressibility behavior of the fiber mat was also incorporated. Two equations were thus formulated, one for the dimensionless fiber mat thickness and one for the dimensionless surface loading of the fiber mat. In order to facilitate the usage of these integral equations, which are complicated enough to obtain any analytical solution, a set of double diagrams was given by Nordén et al. [67]. The graphical method obtained was then applied to a few example cases for calculating the various parameters of pulp mats.

The above method by Nordén was applied for modeling mat formation and compression as well as wash liquid displacement stages in a special type of drum pressure washer (Pro-Feed washer) in study [6]. A simplified dynamic mass balance was combined into the dimensionless equation of surface loading. Upon integration, an expression for the development of the mat surface loading as a function of the mat formation time was obtained. The modeling was then continued for the compression of the formed mat (the Pro-Feed washer is equipped with a compression plate). Finally, the theory was applied for the wash liquid displacement stage by formulating a set of partial differential equations for the mat consistency during the displacement stage.

In the Pro-Feed washer, the inlet consistency is typically 3 – 4 % and the pulp mat formation is enhanced by means of a compression plate. For this reason, the actual free mat formation due to the applied pressure gradient prevailing in the “headbox” is short and does not limit the washer capacity. It is the subsequent wash liquid displacement stage which sets the capacity limit. In such a situation, it is important to study the development of the consistency of the pulp mat during mat formation and especially at the end of it. The reason is the fact that when the mat arrives at the wash liquid displacement stage, the consistency of the mat defines the available velocity of the displacing liquor. The displacement velocity is the basis for the capacity of the washer. In addition, the washing efficiency obtained is related to the displacement velocity, as earlier shown in [4].

Figure 13 illustrates the development of the average pulp mat consistency during the free mat formation and the compaction stages. The compaction stage significantly contributes to the development of the mat consistency. The final consistency of the mat formation and compaction stages is very much affected by the inlet consistency and the length of the formation – compaction zone, which is shown in Figure 14. The net effect of these phenomena can be seen in the average displacement velocity (Figure 15). The higher the mat consistency when entering the wash liquid displacement stage immediately following the compaction zone, the lower the displacement velocity. Selecting the operational conditions resulting in a certain wash liquid displacement velocity results in a tradeoff between the available washer capacity and washing efficiency.

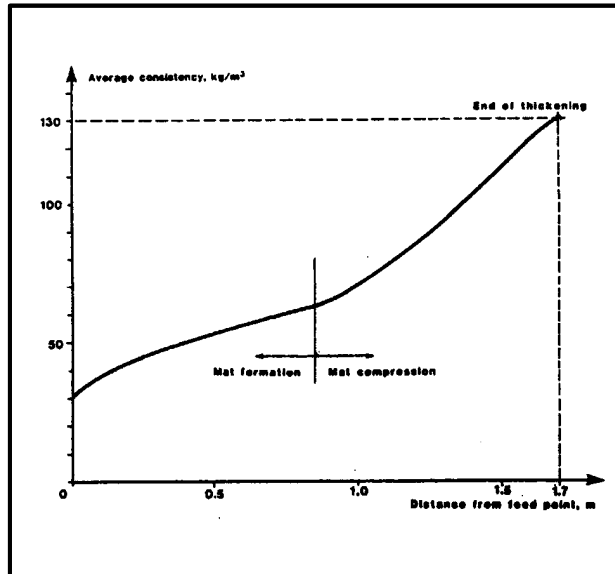


Figure 13: Average mat consistency during the mat formation and compaction stages, [6]

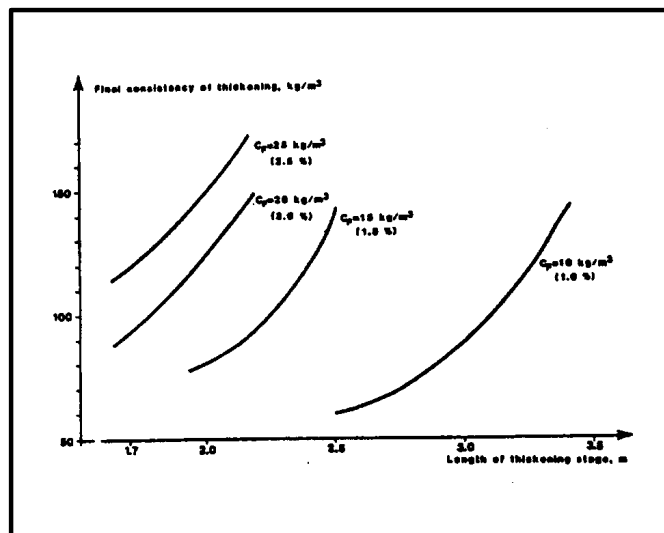


Figure 14: The final consistency after the compaction stage (at the beginning of displacement), [6]

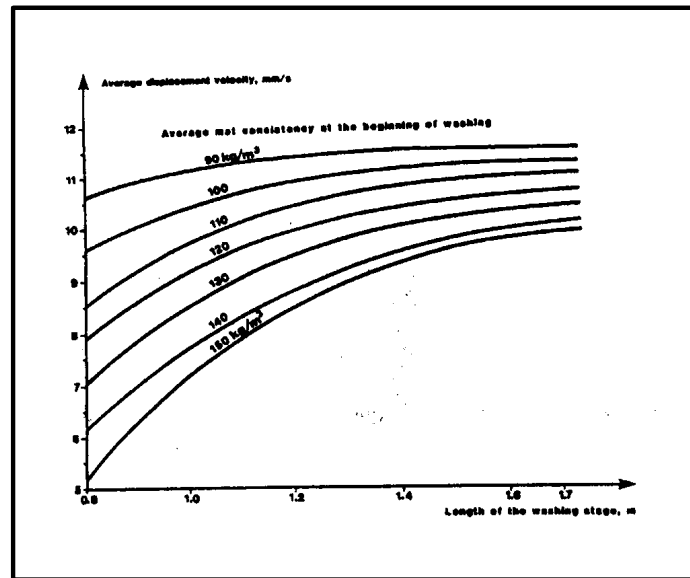


Figure 15: Average displacement velocity as a function of the final compaction consistency, [6]

## 2.5 FACTORS DEFINING THE CAPACITY OF PULP WASHERS, [7]

If the mat formation stage is formulated so that Darcy's law is incorporated into the continuity equation of the filtrate flow, the local mat consistency can be calculated as a function of the distance from the surface of the mat. This was done by Meyer for paper machine conditions, [66]. The same approach was applied in [7] for the mat formation stage in pulp washers under constant pressure conditions. Once the compressibility behavior and the Davies model for the permeability of porous fiber beds are taken into account, a partial differential equation for mat consistency with time and distance can be obtained.

When the mass balance of the fiber material build-up in the mat was incorporated with the partial differential equation describing the consistency behavior, a complete set of model equations was obtained. However, by introducing a dimensionless distance variable it was possible to reduce the original combined partial differential-integral equation into a third order ordinary differential equation. The solving of the equation is easily performed using numerical methods.

When the consistency profile is known, it is possible to further calculate the development of the mat thickness and the specific loading (mass of fibers per unit area of the filter medium) during the mat formation stage. Since the specific loading determines the specific capacity of the washing device (pulp production rate per unit area of the filter medium), the most practical result of the model is directly applicable e.g. for the dimensioning of pulp washers.

By means of the model, it is also possible to study the effects of various parameters and variables on the capacity of pulp washing devices. Basically such calculations serve the same purpose as the purely empirical study of [4].

The only experimental data that is needed to solve the model equations are the specific fiber properties (specific surface and volume) and the compressibility parameters. Such information is available in the literature, e.g. [81]. On the other hand, the determination of these parameters can be readily performed by means of experimental methods.

In study [7], a numerical example was presented in order to model the mat formation stage of a drum washer used for washing unbleached softwood pulp. The effects of inlet (suspension) consistency, available pressure difference and temperature were studied and the selected ranges of the variables were chosen to be representative for brownstock washing and the type of drum filter in question. The first factor to be calculated was the consistency profile in the pulp mat (Figure 16).

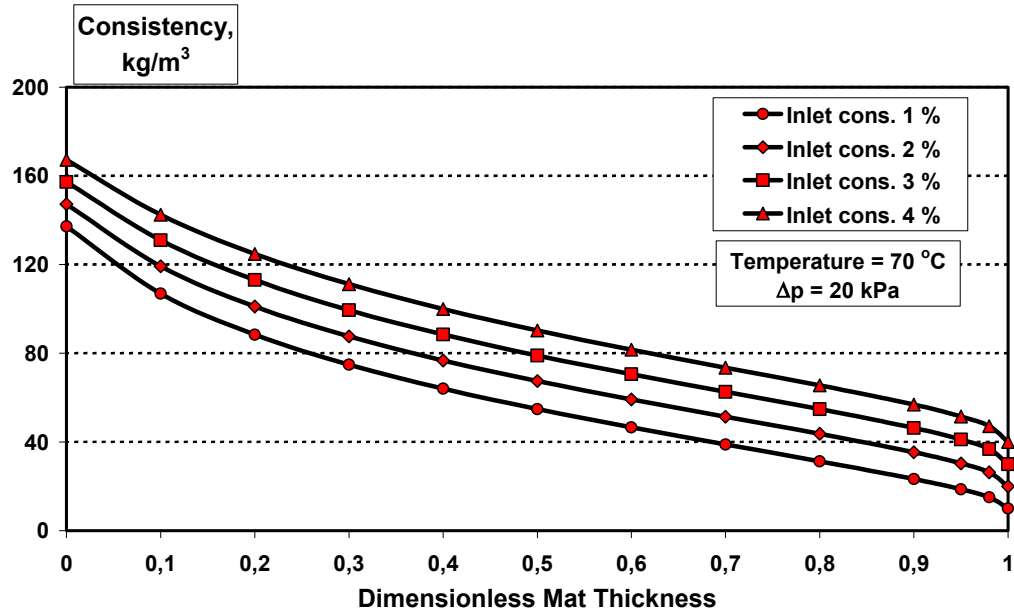


Figure 16. An example of the mat consistency profiles during the mat formation stage as a function of the inlet consistency, [7]

It can be seen that the mat consistency varies a lot as a function of the distance from the mat surface. In addition to that, the profile is quite non-linear. The resulting effect is that the porosity of the mat also has a wide variation, which was illustrated by Figure 2 in [7]. This will further mean that the velocity of the liquor will vary a lot during the subsequent washing displacement stage, thus affecting the local washing efficiency in the pulp mat.

In the study, it was also shown that the washer capacity increases in relation to the square root of the rotation speed of the washer drum. This has also been shown by previous studies, e.g. [5]. Figure 17 illustrates an example of the effect of drum speed and inlet consistency. It is worth mentioning that the diagram is partially misleading regarding the practical possibilities of conventional drum washers, since the drum speed seldom exceeds 2 1/min and the maximum inlet consistency is typically 1.5 – 2 %. In general, increasing the inlet consistency would theoretically be an effective way to increase the washer capacity. Regarding the other variables, the pressure difference and temperature had a lesser effect. The temperature effect is due to the variation in the viscosity of the filtrate. In the studied temperature range (70 – 90 °C) the variation in the viscosity of the filtrate (black liquor) is relatively small.

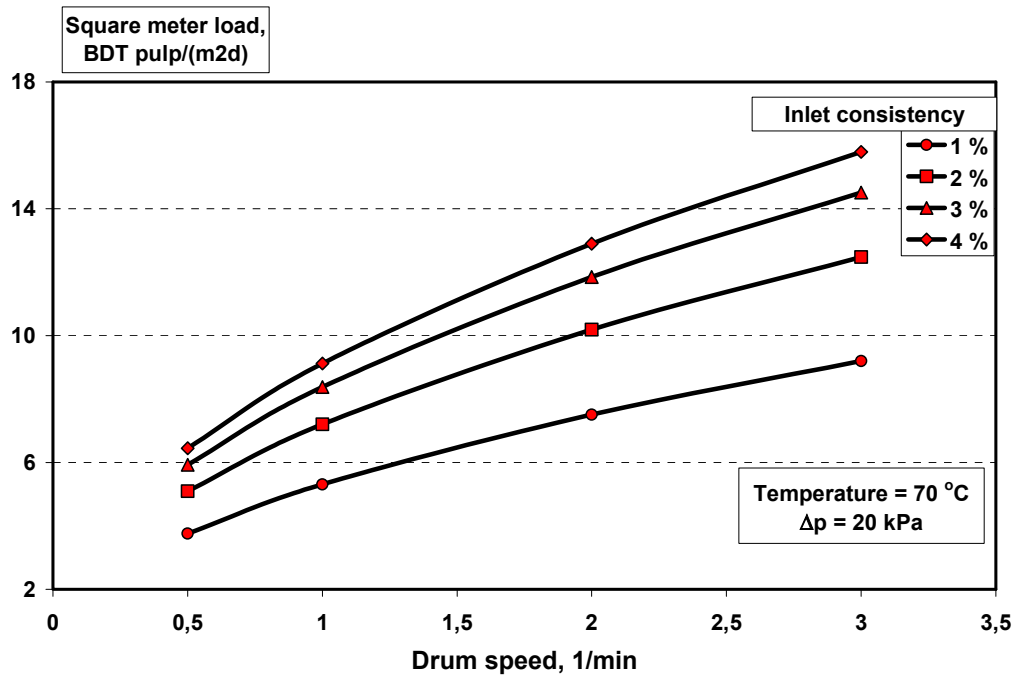


Figure 17: Effect of the inlet consistency and drum speed on the washer capacity, [7]

In addition to the simulations described above on the importance of certain variables, the study also discusses the effect of dispersed gases on washer capacity. In the earlier study, [4], it was shown experimentally that even a small volume percentage share of dispersed air can significantly reduce the available washer capacity. In [7] it was suggested that the additional filtration resistance owing to the dispersed gases is due to the fact that the mat porosity is reduced by the volume fraction of the gases present in the mat. This is expressed mathematically by equation (23):

$$\varepsilon = 1 - a - vc \quad (23)$$

When this new definition of mat porosity is introduced into the model of the mat formation stage, it is possible to model the effect of air on the capacity behavior of a pulp washer. It was shown that the new model predicted a similar reduction in the washer capacity as measured in the experimental study of [4]. However, it should be noted that the air content in the experimental study was based on the initial gas content of the pulp suspension from which the mat was formed. It is reasonable to assume that the gas content in the formed mat is slightly different (probably lower) because some of the smaller gas bubbles are not retained in the mat. Modeling the retention of gas bubbles in the pulp mat was not done in [7] since such a task would have required representative experimental data to confirm the theory.

## 2.6 DISCUSSION OF THE PUBLICATIONS DEALING WITH THE FILTRATION PHENOMENA IN PULP WASHERS

The four studies, [4] to [7], discuss the mat formation stage in pulp washers and thickeners both from an experimental and theoretical point of view. The general idea has been to understand the effects of various parameters on the capacity of washing equipment. In this sense the approach has been technically oriented in order to obtain results for practical purposes, namely the dimensioning of the equipment.



One of the main motivations at the beginning of the studies was to be able to justify the usage of the pulp tester in obtaining a representative small-scale simulation of pulp washers. Since the tester itself and the methods utilized for the experimental simulation of mat formation and washing were seemingly far from industrial practices, it was important to be able to correlate the tester results and theory to actual washing practices. The correlation of the tester results predicting washer capacity with actual production rate figures in identical operating conditions was shown to be good. The regression model obtained for expressing the effect of the most important operational parameters can thus be considered reliable within the range of the studied conditions. The work reported in [4] was extensive and unique in that there have not been any similar experimental studies about the factors affecting the capacity of washing equipment in the published literature.

The second part of the studies presented in [4] dealt with the effect of various operational parameters on washing efficiency. It was shown, for example, that increasing the dilution factor causes a linearly reducing trend in the E-factor. In the same way, increasing the superficial displacement velocity through the pulp mat during the washing stage reduces the E-factor. These effects have been studied by other researchers as well. Kommonen [82] reported mill-scale results about the effect of the dilution factor on the E-factor. Interestingly, his E-factor results showed a slightly increasing trend with a wash filter and a slightly decreasing trend with a diffuser washer and digester Hi-heat washing when the dilution factor was increased. Edwards *et al.* [83] revisited Kommonen's results by modeling pulp washing by means of a dispersion model. They could theoretically confirm the same decreasing E-factor trend as a function of the dilution factor as observed by Kommonen in pulp mill conditions. Cullinan [84] has developed an expression for local washing efficiency using an analogy to local separation efficiency in distillation. The efficiency equation by Cullinan suggests that increasing the displacement velocity means a reduction in local washing efficiency.

Trinh *et al.* [85] have performed an extensive pulp washing study by means of a displacement washing cell. The variables that were investigated, included the superficial displacement velocity. They did not use the E-factor as the measure of washing efficiency but instead expressed the washing efficiency as the recovery yield of the solute. Their observation was that increasing the superficial velocity marginally decreases the washing yield at 15 % consistency (the closest studied consistency compared with the normally used displacement consistency of 10 – 12 %). Finally, Dahllöf and Grén [86] correlated the displacement ratio against the permeability coefficient of Darcy's law. Increasing permeability (which means increasing the superficial displacement velocity in otherwise constant conditions) means the reduction of the displacement ratio.

It can be noticed that there are somewhat contradictory results among various researchers regarding the effect of the dilution factor on the washing efficiency. The comparison of the conclusions is confused by the fact that different expressions of washing efficiency are employed. However, there seems to be a common understanding about the effect of the displacement velocity: the lower the velocity, the higher the washing efficiency that can be obtained. This was also the result of the present study [4].

In study [5], a theoretical approach was presented in order to correlate the filtration results obtained with the pulp tester. A common filtration theory for incompressible fiber beds was applied for the tester as well as for drum filter dimensioning. Even though the assumption of incompressible fiber mats is not correct, the pulp tester results could be explained with an accuracy that is satisfactory from a technical point of view. The assumption of an incompressible fiber bed has also been utilized by von Treiber *et al.* [53] in order to correlate filtration results obtained with a pulp draining apparatus. The filtration data were presented by means of the standard approach by plotting  $t/V$  against  $V$  (where  $t$  is the filtration time and  $V$  the momentary filtrate volume). Hagen and Berg [54] also formulated their filtration model for a flat band filter by assuming an incompressible fiber

bed. The interpretation of their drainage tester results was based on the experimental expression for mat formation time by Wahlström and O'Blenes [55]. A graphical method was presented for determining the permeability parameters based on experimental data.

The benefit of the approach employed in study [5] comes from the fact that by plotting the piston distance of the pulp tester against the square root of the corresponding moment in time, the value for the "filtration coefficient", i.e. the slope of the line, can be directly obtained for use in the washer dimensioning equation, namely equation (12).

The more advanced modeling of the filtration phenomena in the mat formation stage has to be based on the general filtration theories expressed by Darcy's law and the Kozeny-Carman equation for porous beds of particles. In addition to that, compressibility behavior needs to be taken into account. The two presented methods, ([6] and [7]), have a common basis in these theoretical filtration principles. The more complete model presented by study [7] has introduced the equation of continuity for the filtrate flow in a compressible fiber bed. Thus the variation of mat consistency as a function of the distance dimension in the fiber bed can be modeled and calculated. This was not possible in the earlier study [6] where the build-up of the fiber material was expressed as the average mat consistency. However, the approaches are in fact so alike that similar results should be obtained for surface loading.

It is worth mentioning that the most rigorous formulation of the filtration system would also incorporate the continuity equation for the fibers during mat formation, which has been done e.g. in the studies of Stamatakis and Chi Tien [68] as well as Niemenmaa [72]. Also, the model presented by Nordén and Kauppinen [69] considers the velocity gradient of the solids flow. Omitting the continuity equation for the fibers means a simplification in the model of [7] compared with the above-mentioned models. Grén and Hedström [65] have shown that the linear velocity of the fibers during the mat formation stage is about two magnitudes smaller than that of the filtrate flow. Therefore, omitting the continuity equation of the fibers is likely to cause only minor errors in the modeling. Another aspect is the fact that the above-mentioned models become much more complicated to solve compared with the model of [7]. Instead of solving partial differential equations by means of numerical methods, the model of [7] leads to the solving of an ordinary differential equation.

The validity of the model in [7] can be studied by comparing it with the results of the experimental results of study [4]. Comparative calculations have been performed for a situation where:

- Feed consistency = 2 % (20 kg/m<sup>3</sup>)
- Effective pressure difference = 10 kPa
- Temperature = 70 °C

The pulp suspension studied and the corresponding fiber parameters are the same as in the example in [7]. The experimental data of [4] are very closely valid for the same type of fibers and pulp suspension. The correlation shown above (7) was used to obtain the experimental values of the washer capacity. Figure 18 presents a comparison, which shows that the experimental data and the theory of study [7] give almost the same washer capacity figures – especially at smaller drum rotation speeds.

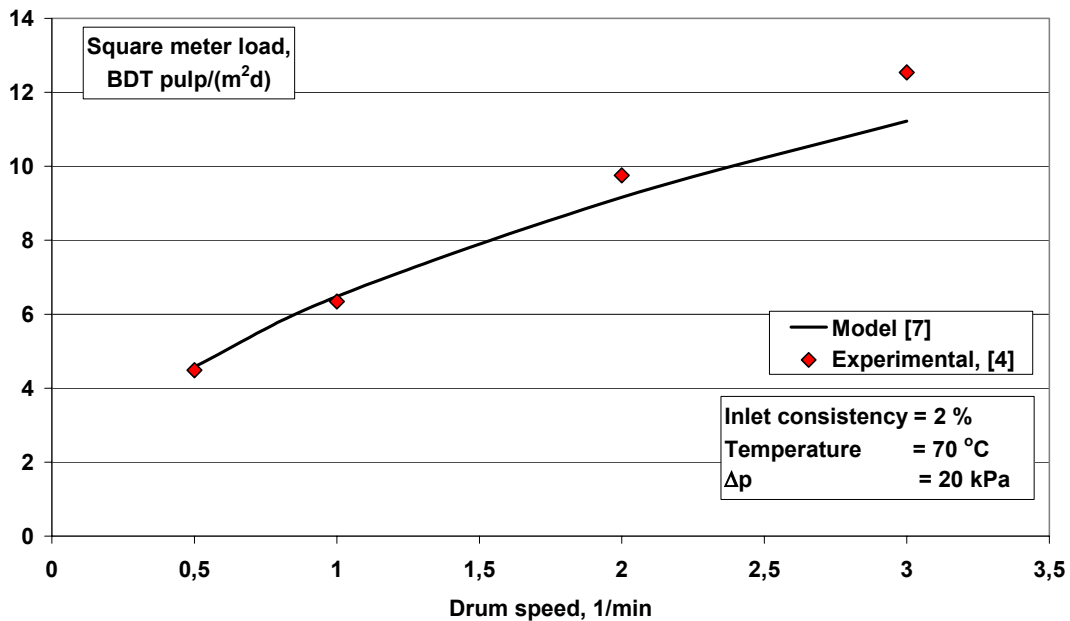


Figure 18: Comparison of the theoretically predicted and experimental drum washer capacity (square meter load) for softwood pulp of kappa number 30.

Another interesting comparison can be made with the numerical example given by Nordén and Kauppinen [69]. They applied their filtration model for calculating the production rate of a drum vacuum filter. Nordén and Kauppinen obtained in their example a production rate of 2.84 kg pulp/s, while the model [7] predicted 2.76 kg pulp/s using exactly the same starting values and pulp parameters. The relative difference is only 3 %.

These two comparisons are certainly not enough to completely validate the model of study [7] even though experimental results and a more rigorous mat formation model could be successfully correlated. More comparisons with experimental data would be needed. Another aspect would be to study other types of pulp washers, not only drum filters. Niemenmaa [72] developed her filtration model for the drum displacer (DD) washer and also compared the predictions with mill data. Unfortunately, the mill data of Niemenmaa gives only capacity figures without any other conditions or parameter values. For this reason, the model of [7] cannot be compared with the mill results of Niemenmaa.

A theoretical approach for taking into account the effect of dispersed air on mat formation was also suggested in [7]. The available literature indicates that the suggested method has not been discussed by other researchers. The presence of gaseous air (or other gases) is assumed to occupy a certain volume in the pulp mat and this volume is not available for the filtrate or fibers. The results of study [4] comprised information about the effect of the dispersed air in the inlet pulp suspension. The air content (as a volumetric share) in the inlet suspension cannot be directly assumed to be the same as that in the pulp mat formed from the suspension. Nevertheless, by assuming that the volumetric share of dispersed air is the same both in the suspension and the mat, a comparison with the modeled air effect and the test results of study [4] can be made. This is presented in Figure 19.

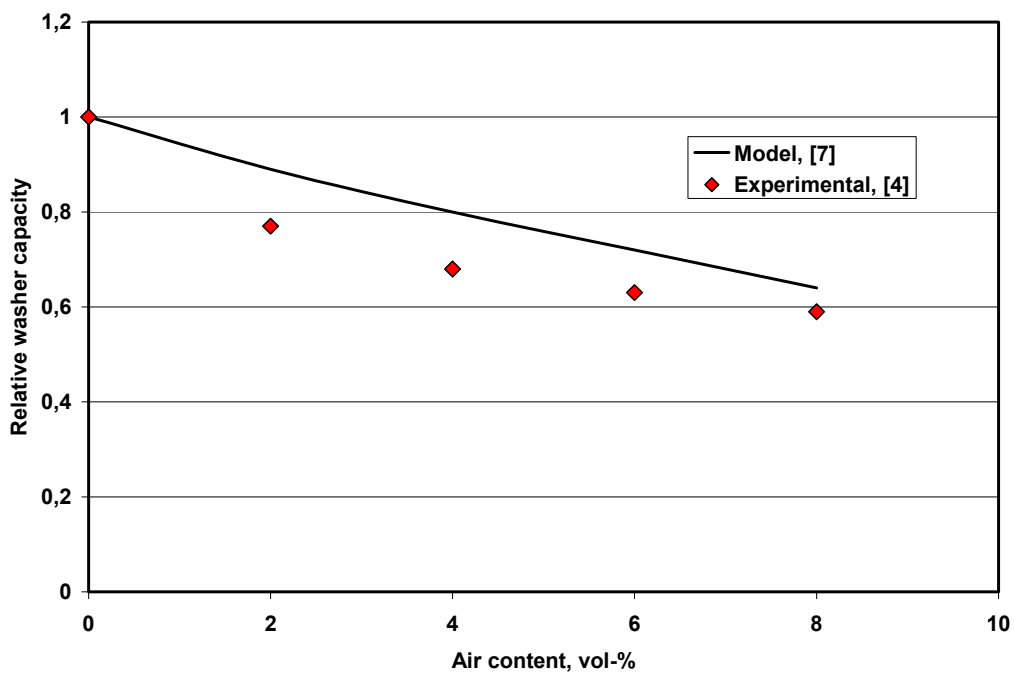


Figure 19: Comparison of the theoretically modeled and experimental effects of dispersed gases on the washer capacity

This part of the new mat formation model also requires more extensive comparison with experimental data. It would be especially important to establish the relation of the gas content in the pulp suspension to that in the mat.

### 3 CONCLUSION

The main results of this thesis can be crystallized into the following statements:

#### Ultrafiltration:

- The mass transfer characteristics of such a macromolecule system as that represented by spent sulfite liquor cannot be expressed in a satisfactory way using the one-dimensional film model. Besides the transversal convection of the solute, the axial convection parallel to the membrane surface also needs to be taken into account. The solving of the more precise model can be performed by means of the presented integral model. The integral model also facilitates a semi-empirical method for correlating experimental ultrafiltration data.
- The mass transfer correlations that have been developed are suitable for the design of industrial ultrafiltration systems.
- The design and optimization of a multistage industrial ultrafiltration system can be effectively performed by means of dynamic programming. Especially in such cases where the optimal number of separation stages increases, the complexity of the problem makes the normally used search algorithms more time-consuming than dynamic programming.

#### Mat formation in pulp washers:

- The pulp tester described provides a representative experimental device and procedure for determining the mat formation characteristics of pulp washers. The simplified theory developed based on the assumption of an incompressible fiber bed can be used for tentative dimensioning of drum filters for pulp washing and thickening.
- The more rigorous filtration model that has been developed, facilitates more accurate dimensioning without the need for any other experimental information than the permeability and compressibility parameters of the fibers in question. The model can be used for studying the effects of the relevant operational parameters on the washer capacity.
- The effect of dispersed gases – such as air – can be taken into account by means of the suggested addition to the developed filtration model. This additional feature is a practical improvement since industrial pulp suspensions very often contain appreciable amounts of dispersed gases, which means additional filtration resistance and a negative factor on the available washer capacity.

The above claims are based on the discussion of the individual articles of the thesis. Regarding the modeling of ultrafiltration, the studied system is challenging because of the fact that the experimental data were obtained on mill scale and with industrial pulp mill spent liquor. Practically all the published work on the mass transfer characteristics of macromolecule ultrafiltration has been based on carefully controlled laboratory studies. Against this background, the modeling results obtained possess such accuracy that the claims presented above can be justified. The recent developments in this field are consistent with the theory developed here. However, the mass transfer models need more cross-checking with experimental ultrafiltration data. In this respect such ultrafiltration systems as e.g. the purification of bleach plant or paper mill effluents would be particularly interesting from an industrial point of view.

The optimization of complicated multistage ultrafiltration systems can be basically performed with many kinds of optimization strategies and algorithms that can be handled by means of modern computer technology. Dynamic programming was shown to be superior to a standard search algorithm. However, the capability of modern computers

needs to be balanced against the benefits of dynamic programming in order to show the real potential of dynamic programming in the situation of today.

The design of industrial washing equipment is an ever-topical matter since new types of washing equipment have been developed, larger pulp mill capacities are called for and also new types of fibers are and will be introduced in the manufacture of pulp and paper . More accurate mathematical models can be made available to describe the filtration phenomena, more reliable designs can be obtained without extensive laboratory and other experimental activities often needed to obtain the required understanding of the design parameters. In this respect, the modeling principles developed here can provide a useful contribution. As mentioned above, the more modern washing equipment (e.g. the drum displacer, washing presses etc.) should be addressed by means of the suggested modeling methods. In order to confirm the validity of the filtration model, mill-scale comparisons would be particularly needed. In general, the filtration model presented here is a simplified one compared with the most rigorous filtration models recently presented. However, the comparisons given above indicate quite good accuracy. In addition, the numerical solution of the model is simple compared with the more complicated filtration models.

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