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MODELLING AND CONTROL
OF COOKING DEGREE
IN CONVENTIONAL AND
MODIFIED CONTINUOUS
PULPING PROCESSES

FACULTY OF TECHNOLOGY,
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TECHNICA



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Abstract

Quality and economical requirements have raised evident need and interest in the industry to further develop continuous kraft cooking. A Kappa number, representing the cooking degree, is one of the few quality measures of cooking, and usually the only one measured on-line. Cooking degree is mainly controlled by temperature, chemical charge, and cooking time. Cooking conditions strongly depend on the packing degree of a chip column in the digester. At the same time, the packing of a chip column is affected by the cooking degree of chips. A typical problem is that the conditions and cooking degree in the process are not known. To achieve better control, more information about the cooking process is required.

The aim of this thesis has been to more accurately describe the cooking conditions and phenomena in the digester scale. Conventional and Downflow Lo-Solids™ continuous cooking processes, producing both softwood and hardwood pulp, were investigated. Information achieved from measurements, and physical and chemical models describing chip scale phenomena, were utilised. Kappa number modelling was based on the use of an optimised and on-line adapted Gustafson's model. Modelling over grade change situations was accomplished by smoothly adjusting the model parameters as a function of temperature change profiles.

Real-time profiles of cooking chemicals, temperature, and lignin and carbohydrates contents of chips within the processes were modelled. These real-time profiles were utilised in the modelling of the chip column's packing degree in the conventional process. Based on the developed models, blow-line Kappa numbers of both processes were predicted. By exploiting the prediction results, a new control strategy for the Kappa number was developed. In the strategy, set points for chemical charge and cooking temperature are iteratively solved by using only the developed prediction models of the blow-line Kappa number.

It was shown that the modelled profiles of wood components and cooking chemicals can give new information regarding the continuous cooking processes. The modelling results are feasible in control purposes, and they also can support the operators' work. In the new control strategy, compared to the widely used *H*-factor based control, chemical concentrations can be more precisely taken into account.

Keywords: chip column, continuous digesters, control, cooking degree, Kappa number, kraft cooking, packing degree, prediction

You miss 100% of the shots you never take.

Wayne Gretzky

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Oulu, July 2006

Rami Rantanen

List of symbols and abbreviations

Roman symbols

$A1-A4$	Measured alkali concentrations, $\frac{g}{dm^3}$ of Na_2O
A, A_1, A_2	Species specific parameters of Arrhenius equation
A_{dig}	Cross-sectional area of digester, m^2
B, B_1, B_2	Species specific parameters of Arrhenius equation
C_i	Concentration of species i , %
C	Carbohydrate or cellulose content, % on wood
Cr	Unreactive cellulose content, % on wood
$C_{i,0}$	Initial concentration of species i , %
$C_{i,bulk}$	Concentration of species i in bulk phase, %
$C_{i,bulk,0}$	Initial concentration of species i in bulk phase, %
CT	One half of chip thickness, m
D_0	Top of digester
D_1-D_6	Calculation zones in digester
$D_{digester}$	Diameter of digester, m
D_i	Diffusivity of species i , $\frac{m^2}{s}$
E_A	Activation energy, $\frac{kJ}{mol}$
H	Hemicellulose content, % on wood
Hr	Unreactive hemicellulose content, % on wood
I_1, I_2, I_x	Calculation zones in impregnation vessel
K	Kappa number
L	Lignin content, % on wood
L_j	Lignin content of species j , % on wood
L^*	Lignin content at intersection, % on wood
LW	Liquor to wood ratio, $\frac{dm^3}{kg}$
P_{chips}	Chip pressure, Pa

$P_{chips,top}$	Chip pressure at the top of calculation zone, Pa
R	Universal gas constant, $8.314 \frac{J}{molK}$
R_1, R_2	Experimental constants in Ergun equation
Ra_i	Reaction rate of species i
T	Temperature, K
$T1-T10$	Measured temperatures, $^{\circ}C$
T_{Dx}	Present temperature in zone D_x , K
$T_{Dx,newmean}$	Mean temperature of new grade in zone D_x , K
$T_{Dx,oldmean}$	Mean temperature of old grade in zone D_x , K
V_b	Volume of bulk liquid phase, m^3
V_c	Volume of chip, m^3
Y	Yield, $\%$
$Y_{correction}$	Constant determining the correction of yield factor
Y_{factor}_{gch}	Yield factor during grade change
Y_{factor}_{new}	Yield factor of new grade
Y_{factor}_{old}	Yield factor of old grade
a	Experimentally determined exponent
b	Experimentally determined exponent
$b_{i,OH}$	Constant relating EA concentration to lignin, $\frac{mol}{dm^3}$
$b_{b,1,OH}, b_{b,2,OH}$	Constants relating EA concentration to lignin, $\frac{mol}{dm^3}$
$diff$	Difference between predicted and target blow-line Kappa numbers
$factorchange$	Degree of temperature change
g	Gravity coefficient, $9.81 \frac{m}{s^2}$
h	Position or height, m
i	Iteration round
k	Calculation moment, min
k_{rc}	Calculation moment in digester's recalculation model, min
k_1, k_2, k_3	Reaction rate constants
k_0, k_1, k_2, k_3	Empirically determined species specific parameters
k_x	Reaction rate constant
k_{il}	Reaction rate constant in initial phase
k_{obl}, k_{1bl}	Reaction rate constants in bulk phase
k_{rl}	Reaction rate constant in residual phase
n_1, n_2	Species specific parameters
p_i	Liquid pressure, Pa
q_i, q_b, q_r	Proportionality constants in initial, bulk and residual phases
$sigma$	Maximum difference allowed between predicted and target blow-line Kappa numbers in the iteration of temperature set point in Kappa number control strategy
t	Time, min

t_{I_n}	Residence time in impregnation vessel's zone I_n , <i>min</i>
t_{dig}	Residence time in digester, <i>min</i>
t_{D_n}	Residence time in digester's zone D_n , <i>min</i>
t_x	Residence time in zone x , <i>min</i>
t_0	Present time, <i>min</i>
v	Superficial velocity, $\frac{m}{s}$
v_{chips}	Chip velocity, $\frac{m}{s}$
x	Distance from chip center, <i>m</i>

Greek symbols

α	Species specific parameter
β	Species specific parameter
ϵ_c	Volume fraction of chips
ρ_c	Chip density, $\frac{kg}{m^3}$
ρ_l	Liquor density, $\frac{kg}{m^3}$
μ	Friction coefficient

Abbreviations

AA	Active alkali
ANFIS	Adaptive neuro-fuzzy inference system
CSTR	Continuous stirred tank reactor
EA	Effective alkali
EMCC	Extended Modified Continuous Cooking TM
EWMA	Exponentially weighted moving average
HW	Hardwood
ITC	Isothermal Cooking TM
LE	Linguistic equation
LS	Least squares regression
ME	Mean error
NIR	Near infrared spectroscopy
NN	Neural network
PCA	Principal component analysis
PDE	Partial differential equation
PLS	Partial least squares
RMSE	Root mean squared error
SW	Softwood
TCF	Totally chlorine free
adt	Air dry ton
rpm	Rounds per minute

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1 Introduction

Pulp is a fibrous material produced either chemically, mechanically or by combining these two methods. Its raw material can be either wood or some other cellulosic material. Most paper and paperboard products in the world are made from wood pulp. Chemical pulp's share of the production is nearly three-quarters (Anon 2006). In Finland, altogether 12.6 million tons of wood pulp was produced in the year 2004, and 61% of that was chemically made (Anon 2006). Sulfate, or kraft process, is the only chemical pulping process used in Finland nowadays. Sodium hydroxide and sodium sulfide are the active cooking chemicals in this process. Most of the kraft pulp is produced in continuously operating cooking vessels called digesters. The success of the continuous kraft cooking process is mainly based on the high quality of the pulp, and good chemical and energy economy.

The main aim in a chemical pulping process is to dissolve lignin from the wood chips to liberate the wood fibers. Delignification is carried out in digesters with the aid of cooking chemicals and heat. The extent of the delignification is typically described with Kappa number, which is a measure of the residual lignin content of pulp. Cooking degree is typically used as a synonym for the Kappa number. In this thesis, cooking degree should rather be understood so, that it covers not only the delignification, but also the degradation of other wood components. During the delignification, other wood components, for example carbohydrates, are also removed to some extent. These undesirable reactions reduce the pulping yield and also affect the papermaking properties of the pulp. Pulping yield is usually calculated as the ratio of oven dry pulp to oven dry input wood (the material is dried to a constant mass at a temperature of about 105°C), expressed as a percent. A total yield in a kraft process is, depending on wood grade and process modification, about 50%.

Both pulping yield and the properties of the pulp, as strength, have an economical importance. In practice, the target value for the Kappa number is specified by the papermaking properties of the pulp. To improve the papermaking properties, various cooking modifications have been developed. In this study, conventional and Downflow Lo-SolidsTM¹ continuous cooking processes are investigated.

¹Lo-SolidsTM is a registered trademark of Andritz Inc.

The kraft pulping process has been widely investigated during recent years, and the optimal cooking conditions in the chip scale are well known. The purpose is, that each individual chip has the proper cooking treatment to ensure uniform and desired quality of the final product. However, a typical problem is that the optimal cooking conditions in the digester scale can not be ensured. This is a consequence from the highly complicated and heterogeneous nature of the cooking process. Large dimensions of the process equipment, highly demanding process conditions, inadequate measurements, quality variations of the raw material and a residence time of several hours are the reasons for this. Thus, the process is more or less like 'a black box', in which conditions are not known.

Vroom's H -factor (Vroom 1957) and Hatton's Kappa number model (Hatton 1973) are widely used in the control of the pulping processes. The H -factor, which describes time and temperature as a single variable, is an input variable to the Hatton's model. Another input variable of the Hatton's model is alkali charge. Despite of the popularity, several weaknesses exist in these models. In Hatton's model, alkali-wood-ratio is used and a real-time chemical profile within the process is not considered. The concept of H -factor and Hatton's Kappa number model requires on-line updating, because of the several, even unmeasurable variations in process conditions and raw material.

1.1 Scope and aims of the research

There has been an evident need and interest in the industry to develop the Kappa number control of continuous kraft cooking. Unquestionably several weaknesses exist in those models, which are widely used in the control nowadays. However, there's also potential to improve the modelling and control of the processes.

The alkali profile is not exactly considered in Hatton's Kappa number model. That has been one reason to improve the modelling of the cooking degree. Another reason has been, that the present models do not give detailed information about the cooking degree profiles. Residence times, for example, are usually calculated based on constant packing degree profile of the chip column, given by equipment supplier. However, packing degree has an important role, when aiming for uniform and desired process conditions. In this study, the packing degree has been described with the volume fraction of chips in the digester. The packing degree is dependent on the compressibility of the chip column. The compressibility depends on chip and liquor levels and flows in the digester, and especially on cooking degree. Thus, it was seen that information about the cooking degree profile would also be useful in the modelling of the residence times.

The primary goal in this study has been to improve the knowledge of the process phenomena in continuous kraft cooking. It was concluded that more information about the cooking degree profile would be essentially important. It looked obvious, that real-time profiles of the cooking degree could enable more accurate modelling of the residence times. Another assumption was, that based on the real-time models, the blow-line Kappa numbers could be predicted. It was also assumed, that

increased information about the processes and resulting models would be feasible in the Kappa number control. The main aims of the research can be listed as following:

1. To model the real-time profile of the cooking degree in two continuous kraft pulping processes.
2. To utilise the cooking degree profile in the modelling of chip column's packing degree and residence times of chips.
3. To predict the blow-line Kappa number based on the real-time models.
4. To develop a new strategy for the control of the Kappa number based on the models of the cooking degree.

Fiber lines of conventional and Downflow Lo-SolidsTM processes, from chips feeding to the blow-line flow after the digester, have been studied. Air removal and impregnation of the chips in the beginning of the fiber lines are assumed to be ideal. However, the non-idealities in these sub-processes have been indirectly taken into account by updating the applied models on-line. Modelling has mainly concentrated in the digester scale, because of the inadequate measurements available of the chip scale. Another reason to define the modelling scope to the digester scale was, that no on-line information about the quality variations of the raw material was available. The accuracy of the present control strategy was not evaluated, and the new control strategy was not compared with the present one. However, the modelling results were compared and verified with the measured blow-line Kappa number.

The research problem is defined and the approach to solve the problem is presented in the next section.

1.2 Research problem

The scope and aims of the study was presented in the previous section. The model presented by Gustafson *et al.* (1983) was chosen to be applied to the modelling of the delignification. It is widely accepted that kinetic theory for the delignification in that model is the most reliable at the moment. The model also has other important features, such as the possibilities to model diffusion kinetics, yield, viscosity of pulp, and the effects of chip size distribution on pulping uniformity and rejects. However, there were questions in the model's usability concerning the studied processes. For instance, the model was originally published for batch cooking of softwood, although it was later applied to continuous cooking by Agarwal (1993).

Input variables of the Gustafson's Kappa number model are: grade, lignin content of chips, chemical concentrations, temperature and cooking time. Because of the lack of the alkali measurements, the alkali profile in the digester was calculated using a model by Pu *et al.* (1991). Consumption of alkali is a function of delignification and carbohydrates degradation. Thus, it was natural to extend the modelling scope to cover the total yield of cooking. The degradation of carbohydrates was calculated using a model by Pu *et al.* (1991). The modelling of packing degree and residence times was based on the model by Härkönen (1984). All models utilised in the study have species and process specific parameters, which had to be optimised and updated.

The research problem was solved based on the following hypotheses:

1. *Phenomena in continuous kraft pulping processes could be modelled on relatively simple models, if kinetic models were optimised, and on-line updated with information obtained from process data.*
2. *Applied pulping models would be feasible for control purposes in the studied pulping processes.*

The research proceeded in a way that at first the phenomena of the cooking processes were modelled in real-time. Process data of two continuous cooking processes was utilised. Promising results of the real-time modelling enabled the development of the prediction models. After that, a new Kappa number control strategy was developed based on the prediction models.

1.3 Contribution of the author

To meet those challenges of the digester scale phenomena in continuous kraft cooking, several research projects have been carried out in the Systems Engineering Laboratory at the University of Oulu. The author has been working as a researcher in those projects. In this thesis, some of the results achieved within the projects are presented. The author has been supervising the M.Sc. thesis of Latvala (2002), Puolakka (2002) and Similä (2003), which were made as a part of the projects. Author's contribution to the results presented in this thesis can be summarised as following:

1. The feasibility of the Gustafson's model has been improved by the author. Optimisations of model parameters, modelling over grade transitions and the model's on-line updating have been developed by the author (Rantanen *et al.* 2003, 2005a). In Latvala (2002), the first version of real-time Kappa number model of conventional process was developed. In that model, the reaction rate parameters were optimised off-line.
2. Experiences of the conventional process were utilised, when Gustafson's model was applied to Downflow Lo-SolidsTM cooking by Similä (2003). Both real-time and prediction models of the Kappa number were developed. The developed models have been further modified by the present author (Rantanen *et al.* 2004, 2005b).
3. The results and optimised parameters regarding to the identification of residence times are contributions of the present author (Rantanen & Kortela 2006). The packing conditions in a conventional digester were studied by Puolakka (2002) and Puolakka & Kortela (2005). In Puolakka, the chips's volume fractions were modelled with simplifying assumptions of the packing conditions.
4. Temperature and alkali profiles of conventional cooking process were modelled by Tervaskanto *et al.* (2003, 2004) and Ahvenlampi *et al.* (2004). Different techniques were used in the modelling of the profiles. The most suitable techniques were chosen and the resulting models were applied and verified by the present author.

5. The modelling of carbohydrates and chemical profiles are contributions of the author.
6. Prediction models of a blow-line Kappa number, in both conventional and Downflow Lo-SolidsTM cooking, were developed by the author (Rantanen *et al.* 2004, 2005a,b).
7. A New Kappa number control strategy, based on real-time and prediction models, was developed by the author (Rantanen *et al.* 2005a). The new strategy is being applied by Metso Automation Oy. A cooking control application, based on the simulation model and control strategy, is being constructed and experimented on in a pulping plant.

Simulation models presented in this thesis can give new information about process conditions. In the new Kappa number control strategy, compared to the present strategy, processes conditions can be more precisely taken into account.

1.4 Structure of the thesis

The structure of the thesis is as follows. Background, scope and aims of the research, research problem and hypotheses, and contribution of the author were introduced above. In Chapter 2, an overview of the continuous kraft cooking is made and the studied processes are presented. Challenges and reaction kinetics of continuous kraft pulping are also discussed in Chapter 2. A literature review of the modelling of the continuous kraft cooking is presented in Chapter 3. Gustafson's pulping model applied in the study, and modelling of chip column's packing degree are also discussed. In Chapter 4, an overview of kraft pulping control is presented and state-of-the-art of Kappa number control is discussed. The development of the simulation framework is presented in Chapter 5. The simulation results of the study are shown and discussed in Chapter 6. Conclusions and suggestions for future work are made in Chapter 7.

2 Continuous kraft pulping

In 1879, Dahl, a German chemist, used sodium sulfate as a makeup chemical for soda pulping to regenerate $NaOH$; actually Na_2S was formed, and unexpectedly, gave a much faster delignification and stronger pulps, since shorter cooking times are used resulting in less carbohydrate degradation. This led to the kraft (or sulfate) process, which is the dominant pulping process. Although related work on the process had been done earlier, Dahl discovered the kraft chemical recovery process, which is perhaps more important than the kraft cooking process. (Kleppe 1970, Biermann 1996)

2.1 Variables and general features

Kraft pulping is a full chemical pulping method using sodium hydroxide and sodium sulfide at pH above 12, at 140–170°C, corresponding to about 800 kPa steam pressure. Total chip residence time is about 3 to 8 hours to dissolve much of the lignin contained in wood fibers. It is useful for any wood species, gives a high strength pulp and is tolerant to bark. Problems with pitch in the pulp are relatively small and process has an efficient energy and chemical recovery cycle. Aside from that sideproducts such as turpentine and tall-oil are valuable. The disadvantages are the difficulty with bleaching compared to sulfite pulps, low yields due to carbohydrate losses, and sulfur in its reduced form provides emissions that are extremely odiferous. (Biermann 1996, Gullichsen 2000a)

Important variables during kraft pulping are (Biermann 1996, Gullichsen 2000a):

1. Wood species (though all species can be pulped), chip geometry and chip cleanliness.
2. Impregnation conditions.
3. Ratio of effective alkali to wood weight.
4. Concentration of effective alkali and liquor-to-wood-ratio.
5. Sulfidity.
6. Time and temperature and their profiles.

Some of the important pulping variables of wood and wood chips are (Biermann 1996, Gullichsen 2000a):

1. Wood species and chemical composition.
2. Age and location in original tree.
3. Length of storage - amount of decay and extractives content.
4. Morphological properties - chip and fiber dimensions and their size distribution.
5. Chip cleanliness.
6. Moisture content.
7. Specific gravity - the density of wood material relative to the density of water.
8. Tension and compression strength properties.

In terms of abundance and suitability for pulping, there are two chief botanical classifications of trees: the softwoods or evergreens and the hardwoods or broad-leaved deciduous trees. The chemistry and anatomy of wood vary somewhat with the species of tree, but there are gross similarities between the two classifications. (Genco 1996)

The feeding and steaming of chips are the first unit operations of the continuous cooking process. Chips are fed from the storage piles or silos into the chip bin in the plant. In the plant, chips are treated with fresh steam, flash steam, or combination of both. The aim of the steaming is to remove entrained air from the chips to ensure optimum digester operations. Accumulation of air and other noncondensable gases in the process can lead to pump cavitation and hydraulic feed line hammering downstream from the pre-steaming operation. In addition, removal of entrained air by pre-steaming promotes uniform penetration of wood chips with the cooking liquor during the impregnation stage of pulping. If the air removal efficiency is low, then liquor will not wet the entire capillary surface area within the chips and nonuniform cooking within the chips will occur. In extreme cases of high entrained air content, the wetted chips will not have sufficient density to generate enough downward force to maintain column movement. In the worst case, the density of impregnated chips is lower than that of the surrounding cooking liquor. This condition causes a buoyant effect that disrupts the chip column movement within the digester. The air removal efficiency of any pre-steaming process will increase with increasing basic chip density and increasing moisture content. (Marcoccia *et al.* 2000)

After air removal chips are impregnated in the impregnation zone. This occurs either in a separate impregnation vessel or in the upper part of digester. Uniform and rapid penetration of cooking chemicals is necessary for two reasons. First, it minimizes within-chip gradients in pulping reaction rates. Second, it increases the density of wood chips. (Marcoccia *et al.* 2000)

The primary role of the cooking zone is to provide retention volume and retention time so that pulping may proceed to the desired extent of reaction. More specifically, the cooking zone must allow sufficient retention time for pulping to proceed to completion without also requiring excessive temperatures, i.e., greater than 165°C. In practice a minimum of 1.5–2.5 hours is necessary. Actual time and temperature requirements are furnish specific. For example, hardwoods generally need less time, temperature, or both for cooking than do softwoods. New digesters equipped with modified continuous cooking technology typically provide more than 5 h of retention time at substantially lower cooking temperatures. (Marcoccia *et al.* 2000)

The aim in various modified pulping processes (Smook 2002), like Extended Modified Continuous CookingTM (*EMCC*), Isothermal CookingTM (*ITC*) and Lo-Solids CookingTM, is to lower the average Kappa number of pulp while maintaining the same pulp strength and yield. Chip preimpregnation, stage wise white liquor addition and lower pulping temperature are all used in these processes (Agarwal *et al.* 1994). Changing the pulping conditions (time, temperature, alkali charge) the uniformity can be improved by affecting the relative rates of lignin reaction and alkali diffusion (Agarwal *et al.* 1994). High concentration of dissolved solids in the cooking liquor is found to retard delignification (Marcoccia 1996a, Lloyd *et al.* 1997, Marcoccia *et al.* 1995). The presence of a high concentrations of dissolved organic solids in the cooking liquor during continuous kraft pulping has been shown to reduce final pulp viscosity, tear resistance, brightness and bleachability (Marcoccia 1996b, Marcoccia *et al.* 1996). A first mill scale application of modified continuous kraft pulping was presented by Johansson *et al.* (1984a,b).

2.2 Studied processes

Two industrial continuous cooking processes producing both softwood and hardwood pulp were studied. In both processes logs are first debarked, chipped and screened in the chip plant before being stored out in chip piles. Chips are fed from the chip piles to the chip bin in the cooking plant either by using air (in the studied conventional process) or a belt conveyor (in the studied Downflow Lo-SolidsTM process). The chips are continuously steamed first in the chip bin at atmospheric pressure and then at low pressure in a steaming vessel where turpentine and gases are vented to the condenser. The chips are then brought to a digester pressure via a high pressure feeder and picked up in a stream of recycled liquor to which a portion of white liquor is added. This stream carries chips to the top of the impregnation vessel, where the recycling feed liquor is extracted from the chips.

Both processes have originally been alike Kamyr processes consisting of a hydraulic impregnation vessel and a steam/liquor phase digester. Production capacities may vary from 1000 to nearly 2000 *adt/day*. Total heights of the digesters are about 60 *m*, diameters 5–6 *m* and volumes about 1300 *m*³. Chips flow co-currently with impregnation liquor through the impregnation vessel, before transferring to the top of the digester. In the following sections the main features of the studied cooking processes are presented. Details of the processes and process control are not presented because of a confidentiality agreement. Dimensions of the process equipment in Figs. 1 and 2 are only indicative. Heights of the calculation zones in the conventional process are, however, comparable. Estimated values for the chemical composition of the wood chips are presented in Table 1.

Table 1. Estimated values for the chemical composition of wood chips in the studied processes (% on wood) (Gullichsen 2000a, Genco 1996).

Wood species	Lignin	Cellulose	Hemicellulose	Others
Softwood	28	45	22	5
Hardwood	22	48	26	4

2.2.1 Conventional cooking process

The conventional Kamyr process is shown in Fig. 1. The process has been substantially simplified by removing almost all of the original liquor circulations and thus only the upper and lower extraction screens in the end part of the cooking zone are used. The digester was examined in six separate zones (D_1 – D_6 in Fig. 1) according to the temperature and alkali measurement points. The first four zones belong to the cooking zone of the digester and the last two to the counter-current washing zone. Softwood chips consist of pine and slightly of spruce. Hardwood chips consist mainly of birch but eucalyptus is also used.

The active alkali concentration of the white liquor ($A1$ in Fig. 1) and the effective alkali concentrations of the digester feed circulation liquor ($A2$) and of the two black liquor extractions from the end of the cooking zone ($A3$ and $A4$) are measured. The white liquor is added to the feed circulations of impregnation vessel and digester. The sulphidity of the white liquor is measured. Before the latest simplifications of the process, alkali measurements were taken from the extraction screens in the upper part of the digester’s cooking zone. These measurements have been utilised in the development of the alkali profile model. All alkali measurements are based on analysed samples.

Temperatures are measured from the liquor circulations, from the heated steam and from several vertical locations of the digester near the digester wall. The temperature profile of the digester’s cooking zone was developed using separate models for all subzones (D_1 – D_4 , Fig. 1). In the digester’s washing zone, there are two temperature sensor circles around the digester. Both circles consist of six temperature sensors on the outside wall of the digester. One circle is right under the lower extraction screens (between D_4 and D_5) and another circle is in the lower part of the washing zone (between D_5 and D_6). These sensors enable the detection of the temperature changes in both the horizontal cross-section and vertical direction of the digester.

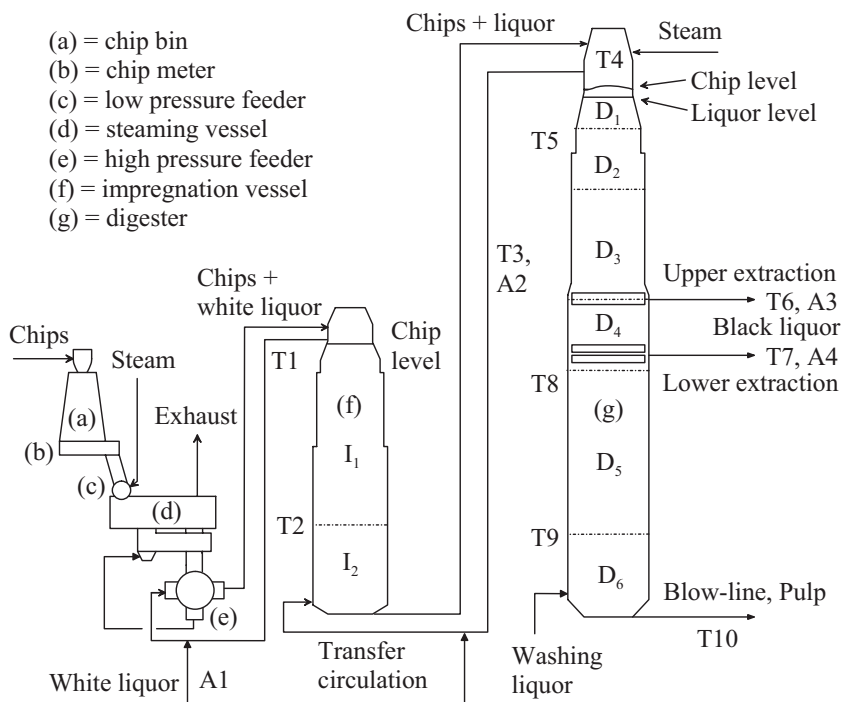


Fig. 1. Equipment, measurements and flows of conventional continuous cooking process from the feeding of chips to the blow-line.

2.2.2 Downflow Lo-SolidsTM cooking process

The Downflow Lo-SolidsTM process is shown in Fig. 2. The digester was examined in four separate zones according to the extractions.

The chips are impregnated in the impregnation vessel and in the first zone (D_1) in the digester down to the upper extraction screens. Between upper extraction and cooking circulation there is a counter-current displacement heating zone (D_2). In this zone, black liquor is displaced with cooking circulation liquor, of which the temperature and alkali concentration are high. The lignin is mainly removed in the comparatively long co-current cooking zone (D_3). At the bottom of the digester is a short washing zone. Softwood chips mainly consist of pine chips with a small amount of spruce chips. Hardwood chips mainly consist of birch chips with a small addition of aspen chips.

The active alkali concentration of the white liquor (A1 in Fig. 2) and effective alkali concentrations of the digester feed circulation liquor (A2), two black liquor extractions (A3 and A5) and cooking circulation (A4) are measured. The white liquor is added to the impregnation vessel's feed circulation, to the digester's feed circulation and to the cooking circulation. The sulphidity of the white liquor is

measured. Temperatures are measured from the liquor circulations and from the heated steam at the top of the digester. A temperature profile from the top of the digester to the cooking circulation was constructed emphasizing the measured temperatures suitably. The temperature profile from the cooking circulation to the blow-line was based on the temperature of cooking circulation.

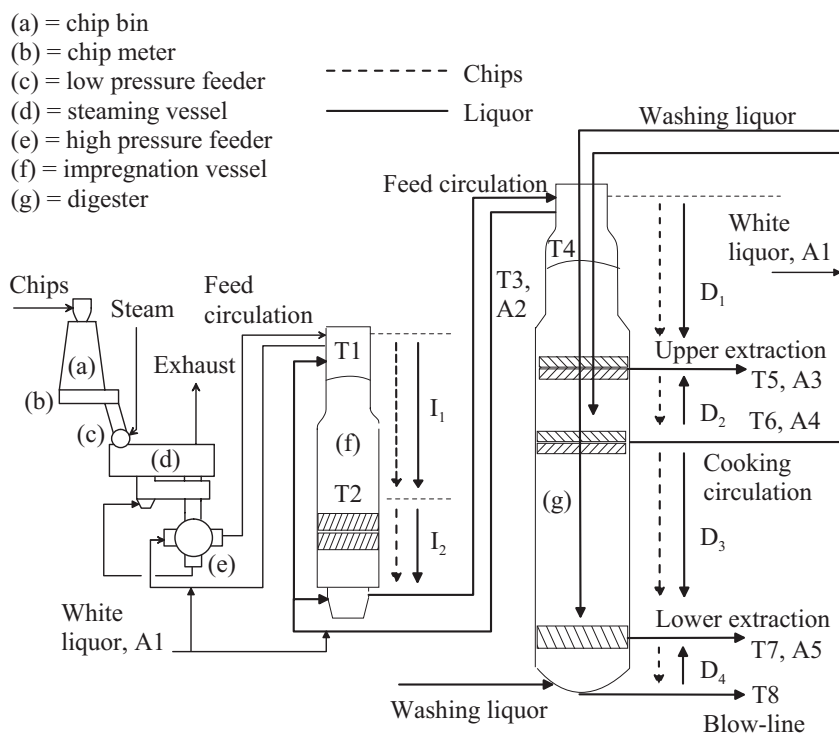


Fig. 2. Equipment, measurements and flows of continuous Downflow Lo-SolidsTM cooking process from the feeding of chips to the blow-line.

2.3 Challenges

Numerous factors affect on the pulping process and pulp's quality. In this section, some of them are addressed. A summary of selected variables, their affects on the process behaviour, and possibilities to affect on those variables is shown in Table 2.

2.3.1 Importance of quality of raw materials

The uniformity and cleanliness of chips has an important economical feature. The biggest disadvantages of poor chip quality are: less usability of fiber sources, non-uniformity of pulping operations, yield and quality losses, offgrade pulp and increased production costs (Akhtaruzzaman & Virkola 1979a, Hatton & Keays 1973). If the size distribution of chips is narrow enough and the amount of small fractions is minor, no screening is needed and obtaining a higher yield from single tree is possible. The chips' size distribution also has a major effect on the pulping uniformity.

Because of the heterogeneous nature of wood, the chemical and morphological characteristics of chips are not uniform. For example, the heartwood, compression wood and branch wood may, because of their denser structure, remain poorly cut and appear in a comparatively higher percentage in thicker rather than in thinner chip fractions. Total pulp yield at a given extent of delignification was found to be 3.5–4.5% higher for pulps from high density chips than for those from low density chips (Lloyd *et al.* 1998). A description of wood heterogeneity would improve predictive capability for rejects and screened Kappa number. (Agarwal *et al.* 1994, Akhtaruzzaman & Virkola 1979c)

Chip thickness is the most critical dimension in kraft pulping, and even more critical at a higher Kappa number. Chip length has a less pronounced influence and the influence of chip width is negligible. (Akhtaruzzaman & Virkola 1979c) Thin chips can be cooked down to very low residual lignin contents without a significant loss of total yield, screened yield, viscosity or strength properties. Also, thin chips need less chemicals, produce less rejects and give better yield selectivity than thick chips (Gullichsen *et al.* 1992, 1995). Chips of different thickness are delignified nonuniformly even in the laboratory conditions, and only sufficiently thin chips (<2 mm) can be uniformly delignified under normal cooking conditions (Gullichsen *et al.* 1992). Over 3 mm thick chips always produce rejects and rejects increase exponentially with increasing chip thickness (Gullichsen *et al.* 1992). Thick chips show steep delignification gradients, because of poor penetration and slow diffusion of reactants and reaction products to and from the inner sections of chips (Gullichsen *et al.* 1992). For commercial chips, the rate limiting penetration and diffusion rates are in the thickness plane of the chip. Commercial chips have variable thickness distributions. Chip thickness values typically are 1–10 mm with the majority of chips in the 4–8 mm range (Marcoccia *et al.* 2000).

With a constant cooking schedule the EA consumption decreases little with growing chip dimensions. When compared at the same Kappa number, the EA consumption increases with increasing chip length or thickness. Thickness has a greater effect

than chip length. At a lower Kappa number, thickness is more critical for alkali consumption. A higher alkali charge leads to a higher consumption of alkali at the same point of delignification. (Akhtaruzzaman & Virkola 1979c)

Chip thickness affects in a way that thick chips are more damaged in industrial chipping than thin chips (Akhtaruzzaman & Virkola 1980b). Chemicals penetrate easier into commercial chips of comparable thickness than into chips cut in the laboratory. This is explained by the finding that commercial chips are uneven on the surface and have more longitudinal cracks than hand made chips (Gullichsen *et al.* 1992). It has been found that the fiber length of unbleached pulp increases with longer chips and decreases with thicker chips under constant cooking conditions (Akhtaruzzaman & Virkola 1980b). The overall delignification is affected only slightly by chip damage attributable to the chipper blade and chip damage has no effect on pulp yield (Lloyd *et al.* 1998), however yield is lower for mill chips (Gullichsen *et al.* 1992). Also, bleached pulp made from mill chips has poorer strength properties than pulp cooked from thin chips (Gullichsen *et al.* 1992).

When chips of different dimensions are cooked together, the smaller chips are easily impregnated and delignification proceeds at a more rapid rate, consuming the chemicals. As a result, the effective concentration of chemicals available for reaction in bigger chips is lower than when different sized chips are cooked separately. The delignification is then dominated by the smaller chips (Akhtaruzzaman & Virkola 1979b), leaving the bigger chips undercooked which appear as screening rejects in the cooking of chip mixtures. The greater amount of rejects leads to a higher total yield with chip mixtures. (Akhtaruzzaman & Virkola 1980a)

Chip size affects the mill digester operation in many ways: 1) it is harder to remove all of the air from thick chips with presteaming and this can cause plug flow problems in a mill continuous digester 2) chip fines and pin chips can interfere with liquor circulation in mill digesters and 3) chip fines will decrease pulp drainage on pulp washers (Becker 1992).

2.3.2 Factors affecting the pulping yield

Heterogeneities in kraft delignification (causing *e.g.* more rejects) may result either from gradients in the pulping chemical concentrations and in temperature, or from variations in wood properties (Hartler & Onisko 1962). Due to diffusion theory, the heterogeneity increases upon an increase in temperature, because the rate of delignification increases relatively more than the rate of diffusion. The former can be expected to be approximately doubled upon a 10° increase in temperature, whereas the latter increases only in proportion to the temperature given in absolute degrees (Hartler & Onisko 1962). Pulping uniformity can be improved by several ways: reducing the pulping temperature, increasing the alkali charge, impregnating standard chips with strong enough white liquor before cooking, prolonging cooking time, cooking only thin chips or screening and/or slicing or crushing the chips (Agarwal *et al.* 1994, Gullichsen *et al.* 1995, Akhtaruzzaman & Virkola

1980a). More uniform pulping opens up a possibility of lowering the target for the average Kappa number without compromising pulp strength (Agarwal *et al.* 1994).

In pulping, both screened yield and total yield are considered. Total yield includes the screening rejects, which are recycled to the digester. At a constant EA charge and H -factor, the screened pulp yield decreases in proportion to the square of the chip length, and in proportion to the square of thickness. At a certain Kappa number level thinner chips always give a higher screened yield and the effect is more marked at a higher Kappa number. An increase in the EA charge results in a lower screened yield at the same Kappa number. (Akhtaruzzaman & Virkola 1979c)

Decreased chip thickness results in an increased rate of delignification with the effect less pronounced the higher the temperature. A higher temperature gives a lower screened yield (Bäckström *et al.* 1996) with the effect more pronounced the higher the Kappa number. Screened yield markedly decreases upon an increase in chip thickness. A higher cooking temperature results in a more pronounced heterogeneity. (Hartler & Onisko 1962)

The total yield at a particular Kappa number of screened pulp is greater above a certain critical point as the thickness increases. As delignification proceeds, the yield drops rapidly with thicker chips, and below the critical point is lower than with thinner chips. It can further be seen that the yield loss below this critical point is much greater for thicker chips. According to Akhtaruzzaman & Virkola (1979b), this is because the thicker chips may reach the residual delignification phase from a higher lignin content. Increasing chip length leads to the same observation as increasing thickness. Longer chips result in a slightly higher yield at a particular Kappa number, above a critical point. (Akhtaruzzaman & Virkola 1980a)

Low alkali concentration is required in the free liquor outside chips if they are uniformly impregnated with enough alkali. Considering rejects and viscosity, the best results are achieved when the liquor concentration outside well-impregnated chips is reduced to the point where thick chips start to produce rejects. Measured data has been compared with theoretically calculated results (Agarwal *et al.* 1994) based on reaction kinetics and mass transfer models. (Gullichsen *et al.* 1995) However, a critical requirement during bulk delignification is that available alkali should never be fully depleted (Marcoccia *et al.* 2000). If the reaction mixture pH falls much below 11, condensation reactions and lignin precipitation will occur.

2.3.3 Quality of pulp

In the production of bleached pulp, a low lignin content is aimed for after kraft pulping as well as after subsequent bleaching. The limit for delignification is often set by pulp viscosity determining the strength properties. The possible extent of delignification is thus determined by the selectivity of the process. (Johansson *et al.* 1984b) The selectivity of a kraft cook can be defined as the ratio of the rate of carbohydrate degradation to the rate of delignification and is usually illustrated as pulp yield or pulp viscosity versus the Kappa number. (Bäckström *et al.* 1996) In the best case, lignin should be selectively degraded but not the carbohydrates (Johansson *et al.*

1984b). The increased pulp viscosity at a given Kappa number makes it possible to extend the kraft pulping without decreasing the pulp viscosity of the fully bleached pulp (Johansson *et al.* 1984b). The higher delignification selectivity in the kraft cook can also be utilized to extend the delignification in a subsequent oxygen bleaching stage, resulting in substantial benefits to both the bleaching economy and the environmental load from the bleach plant effluent (Johansson *et al.* 1984a).

To improve the selectivity of kraft pulping, it has been shown that the following four principles should be satisfied (Teder & Olm 1981, Nordén & Teder 1979, Kubes *et al.* 1983, Sjöblom *et al.* 1983)(Johansson *et al.* 1984b):

1. Alkali concentration should be levelled out, *i.e.* decreased at the beginning of cooking and increased at the end.
2. The concentration of hydrogen sulfide ions should be as high as possible, especially at the beginning of the bulk delignification phase.
3. The concentration of dissolved lignin and sodium ions in the liquor should be as low as possible, especially in the final phase of the kraft cook.
4. The temperature should be low, especially at the beginning and at the end of the cook.

In Lo-SolidsTM process, the sulfide ion concentration is low in the initial phase and at the beginning of the bulk delignification phase. This is a drawback of Lo-SolidsTM process, and would be compensated for by a new chemical recovery process to make two kinds of white liquor with different sulfide ion concentration and to feed them to the different points of the process. (Miyanishi & Shimada 2001)

Kinetic data describing the loss in pulp viscosity during kraft and soda pulping has been reported by Kubes *et al.* (1983). The decrease in viscosity is of first order with respect to the hydroxide ion concentration in all three delignification phases and is independent of the concentration of hydrogen sulfide ions. A decrease in the concentration of dissolved lignin and sodium ions increases the rate of delignification (Teder & Olm 1981, Kubes *et al.* 1983) but does not affect carbohydrate degradation. (Johansson *et al.* 1984b)

The level of dissolved solids in pulping liquors affects the proportion of different carbohydrates in pulp without affecting overall pulp yield (Lloyd *et al.* 1998). In the experiments by Lloyd *et al.* (1998), the pulping selectivity was significantly higher for pulps prepared with laboratory liquors containing low concentrations of dissolved solids. According to the results by Bäckström *et al.* (1996), the cooking temperature has only minor effects to the strength properties of unbleached pulp. The pulp produced at 155°C in both the bulk and residual phases reached a higher brightness level at a lower hydrogen peroxide consumption than the pulp produced at 175°C. The viscosity at a given brightness level after TCF-bleaching was also favoured by low temperature during cooking (Bäckström *et al.* 1996).

In the experiments by Bäckström *et al.* (1996), the viscosity of pulp improved when a low cooking temperature was used. The temperature in the residual phase has a smaller effect on the cooking selectivity. The difference in pulp viscosity resulting of a decrease in cooking temperature in the bulk and residual phases can be explained by the difference in activation energies of the delignification and alkaline hydrolysis. According to the activation energies, a decrease in temperature in the

residual phase is more beneficial for the selectivity than a temperature decrease in the bulk phase (Johansson *et al.* 1984b). However, the results by Bäckström *et al.* (1996) show that the large extent of delignification in the bulk phase determines the total selectivity of the kraft cook. (Bäckström *et al.* 1996)

An increase in the length of chips increases the viscosity of unbleached pulp under constant cooking conditions (Akhtaruzzaman & Virkola 1980b). The viscosity of unbleached pulp is independent of thickness under constant cooking conditions (Hartler & Onisko 1962, Akhtaruzzaman & Virkola 1980b). According to Hartler & Onisko (1962), decreased Kappa number and increased cooking temperature decreases viscosity. In the experiments by Akhtaruzzaman & Virkola (1980b), viscosity suddenly dropped at a higher Kappa number as the thickness increased. That was due to the shift of the transition point between the bulk and residual delignification phases towards a higher lignin content with increasing chip dimensions. Severe cooking with a higher EA charge resulted in a lower viscosity at a particular Kappa number level, implying a higher content of degraded carbohydrates. (Akhtaruzzaman & Virkola 1980b)

A decrease in Na^+ concentration, *i.e.* ionic strength, increases the rate of delignification in the bulk phase (Lémon & Teder 1973) as well as in the final phase. The carbohydrate degradation is almost unaffected by the ionic strength. The ionic strength in the initial phase affects neither the delignification rates in the bulk and final phases nor the amount of residual lignin. In order to obtain good selectivity it is essential to achieve a low concentration of Na^+ in the final part of cooking. (Teder & Olm 1981)

The handsheet tear-tensile relationships are considered appropriate indicators of overall pulp strength. In experiments by Lloyd *et al.* (1998), the pulps prepared from low solid laboratory liquors had over 10% greater tear resistance and on average 10 units higher pulp brightness than corresponding pulps prepared from high solids mill liquors.

Table 2. Summary of selected challenges in kraft pulping and how these can be affected.

Challenges and questions	Answers
Uniformity, dimensions and size distribution of chips	
Has an effect on:	Air removal, penetration and diffusion; uniformity of delignification; fiber and pulp properties
How this can be affected?	With chipping and screening technology, chip storage and chip feeding technology
Possibilities to be affected?	Can be handled with modern chipping and chip feeding technology
Chemical and morphological characteristics of wood	
Has an effect on:	Set special requirements for the temperature and chemical treatment
How this can be affected?	By ensuring equal storage history for chips of same origin
Possibilities to be affected?	It is difficult to measure and control at the plant; How to get enough raw material with uniform quality?
Pulping yield	
Has an effect on:	Economical issues by raw material consumption and capacity of process
How this can be affected?	By improving pulping uniformity and selectivity; by using thinner chips
Possibilities to be affected?	Can be affected with various cooking modifications, and by increasing the information about the cooking degree
Viscosity determining strength properties	
Has an effect on:	Paper making properties of pulp
How this can be affected?	By improving selectivity of cooking; It is favourable to level out alkali concentration, increase hydrogen ion concentration, lower dissolved lignin and sodium ion concentration, and to use lower temperature; By using longer chips
Possibilities to be affected?	Can be affected with various cooking modifications and concepts

Table 3. Exponents and activation energies of Eq. 1. (Teder & Olm 1981)

Phase	a	b	$E_A, kJ/mol$
Initial	0	0	40–60
Bulk	0.7–0.8	0.1–0.4	150
Residual	0.7	0	120

2.4 Reaction kinetics

A chemical reaction in a system involving solid and liquid phases can be divided into five consecutive steps as follows: 1) Diffusion of reactants to the site of reaction. 2) Adsorption of reactants. 3) Chemical reaction. 4) Desorption of products. 5) Diffusion of products away from the site of reaction. In a kraft cook, the reaction takes place in the swollen phase and the reaction sequence above can be simplified so that the dissolution of lignin can be regarded as the result of three consecutive reactions: 1) The diffusion of alkali to the interior of chips. 2) The chemical reaction between alkali and the wood components. 3) The diffusion of products out of the chips. The overall reaction rate is controlled by the slowest of the steps. (Olm & Tistad 1979)

The kraft pulping process can be divided into three separate phases: the initial, bulk and residual phases (Kleinert 1966, Lémon & Teder 1973, Olm & Tistad 1979). In all three phases, the delignification is of apparent first order with respect to the residual lignin in the wood, *i.e.* if the pulping experiments are carried out at a constant liquor composition, straight lines are obtained when the logarithm of lignin content of the wood residue is plotted versus reaction time (Teder & Olm 1981). The slopes, however, differ in the three phases (Axegård *et al.* 1978). The rates of delignification are dependent on the temperature and have dissimilar activation energies (Johansson *et al.* 1984b). The three phases of the cook are obviously governed by various chemical and physical reactions and must be treated separately when the kinetics of kraft pulping are considered (Olm & Tistad 1979).

Chemical reactions in kraft pulping are exothermic. The exothermic heat of reaction for dissolution of wood during kraft pulping is approximately 400 kJ/kg wood dissolved. This translates to a 5–15°C temperature increase. The actual temperature increase because of exothermic heat of reaction depends largely on the liquor-to-wood ratio. (Maccoccia *et al.* 2000)

The effects of OH^- and HS^- concentrations and temperature on the rate of delignification of a kraft cook can be expressed of the type

$$-\frac{dL}{dt} = A \cdot e^{-E_A/RT} \cdot [OH^-]^a \cdot [HS^-]^b \cdot L \quad (1)$$

where L is lignin content of the wood residue (calculated with respect to the original amount of wood), t is time, A is frequency factor, T is absolute temperature (K), E_A is activation energy, R is universal gas constant, and a and b are exponents determined experimentally. The exponents a and b and the activation energy E_A vary from phase to phase; see Table 3 (Teder & Olm 1981).

In the initial phase the amount of carbohydrates decreases rapidly. The dissolution of lignin is small and the extent of delignification seems to be unaffected by process conditions. For a wide range of hardwood and softwood species, testing has shown that 20–30% of the wood dissolves and that 6–7% effective alkali (EA) on wood (as Na_2O) is consumed during the impregnation phase (Marcoccia *et al.* 2000). This represents 40–60% of all wood removal and all alkali consumption in the entire kraft pulping process (Marcoccia *et al.* 2000, Olm & Tistad 1979). The carbohydrate reactions mainly have the characteristic of unavoidable undesirable side reactions (Rydholm 1967). In alkaline media, there are two reactions of importance, namely the hydrolysis of glycosidic bonds, and the successive degradation of the molecules from the end monomers, called 'peeling' (Rydholm 1967). The degradation because of alkaline hydrolysis is affected by the temperature (Kubes *et al.* 1981, 1983); alkaline peeling is not significantly affected by the cooking temperature (Kleppe 1970). Although alkaline degradation by direct hydrolysis has technical importance, peeling is the dominating reaction. The rate of carbohydrate degradation in the initial phase is almost completely determined by the temperature and the OH^- ion concentration, whereas other variables of importance for delignification such as the HS^- ion concentration and the ionic strength have no significant influence (Teder & Olm 1981). (Olm & Tistad 1979)

The change from the initial phase to the bulk phase can be seen when the carbohydrate yield or the alkali consumption is plotted versus the lignin yield in the wood residue (See Figs. 3 and 4.) (Teder & Olm 1981). The limit (in % on wood) between the initial and bulk delignification phase is independent of temperature and effective alkali concentration; but is lowered with increased hydrosulfide concentration. Bulk delignification starts after the solid/liquid reaction mixture of chips and cooking liquor reaches temperatures above 145–150°C (Marcoccia *et al.* 2000). In the bulk delignification phase, the main dissolution of lignin takes place (80% of the lignin in the wood is removed (Bäckström *et al.* 1996)) and the amount of carbohydrates and the alkali concentration in the cooking liquor decrease only slightly; see Figs. 3 and 4. In the bulk delignification phase, the alkali concentration affects both the delignification and the dissolution of carbohydrates. (Olm & Tistad 1979)

The transition point between the bulk and residual phases shifts to a higher lignin content or lignin to carbohydrate ratio when the cooking temperature (Kleinert 1966, Kleppe 1970), the EA charge or the sulphidity (Kleppe 1970) of the liquor is decreased. The transition point shifts to a higher lignin content also with increasing chip dimensions, which can be seen from a sudden drop in viscosity at a higher Kappa number as the thickness increases (Akhtaruzzaman & Virkola 1980b).

In the residual delignification, the carbohydrate content again decreases significantly accompanied by an increased alkali consumption while the delignification rate decreases (Olm & Tistad 1979). Sulphite added to the kraft cooking liquor before the start of the final phase decreases the amount of residual lignin. At low alkalinity, the delignification rate in the final phase is accelerated significantly by sulphite. Sulphite also has the ability to degrade carbohydrates at kraft pulping conditions. Sulphite added in the proper place can, however, improve a modified kraft cook. (Teder & Olm 1981)

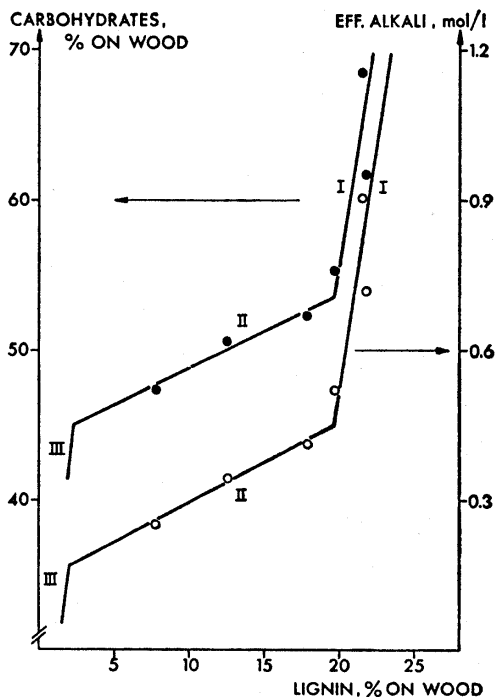


Fig. 3. Carbohydrate yield (% on dry wood) and effective alkali concentration in cooking liquor as a function of lignin content (% on dry wood) (Olm & Tistad 1979).

Lignin remaining in the wood after the bulk phase is a sum of native lignin of chips, and possibly condensed lignin as a result of unfavourable reactions. In the studies by Blixt & Gustavsson (2000), residual lignin was found to be native lignin and unfavourable reactions did not appear in normal cooking conditions. Blixt & Gustavsson (2000) also found out that the activation energy of the residual phase ($152\text{--}156 \frac{\text{kJ}}{\text{mol}}$) is higher than in the bulk phase ($136 \frac{\text{kJ}}{\text{mol}}$). These different activation energies compared to *e.g.* Teder & Olm (1981) (See Table 3.) were justified because lignin solubilisation is more difficult during the residual phase than the bulk phase delignification.

The dependence in the amount of residual phase lignin (*i.e.* lignin content after the bulk phase) in softwood (Norwegian spruce, *Picea abies*) on the conditions in the earlier phases has been studied by Gustavsson *et al.* (1997). An equation for the amount of residual phase lignin was presented, when high liquor-to-wood ratio was used. It was found out that an increase in hydroxide ion concentration and/or hydrogen sulfide ion concentration leads to a decrease in the amount of residual phase lignin, while an increase in ionic strength leads to an increase. Hydrogen sulfide

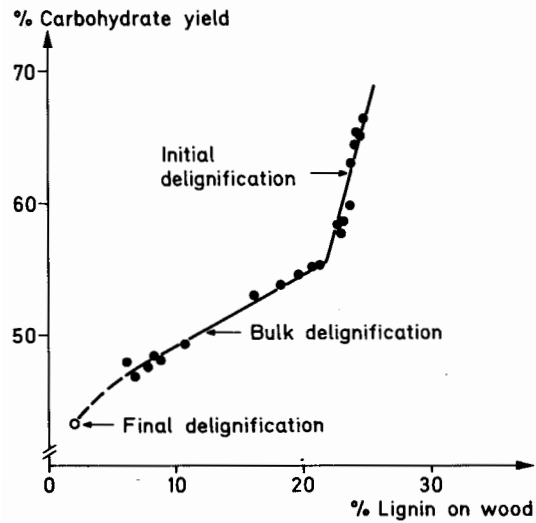


Fig. 4. Carbohydrate yield plotted versus residual lignin content for a kraft cook (Axegård *et al.* 1978).

concentration had a pronounced influence on the amount of residual phase lignin, when hydroxide ion concentration was low. Similar results were also achieved in a previous study by Lindgren & Lindström (1996). Results by Lindgren & Lindström (1996) also indicated that only the hydroxide ion concentration and the temperature have an influence to the rate of delignification in the residual phase.

3 Review of kraft pulping modelling

Kraft pulping has been widely modelled during the last few decades. Efforts have been made to model chemical and physical phenomena, subprocesses, or the whole cooking process in various complexity. Chemical phenomena include *e.g.* penetration and diffusion of cooking chemicals and consumption of cooking chemicals via chemical reactions. Physical phenomena include *e.g.* flows of chips and liquor, chip and liquor levels, and chip column's packing degree. Review of pulping models is made in Section 3.1. Kinetics of kraft pulping are described in more detail in Section 2.4. Applied pulping model by Gustafson *et al.* (1983), is presented in Section 3.2. Modelling of chip column's packing degree is presented in Section 3.3.

3.1 Pulping models

Early investigations of kraft pulping kinetics have been made by Vroom (1957) (See Section 4.2.1.), Hatton (1973) (See Section 4.2.2.), Wilder & Daleski (1965), Kleinert (1966), Kerr (1970), Kerr & Uprichard (1976) and Lémon & Teder (1973). Lignin and carbohydrate kinetics were studied as a function of temperature and alkali charge by Kleinert (1966). The results showed that two overall reaction mechanisms of first order exist in the kraft pulping, namely bulk and residual delignification having different rate constants. Wilder & Daleski (1965) presented an equation for kraft delignification, in which Arrhenius equation was used to present the temperature dependence. Other variables were $[OH^-]$ and $[S^{2-}]$ ion concentrations. In the model by Kerr (1970) and Kerr & Uprichard (1976), Kappa numbers were predicted as a function of H -factor (Vroom 1957). Initial effective alkali concentration, sulfidity, chip size, chip moisture content, and liquor-to-wood ratio were also used as input variables.

Two broad main approaches to model the reaction kinetics can be found. These approaches can be divided into families of '3-stage models' derived at the University of Washington and 'Purdue models' derived at the Purdue University. The pioneering work of both approaches was done by Lémon & Teder (1973) (Andersson 2003). In the '3-stage models', a lignin and carbohydrate dissolution is modelled in three

phases: initial, bulk and residual. In the 'Purdue models', the solid materials of wood chip (lignin, hemicelluloses and celluloses) are divided into several components, and each component is treated separately. The development of Gustafson's model (Gustafson *et al.* 1983) has been based on the studies of several researchers: Pu (1991), Agarwal & Gustafson (1997), Walkush & Gustafson (2002). Gustafson's model, which belongs to the family of 3-stage models, is presented in more detail in Section 3.2.

The original Purdue model was developed by Smith & Williams (1974, 1975a,b), where the digester was approximated by a series of continuous stirred-tank reactors (CSTRs), with the external flows entering and exiting those CSTRs where the heaters and extraction screens were located. Three phases (solid, entrapped liquor, free liquor) were defined for each CSTR. The original model was further improved by Christensen *et al.* (1982, 1983a). Improvements were related to kinetic parameters, unreactive concentrations of carbohydrates and reactant consumption. The wood species were divided into hardwood and softwood, and a rate multiplier was used to adjust the kinetics for the different species. As an extension of above mentioned models, Wisnewski *et al.* (1997) developed a model of continuous digester. What was derived was a lumped-parameter approximation. This was used to describe the flow transport mechanism, and a model based on the fundamental principles of mass and energy was developed. In that model, fewer simplifying assumptions in the model derivation are needed, because of definition of mass bases and volume fractions. Today, the Purdue model is the industrially accepted digester model that can be used to predict temperature and mass component profiles (Malkov & Leavitt 2003).

A fundamental model providing dynamic characteristics required to track both species and transition behaviour without severe simplifying assumptions on solid phase behaviour has been presented by Kayihan (2002), Kayihan *et al.* (2005). Physical and chemical phenomena, including chip size distribution, dynamics of chip column movement, diffusion and reaction fluxes between entrapped and free liquor, and relative velocity differences between chip column and liquor flows are included in the model. An approach of moving coordinates of the moving plug instead of stationary coordinates (like in the CSTR approach) has been used to achieve an efficient numerical procedure. Reaction kinetics are calculated like in Wisnewski *et al.* (1997). Inter-particle solid and liquor density gradients are calculated in a manner similar to Gustafson *et al.* (1983). Dynamic compaction calculations in that model are based on Michelsen (1995).

The extended Purdue model (Wisnewski *et al.* 1997) is further developed and integrated into dynamic process simulation software (Savolainen 2004). The extensions made are: 1) the chip compaction calculation as described by Härkönen (1987), 2) chip and liquor level calculations and 3) an alternative reaction kinetic presented by Gustafson *et al.* (1983). The chip compaction calculation uses the Kappa number profile and chip column pressure to calculate new values for the chip volume fractions in each calculation volume of the digester. This new calculation of the compaction profile then affects the chip and liquor levels of the digester.

In Funkquist (1995, 1997), grey-box modelling is used to identify a continuous Kamyrgester. A basic deterministic partial differential equation (PDE) digester

model is derived from first principles including mass and energy balances. The description of overall mass movement is simplified, including no momentum balances. The chip compaction profile is modelled as piecewise linear, and it is compared to the profile presented by Härkönen (1987). The method of orthogonal collocation is utilised to reduce the infinite dimensional system of modelling equations into a finite dimensional state space approximation, which is suitable for the numerical calculations involved in identification. The model parameters were estimated to get a deterministic model. Physically based stochastic disturbances were introduced to explain some of the discrepancies between the model and measurements over a large operating range.

The physical flows and packing degree in the digester have been studied by Johnsson (1971) (first model in terms of mass and energy balances), Härkönen (1987) (first model considering fluid dynamics in the continuous digester; see Section 3.3), Michelsen (1995) and Laakso (2004).

Michelsen (1995), Michelsen & Foss (1996a,b) have developed a dynamic model to improve the understanding of the Kamyrdigester, especially the complex interactions between the vertical motion of the mass and the chemical reactions. The model is a dynamic first principles model based on mass, momentum, and energy balances. The vertical motion of chip column is described by the two-phase flow concept, first proposed by Härkönen (1987). The kinetic model may be viewed as a simplified version of the model by Christensen *et al.* (1983a). The kinetic model is rather simple in that the wood solids are divided into one lignin and one carbohydrate component. Further, the liquor consists of one alkali component, a component of dissolved solids, and water. The liquor is divided into entrapped and free liquor. The chips may be only partially penetrated by liquor, that is air may be entrapped in the chips as well. The chemical reactions inside the chips are of first order as a function of mass densities of free and entrapped liquor, and mass fractions of lignin and carbohydrates in wood. The interphase diffusion rate coefficients for the alkali and dissolved solids are derived from Gustafson *et al.* (1983) and Christensen *et al.* (1983a). The simplification in the kinetics make the model only valid for Kappa numbers in the range between 50 and 150 (Wisniewski *et al.* 1997). The model predicts Kappa number, the compaction of chips, velocities of chips and free liquor, and the chip level.

Based on the mass and energy flows in the digester and the integration of the ideas behind the work done by Smith & Williams (1974), Christensen *et al.* (1982), Härkönen (1987), Saltin (1992), Agarwal (1993) and Gustafson *et al.* (1983) a simulation model of Lo-SolidsTM cooking was developed by Miyanishi & Shimada (2001). The model is a static one that can be used for steady-state simulation of several types of modified continuous cooking systems. The kinetic model is based on the Purdue model (Smith & Williams 1975a, Christensen *et al.* 1983a). A chip is assumed to have six components: high and low reactivity lignin, cellulose, galactoglucomannan, xylan and extractives. Extractives are assumed to dissolve immediately at the beginning of a cook. Removal of other five components is modelled by five differential equations in the phases of initial, bulk and residual delignification. The three-phase (wood components, entrapped liquor, free liquor) model in calculation sections is used. The concentration in the chips is assumed to be constant,

because mill chips have a wide range of chip thickness, and concentration gradient measurements to verify the results are difficult to perform. Flow characteristics in the digester are modelled using simplified one-dimensional model, derived by Saltin (1992) based on Härkönen's data and theory. Void fraction in the chip column, the liquid pressure, pressure on the chip column, the flow of chips and the flow of liquid are calculated. (Miyaniishi & Shimada 2001)

Most of the pulping models consider the kinetics of softwoods. In few studies, hardwood pulping is investigated, and in the most cases the species has been eucalyptus. Hardwood pulping is studied by Mortha *et al.* (1992) (hybrid poplar), Giudici & Park (1996) (*Eucalyptus saligna*), Saucedo *et al.* (2002) (carbohydrates degradation of red oak) and Saucedo & Krishnagopalan (2002) (delignification and alkali kinetics of oak). Grade transitions are studied by Christensen *et al.* (1983b), Puig *et al.* (2000), Doyle & Puig (2001), Bhartiya *et al.* (2003) and Kayihan *et al.* (2005).

Soft computing methods have been used in several studies. Dayal *et al.* (1994) have developed empirical predictive models for Kappa number using both methods of neural networks and partial least squares (PLS). Dynamics of 22 process measurements, including a Kappa number at lag $k - 1$, were incorporated into the models. Based on the results of PLS modelling (only two latent vectors were necessary to explain most of the Kappa number sum of squares), the number of input variables was reduced to 5, and the new PLS and neural network models were developed. These variables were Kappa number at lag $k - 1$, active alkali concentration of upper cooking zone, white liquor's flow rate, feed rate of chips and flow rate of blow-line. When predicting the blow-line Kappa number, similar results were achieved with both PLS and neural network models. Based on results achieved, a new strategy for chip level control was developed, resulting as smaller variations of upper cook zone active alkali, and Kappa number.

The use of neural networks and linguistic equations (LE, a non-linear scaling method) in Kappa number modelling was compared by Murtovaara *et al.* (1999). In the study, alkali, total dissolved solids, lignin content, and temperature were measured on-line from several circulation flows in a conventional continuous digester. It was found that in training, both methods seem to learn process behaviour in a similar manner. Differences come out in using the models in a process environment. Neural network models are suitable for processes where process conditions are stable and there is a lot data available. The linguistic equation model works better and it is more suitable for the prediction of the Kappa number, because it is not as sensitive for the changes in process conditions. Linguistic equations also offer tools for modelling the system in several operating points and thus developing adaptive models.

In Musavi *et al.* (1999), the blow-line Kappa number was predicted using a neuro-fuzzy system. The system consists of five layers: two crisp inputs, input fuzzy sets, IF-THEN-rules, output fuzzy sets, and one crisp output. Inputs are digester chip level and blow flow, output is the blow-line Kappa number. Past values of 1.5 hours of each variable were used. A hybrid model was used in Aguiar & Filho (2001) to model the blow-line Kappa number. First principles kinetic equations were used to

model the Kappa number. The Kappa number was fed with other process variables to the neural net, yielding final Kappa number.

A dynamic radial basis function neural network model based on genetic algorithms was used to predict blow-line Kappa number in a continuous digester (Alexandridis *et al.* 2002, Sarimveis *et al.* 2004). The model presented was used to build a discrete dynamic model that can predict the value of the Kappa number on a hourly basis. Although there are numerous variables that affect the process, only the most important of them were selected as inputs to the model. These were the hourly measured temperatures in three different zones of the digester. Past values of up to 12 h for each variable were used, summing up to a total of 36 input variables. The results shown in the papers have good correlation between measured and predicted Kappa numbers.

Raw material variations have a great impact on the Kappa number in blow-line. A neural network-based strategy for detection of feedstock variations in a continuous pulp digester is presented by Dufour *et al.* (2005). The following feedstock properties were considered: moisture content of the wood chips fed to the chip bin (only measured property), densities of five wood chip components, and densities of EA and HS in white liquor. The measured EA and HS content of the upper extraction liquor were selected to infer the magnitudes of the eight quality descriptors that affect the Kappa number. The digester model, used as a plant, and modelling assumptions are similar to those presented by Wisniewski *et al.* (1997). In the study, the generation of training and validation data sets are presented. The neural network-based software sensor has been simulated with raw material variations, and changes in the manipulated process variables (chip flowrate, upper extraction flowrate, and cook temperature).

3.2 Gustafson's model

At the beginning of cooking, it is assumed that the chip phase is full of cooking liquor and the alkali and sulfide concentrations are uniform throughout. During the heating of chips and liquor, pulping reactions begin that produce carbohydrate degradation products that neutralize alkali as they diffuse out of the wood (Hartler & Onisko 1962). Alkali is transported from the liquor phase to the chip surface and then diffused into the chip to replace the alkali consumed by the degradation products. The sulfide ion concentration is assumed to remain constant, and therefore it does not diffuse into the chip. Because of the low consumption of sulfur (Rydholm 1965) and because of the low delignification dependence on sulfur (Eq. 11) the assumption that the sulfide concentration is constant is good, provided that initial sulfide concentration is above a value corresponding to about 20% sulfidity. Both the sulfide ions and hydrosulfide ions are regarded as the sulfide. This is done because $[S]$ in the bulk delignification rate equation, Eq. 11, is the total sulfide sulfur concentration (Lémon & Teder 1973). At the end of cooking, and after the pulp has been washed, all that remains in the pulp are the bound wood components. (Gustafson *et al.* 1983)

The original model (Gustafson *et al.* 1983) was developed and verified against the data of softwoods. Mortha *et al.* (1992) have presented an equation for the bulk delignification of hybrid poplar. In the model development, several simplifying assumptions were made (Gustafson *et al.* 1983):

1. The liquor penetration rate is infinite.
2. The chips are isothermal.
3. The pulp chips are one-dimensional and chip thickness is the critical dimension.
4. The liquor phase is homogeneous and well stirred.
5. Wood is divided into lignin, carbohydrate, and acetyl.
6. Pulping reactions are irreversible.

A mass balance on the chip, with the above assumptions, gives equations

$$\frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x} \left(D_i \frac{\partial C_i}{\partial x} \right) - Ra_i \quad (2)$$

$$\frac{\partial C_i}{\partial x} = 0 \quad (\text{chip center}) \quad (3)$$

$$D_i \frac{\partial C_i}{\partial x} = k_i (C_{i,bulk} - C_i) \quad (\text{chip edge}) \quad (4)$$

$$C_i = C_{i0} \quad (t = 0) \quad (5)$$

where C_i is concentration of the i species, $C_{i,0}$ is initial concentration of the i species, $C_{i,bulk}$ is concentration of the i species in the bulk liquid phase, D_i is diffusivity of species i , k_i is mass transfer coefficient for species i , Ra_i is reaction rate of species i , t is time, and x is distance from the chip center. (Gustafson *et al.* 1983)

A mass balance on the bulk liquid phase gives equations

$$\frac{dC_{i,bulk}}{dt} = -D_i \frac{\partial C_i}{\partial x} \left(\frac{V_c}{CT \cdot V_b} \right) \quad (6)$$

$$C_{i,bulk} = C_{i,bulk,0} \quad (t = 0) \quad (7)$$

where V_c is volume of the chip, V_b is volume of the bulk liquid phase, CT is one-half of the chip thickness, and $C_{i,bulk,0}$ is the initial concentration of species i in bulk phase. (Gustafson *et al.* 1983)

Gustafson *et al.* (1983) also present an equation for the diffusivity of alkali. This information could be used to investigate the conditions inside the chips, and to predict the amount of yield and rejects.

Table 4. Species specific parameters in Eqs. 10, 11 and 14 (Gullichsen 2000a).

Phase of delignification	Parameter	Value
Initial	k_{il}	1
Bulk	k_{obl}	0.15
	k_{1bl}	1.65
Residual	k_{rl}	2.2

3.2.1 Delignification

The kinetics of kraft pulping is divided into three phases: initial, bulk, and residual (Kleppe 1970, Kleinert 1966, Nordén & Teder 1979). The initial phase is characterized by rapid delignification, significant hemicellulose degradation, and large alkali consumption.

The delignification rate in the initial cooking phase is independent of the effective alkali and hydrosulfide concentration (Olm & Tistad 1979). Thus, it is a zero order reaction with respect to the effective alkali and hydrosulfide concentration, presented as

$$-\frac{dL}{dt} = k_x \cdot L \quad (8)$$

where $-\frac{dL}{dt}$ is the rate of delignification, L is residual lignin content calculated on the original wood weight % on wood and k_x is rate constant which is independent of the lignin content but a function of the liquor composition and temperature (Olm & Tistad 1979).

The lack of alkali or sulfide dependence in Eq. 8 can be explained by the initial phase chemistry. Initial phase delignification is attributed to the cleavage of phenolic α - or β -aryl ether groups. This cleavage rapidly takes place at pulping temperatures provided the pH is above 12 (Gierer 1980).

Based on data by Olm & Tistad (1979), rate equation for the initial phase delignification is presented in Gustafson *et al.* (1983) to be

$$\frac{dL}{dt} = 36.2\sqrt{T}e^{(-4807.69/T)}L \quad (9)$$

where T is temperature (K).

The delignification rate equations applied in this thesis, are shown in Eqs. 10, 11 and 14. The original values of the model parameters k , A and B are shown in Tables 4 and 5. The rate equation for the initial phase delignification is (Gullichsen 2000a)

$$\frac{\partial L}{\partial t} = k_{il}e^{(17.5-8760/T)}L \quad (10)$$

where k_{il} is a species specific constant.

Table 5. Original parameters of Arrhenius equation (Eq. 11) in bulk phase. (Gullichsen 2000a)

Parameter	Original value, SW
A_1	35.5
B_1	17200
A_2	29.4
B_2	14400

The transition from the initial phase to the bulk phase in kraft cooking takes place at a lignin content of about 22% of wood, and it is independent of temperature, sulphide concentration and alkali concentration (Olm & Tistad 1979); (Rekunen *et al.* 1980). The rate equation for the bulk phase delignification of softwood, based on Teder & Tormund (1973), is

$$\frac{\partial L}{\partial t} = k_{obl}e^{(A_1-B_1/T)} [OH^-] L + k_{1bl}e^{(A_2-B_2/T)} [OH^-]^{0.5} [S^{2-}]^{0.4} L \quad (11)$$

where $[OH^-]$ is hydroxyl ion and $[S^{2-}]$ is sulphide ion concentration ($\frac{mol}{dm^3}$) and k_{obl} , k_{1bl} , A_1 , A_2 , B_1 and B_2 are species specific constants (Gustafson *et al.* 1983). The equilibrium



is always shifted so far to the right that the presence of sulphide ions can be ignored (Teder & Tormund 1973). The concentration of effective alkali determined is thus equal to OH^- concentration and the concentration of sulphide determined equal to HS^- concentration. (Teder & Olm 1981)

The rate equation for the bulk delignification of hybrid poplar is

$$\frac{\partial L}{\partial t} = 6.45 \cdot 10^{17} e^{(-36500/RT)} [OH^-] [HS^-]^{0.62} L \quad (13)$$

where (HS^-) is hydrosulphide ion concentration ($\frac{mol}{dm^3}$) (Mortha *et al.* 1992).

In the study by Mortha *et al.* (1992), the activation energy for kraft pulping was found to be $36.5 \frac{kcal}{mol}$. The relatively high reaction order and activation energy were explained so, that they may be attributable to the absence of diffusion limitations common in kraft pulping. The low density and thin cell walls of the hybrid poplar will minimise any diffusion resistance.

The relative reaction rate and the activation energy are highest in the bulk phase. The hydroxyl ion and hydrosulphide ion concentrations have a considerable impact on the rate. The lignin content of the pulp at the transition point from the bulk phase to the residual phase may vary between 2.5% on wood (Rekunen *et al.* 1980) and 1.1% on wood (Kleinert 1966). The relative rate decreases, and the effect of hydroxyl ion concentration decreases in the residual phase.

The rate equation for the residual phase delignification (Nordén & Teder (1979), data from Kleinert (1966)) is

$$\frac{\partial L}{\partial t} = k_{rl}e^{(19.64-10804/T)} [OH^-]^{0.7} L \quad (14)$$

where k_{rl} is a species specific constant for residual delignification (Gustafson *et al.* 1983).

The reaction rates presented in this section will not apply for all species, even among softwoods, under all conditions (Gustafson *et al.* 1983). The conditions in the initial phase are found to significantly affect the delignification rates in the bulk phase (Wilder & Daleski 1965, Lémon & Teder 1973). The bulk delignification rate measured for spruce (Kleinert 1966) is about 20% higher than the value measured for pine (Wilder & Daleski 1965, Lémon & Teder 1973). The form of the rate equations is consistent with the wood chemistry for softwoods. That is that differences in the kinetics between species may be accounted for by adjusting the rate constants (Gustafson *et al.* 1983).

3.2.2 Carbohydrate kinetics

According to Yllner *et al.* (1957), Aurell & Hartler (1965), Kleinert (1966) and Olm & Tistad (1979), the carbohydrate reaction rate for each phase is a linear function of the lignin reaction rate. The carbohydrate degradation rate is almost completely determined by the OH^- ion concentration and the temperature, whereas other variables of importance for delignification such as the HS^- ion concentration and the ionic strength have no significant influence (Teder & Olm 1981). In Gustafson *et al.* (1983), rate equations for carbohydrate degradation are presented for initial (Eq. 15), bulk (Eq. 16) and residual (Eq. 17) phases as

$$\frac{dC}{dt} = q_i[OH]^{0.11} \frac{dL}{dt} \quad (15)$$

$$\frac{dC}{dt} = q_b \frac{dL}{dt} \quad (16)$$

$$\frac{dC}{dt} = q_r \frac{dL}{dt} \quad (17)$$

where C is carbohydrate content (% on wood) and q_i , q_b and q_r are proportionality constants in the initial, bulk, and residual phases (See Table 6.).

The theoretical model presented by Gustafson *et al.* (1983) was further developed by Pu *et al.* (1991), especially to handle the alkali consumption and carbohydrate degradation more accurately. Industrial loblolly pine chips were used. Pu *et al.* (1991) presented separate models for cellulose and hemicellulose degradation, after analysing the results for red spruce by Genco *et al.* (1989). Rate equations for initial phase carbohydrate degradation are (Pu *et al.* 1991)

Table 6. Proportionality constants in Eqs. 15–17.

Phase	Parameter	Value ^(*)	Value ^(**)
Initial phase	q_i	2.53	2.12
Bulk phase	q_b	0.47	0.665
Residual phase	q_r	2.19	0.55

(*) Gustafson *et al.* (1983)

(**) Vanchinathan & Krishnagopalan (1997)

$$\frac{dC}{dt} = -e^{(4.50-4209.8/T)}[OH^-]^{1.5}(C - Cr)^{1.5} \quad (18)$$

$$\frac{dH}{dt} = -e^{(6.02-4209.8/T)}[OH^-]^{1.5}(H - Hr)^{1.5}. \quad (19)$$

Rate equations for bulk phase carbohydrate degradation are (Pu *et al.* 1991)

$$\frac{dC}{dt} = -e^{(36.08-18041.9/T)}[OH^-](C - Cr) \quad (20)$$

$$\frac{dH}{dt} = -e^{(21.42-11186.0/T)}[OH^-](H - Hr) \quad (21)$$

where C and H are cellulose and hemicellulose contents (% on wood), Cr and Hr are unreactive cellulose and hemicellulose contents (% on wood). Based on data by Genco *et al.* (1989), Cr and Hr were approximated to be 32% and 5% on wood (Pu *et al.* 1991).

3.2.3 Alkali kinetics

According to Vanchinathan & Krishnagopalan (1997), the effective alkali consumption in the initial phase as a function of lignin is

$$-\frac{d[OH]}{dt} = b_{i,OH}L \quad (22)$$

where $b_{i,OH}$ is a constant relating effective alkali concentration to lignin ($\frac{mol}{dm^3}$). The irreversible consumption of effective alkali in bulk and residual phases is

$$\frac{dEA}{dt} = \left[b_{b,1,OH} \frac{dL}{dt} + b_{b,2,OH} \left(\frac{dC}{dt} + \frac{dH}{dt} \right) \right] / (0.1 \cdot LW) \quad (23)$$

where EA is effective alkali concentration of pulping liquor without considering the wood-alkali equilibrium ($\frac{mol}{dm^3}$) and LW is liquor-to-wood ratio ($\frac{dm^3}{kg}$). $b_{b,1,OH}$ and $b_{b,2,OH}$ are constants relating effective alkali concentration to lignin ($\frac{mol}{dm^3}$) (Pu *et al.* 1991); See Table 7.

Table 7. Constants relating effective alkali concentration to lignin in Eqs. 22 and 23.

Phase	Parameter	Value ^(*)	Value ^(**)
Initial phase	$b_{i,OH}$	$7.5 \cdot 10^{-4}$	-
Bulk phase	$b_{b,1,OH}$	$5 \cdot 10^{-3}$	$2.1 \cdot 10^{-3}$
	$b_{b,2,OH}$	$1.2 \cdot 10^{-2}$	$6.1 \cdot 10^{-3}$

(*) Vanchinathan & Krishnagopalan (1997)

(**) Pu *et al.* (1991) from data by Genco *et al.* (1989)

3.3 Chip column's packing degree

The chips form an elastic and compressible column in the digester. The flexibility of the column affects the void space in it. The force acting on the points of contact between chips is transferred via a chip to the next point of contact. Because the number and size of these points vary, it is not possible to define the force exactly. Therefore, a pressure known as 'chip pressure' is defined instead of force. The chip pressure is the average of these contact forces over a reference surface. Chip pressure defines the volume fractions of the chips and liquor in the digester. Chip pressure is not the same as liquor pressure and can deviate greatly from liquor pressure in magnitude. (Härkönen 1987)

Härkönen (1984) has applied the Ergun equation (Ergun 1952) to describe the liquor flow resistance of a compressed chip column as

$$\frac{\partial p_l}{\partial h} = R_1 \left[\frac{\epsilon_c^2}{(1 - \epsilon_c)^3} \right] v + R_2 \left[\frac{\epsilon_c}{(1 - \epsilon_c)^3} \right] v^2 \quad (24)$$

where p_l is liquor pressure (Pa), ∂h is corresponding change in the position coordinate (m), ϵ_c is the volume fraction of chips and v is superficial velocity in packed bed, *i.e.* the volumetric flow rate of free liquor divided by the cross-sectional area of the empty digester.

Equation 24 considers laminar and turbulent flows. Härkönen (1987) determined values for constants R_1 and R_2 through experiments. The constants, R_1 and R_2 , depend on species, chip particle size and size distribution as the examples in Table 8 show. It has to be noted, that in the case of Lammi (1996) a simplified equation for flow resistance has been used.

Uncooked chip columns are compressible, because individual chips bend and conform under stress. The compressibility increases with reduced lignin content during the cooking. Chips in the cooking reactor experience compressive forces such as static head and fluid drag. The chip volume fraction, ϵ_c , depends on residual lignin content (Kappa number) and compacting pressure. (Gullichsen 2000a)

Table 8. Some examples of chip column compressibility constants, Eq. 24.

Wood species	R_1	R_2
Scandinavian Pine (Härkönen 1987)	4600	$3.9 \cdot 10^6$
Scandinavian Birch (Lammi 1996)	-17	$1.7 \cdot 10^6$
Eucalyptus Camaldulensis (Lammi 1996)	3.2	$3.7 \cdot 10^6$

Table 9. Some examples of chips' void fraction constants, Eq. 25.

Wood species	k_0	k_1	k_2	k_3
Scandinavian Pine (Härkönen 1987)	0.644	0.59	0.139	0.831
Scandinavian Birch (Lammi 1996)	0.630	0.64	0.151	0.697
Eucalyptus Camaldulensis (Lammi 1996)	0.591	0.56	0.148	0.675

According to Härkönen (1987), the void fraction of the chips in chip column changed as

$$\epsilon_c = k_0 - \left(\frac{P_{chips}}{10^4} \right)^{k_1} (k_2 \ln K - k_3) \quad (25)$$

where the chip pressure, P_{chips} (Pa), is the contact force acting on the chip column divided by the cross-sectional area of the chip column, K is Kappa number, k_0 , k_1 , k_2 and k_3 are species specific constants determined empirically. Different values for k parameters have been presented in the literature, see Table 9.

The chip pressure is a factor, which mainly defines the movement of the chip column in the digester. Chip pressure can be calculated using the force balance on the chip column. The acting forces are gravity, buoyancy, friction between the digester wall and the chip column, and the flow's friction between the chip column and the liquor. Saltin (1992) has calculated the chip pressure using a force balance on the chip column. This force balance is

$$\frac{\partial P_{chips}}{\partial h} = (\rho_c - \rho_l)\epsilon_c g - \mu \frac{P_{chips}}{D_{digester}} \pm \frac{\partial p_l}{\partial h} \quad (26)$$

where ρ_c is chip density ($\frac{kg}{m^3}$), ρ_l is liquor density ($\frac{kg}{m^3}$), g is gravity coefficient, μ is friction coefficient (0.5 between wood and steel (Saltin 1992)) and $D_{digester}$ is digester's diameter (m). The sign " \pm " in the flow friction term depends on the directions and relative values of flows of chips and liquor inside the digester. For example in the counter-current washing zone the sign is " $-$ ".

Usually in the industrial control systems, the residence time of chips is calculated based on production rate, digester geometry and given constant compaction profile. In this study, the residence time of chips in the digester is calculated utilising also the chips' volume fraction, see Section 6.1.

4 Overview of kraft pulping control

In this chapter, an overview on the control of continuous kraft pulping is made, and the state-of-the-art of Kappa number control is discussed.

4.1 Control of continuous pulping process

The first unit operations in continuous pulping, *i.e.* pre-steaming and impregnation, are described in Chapter 2. The main features of conventional and Downflow Lo-SolidsTM processes are presented in Section 2.2. In this section, an overview of pulping control is given.

Important variables of kraft cooking are listed in Section 2.1. Main challenges in digester control are (Leiviskä 2000, Sutinen *et al.* 1990):

1. Variations in chip quality. See Section 2.3.1.
2. Measurement problems. A serious problem is that the controllable quality variable, Kappa number, is not directly measurable while the cooking occurs.
3. Long process delays.

Production rate is controlled by adjusting the chip meter speed (or chip screw rpm) according to the production rate target. The production rate also depends on the pocket filling degree of the chip meter, bulk density of the chips, and the yield. The residence time of chips is mainly affected by production rate and the chip column's compaction; see Section 3.3. Compaction is adjusted by chip and liquor levels, extraction and blow-line flows, and by blow-line consistency. To ensure as steady conditions in the digester as possible, the most feasible strategy is to keep the blow-line's flow rate and consistency constant. Pulp quality (Kappa number, yield, strength, viscosity) is mainly controlled by temperature and chemical profiles. A high internal pressure is required in the process to eliminate flashing and boiling of the liquid within the different temperature zones (Smook 2002). In the chemical profile control, the aim is to ensure the correct alkali-to-wood ratio based on an alkali concentration of white liquor and the amount of dry wood. The liquor-to-wood ratio is controlled by the black liquor. Pulp is washed in counter-current washing zone(s), which also has an important role in the resulting pulp quality.

4.2 State-of-the-art of Kappa number control

Some of the earliest surveys of cooking control strategies and models are presented by Perron & Ramaz (1977) and Jutila (1979). During the 1970's the first automatic control systems were introduced to the cooking control (Perron & Ramaz 1977). A typical control strategy usually included, as it does also nowadays, the following parts: chip level control, chemical-to-wood ratio control, liquor-to-wood ratio control and temperature control.

Final quality control (Kappa number control) has been used to compensate disturbances, particularly wood quality variations. In the Kappa number control, a set point for temperature or chemical charge has been calculated. Improvements to the feedback control were achieved, when alkali concentration measurements during the cooking were used in the control. The Kappa number control based on cooking liquor measurements is considered in Section 4.2.3.

Automation systems have been developed and computing capacity has been substantially increased during the last decades, especially during the last few years. This has enabled the implementation of advanced control methods into the industrial automation systems. Purdue model (Smith & Williams (1974, 1975a,b), see Section 3.1) is found to be an industrially accepted pulping model. However, nowadays the models presented by Vroom (1957) (See Section 4.2.1.) and Hatton (1973) (See Section 4.2.2.) are still widely used in the cooking control.

4.2.1 Vroom's H -factor model

Vroom (1957) established H -factor, a model combining cooking time and temperature as a single variable. Model development was based on work by Vroom and some other researchers. The aim of the model development was to determine a factor, which satisfactorily describes the temperature dependence of reaction rates, but not any absolute reaction rate. Thus, changing the temperature from one level to another would change the pulping rate by a given factor, regardless of the absolute velocity of the reaction at that time.

Vroom (1957)'s idea was that if a single numerical value for times and temperatures of any cooking cycle could be established, then under otherwise constant pulping conditions, all cycles having that same numerical value would produce pulps of equivalent yield - regardless of differences in the rate of rise to temperature or in the level of maximum temperature. However, it has been reported that an improved yield can be achieved at any given Kappa number when the cooking temperature is decreased (Dillner 1993, Bäckström *et al.* 1996).

The reaction rate at 100°C was chosen as unity and rates at all other temperatures relate to this standard. The Arrhenius equation was used in form

$$\ln k = B - \frac{A}{T} \quad (27)$$

where k is reaction rate, T is temperature (°C), B and A are constants.

A value of 16113 for A , based on work reported, was selected. The value for constant B can now be solved and the relative rate at any other temperature

$$k = e^{43.20 - \frac{16113}{T}}. \quad (28)$$

Tables of reaction rate values for any desired temperature range were calculated. Employing those relative reaction rate values, a curve of rate vs. time in hours can be plotted for any cooking cycle, and the area under such a curve is then designated as the H -factor. H -factor does not represent any absolute prediction of the yield to be obtained with a given set of pulping conditions. It's major use is visualized as a means of predicting the required change in cooking time to compensate for some change in temperature, or *vice versa*. However, once the H -factor vs. yield relationship has been established for any given set of conditions, the factor may be used to estimate the cycle required for any degree of pulping with those conditions; but a change in, *e.g.*, liquor concentration, or wood species, would require the establishment of a new H -factor vs. yield relationship before the factor could be used in this manner under the new conditions. (Vroom 1957)

H -factor has been applied in the modelling of reaction kinetics *e.g.* by Kerr (1970), Kerr & Uprichard (1976) and Hatton (1975). Kerr developed a model, which enables the Kappa number to be predicted from the wood lignin content, the initial effective alkali level applied, and the H -factor.

If the effective alkali concentration and the lignin content at the beginning of the bulk delignification are known, a unique relationship exists for each alkali concentration between the lignin content at any stage of the bulk delignification and the H -factor (Vroom 1957). (Olm & Tistad 1979)

4.2.2 Hatton's Kappa number model

Hatton (1973, 1975, 1976) succeeded in correlating the kraft pulping data with simple relationships that relate pulp yield and Kappa number to H -factor and effective alkali charge. These equations are applicable to the pulping of thin chips at unspecified and presumably constant values of sulfidity and liquor-to-wood ratio. (Clayton *et al.* 1989)

The form of the relationship is

$$Y = \alpha - \beta \cdot (\log_{10} H) \cdot (EA^{n_1}) \quad (29)$$

$$K = A - B \cdot (\log_{10} H) \cdot (EA^{n_2}) \quad (30)$$

where Y is yield, K is Kappa number, H is H -factor, EA is effective alkali charge and A , B , α , β , n_1 , and n_2 are parameters assumed to be constant for a given species. Values of the parameters were determined for several species (Hatton 1973, 1976). It was found that the softwoods all behave similarly with regard to alkali sensitivity (n), while the hardwoods exhibited a much wider range of n values.

This is presumably a result of the greater diversity of the hardwoods, with respect to both chemical composition and wood morphology.

Hatton's equations provide a compact description of the pulping behaviour of several species. Their most obvious shortcomings are that they cannot predict the effects of changing sulfidity or liquor-to-wood ratio, and that their predictive power for hardwoods is not very good. Nevertheless, they are useful for quickly estimating the Kappa number and yield of a given cook or for calculating tradeoffs between chemical charge and H -factor. (Clayton *et al.* 1989)

Despite some weaknesses mentioned, Vroom's H -factor and Hatton's Kappa number model are still widely used in the industry. The basic strategy for Kappa number control is shown in Fig. 5. In the strategy, the difference between the estimated Kappa number and the Kappa number target is used to correct the H -factor target and the temperature profile. The Kappa number model gives estimates of the cooking result inside the cooking zone that are faster than using conventional control with laboratory Kappa number measurements. Temporary variations in the cooking phase are observed and compensated for before the pulp reaches the washing zone. The model is updated with blow-line or laboratory Kappa numbers. Statistical methods filter variations in laboratory Kappa number analysis caused by analyzing errors or process fluctuations. (Leiviskä 2000)

4.2.3 Control based on cooking liquor measurements

Several techniques to measure the cooking liquor composition exist: conductivity measurements (Vanchinathan & Krishnagopalan 1995), near infrared spectroscopy (NIR) (Andersson 2003, Shen *et al.* 2004), Fourier transform infrared (FTIR) spectrophotometer (Li & Mui 1999), gas-liquid chromatography (Alén *et al.* 1988), ion chromatography (Vanchinathan & Krishnagopalan 1995), refractive index measurement (Vanchinathan & Krishnagopalan 1995), UV-light absorption measurement (Vanchinathan & Krishnagopalan 1997, Saucedo & Krishnagopalan 2002), and titration.

Cooking liquor measurements give indirect information about the extent of the cooking degree. The information about the cooking liquor composition within the process can be utilised in the profiling of cooking chemicals into the process.

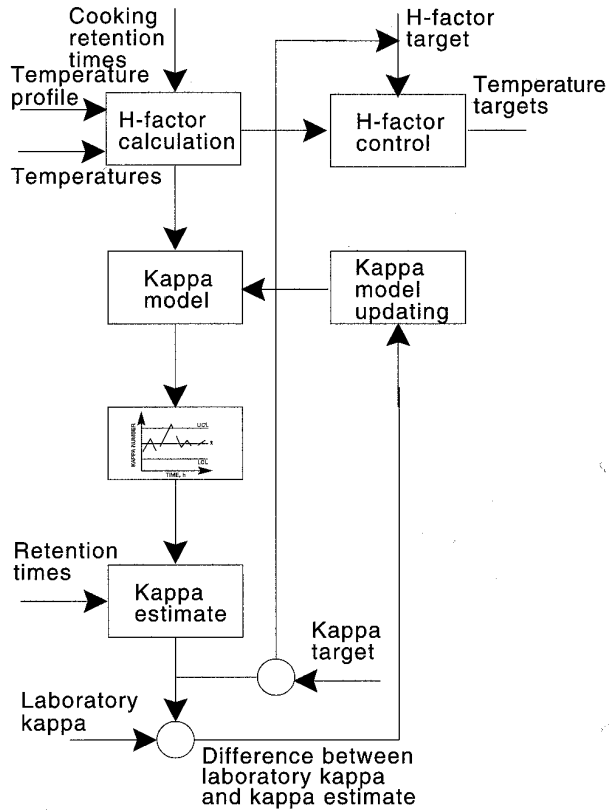


Fig. 5. Basic strategy for Kappa number control (Leiviskä 2000).

5 Development of simulation framework

In this chapter, the implementation of the applied models as a part of the simulation model is described. In addition, the modelling procedures of the real-time and prediction models are presented. Finally, new Kappa number control strategy is introduced.

Yields of lignin and carbohydrates of wood chips were modelled in the studied processes. The yields are the amounts (% on wood) of the lignin, cellulose and hemicellulose dissolved in each calculation zone of the digester. The yields should be understood as absolute amounts of each wood component, not as a percentage of *e.g.* lignin content of chips, at the calculation moment. Let us assume that a lignin content of the chips when entering a calculation zone is 20% in wood. Now, if the yield is 3% in wood, the lignin content after the zone will be 17% in wood. The total yield (%) is the sum of all wood components left in the chips or pulp, compared with uncooked chips.

5.1 Utilisation of Gustafson's Kappa number model

Several kinetic models, including the two principle model families, Purdue and 3-stage (Gustafson) model, were compared in Andersson *et al.* (2002). When the original model parameters were used in the conventional cooking scheme, only slight differences within, and across the families were found. After validating the re-regressed models against data from ITCTM (isothermal cooking) scheme, the models' ability to predict outside the conditions of the parameter fitting was examined. Gustafson's model was found to capture the end point for lignin, and the Purdue model was found to follow the trajectory better for the majority of the cook.

The model by Gustafson *et al.* (1983) was chosen to be applied in this study, because the kinetics of delignification in that model are widely accepted to be the most reliable. The model also has other important features, like the possibility to model diffusion kinetics, yield, viscosity of pulp, and the effects of chip size distribution on pulping uniformity and rejects. However, difficulties were met when industrial cooking processes were modelled. Problems mainly occurred in the cooking

of hardwood, but also in softwood cooking in the case of Downflow Lo-SolidsTM process. The main observation was that the model with the original parameters was too sensitive to temperature changes. It was also found, that there existed unmeasurable variables and disturbances in process conditions and raw material needed to be taken into account on-line. Some problems also occurred because of non-ideal process conditions during the grade transitions.

To solve the problems mentioned above, the model parameters were optimised, updated on-line, and smoothly adjusted over grade transitions as will be described in the following sections.

5.1.1 Parameter optimisations of Arrhenius equation

The model was noticed to be too sensitive to temperature changes when using the original model parameters (See Table 5 in p. 43.). This was seen especially in hardwood but also in softwood cooking. This can be seen *e.g.* from the modelling errors between predicted and measured blow-line Kappa numbers of HW cooking presented in Table 22 in p. 117. With optimised HW parameters (Fig. 60 in p. 112) the modelling errors are significantly smaller than with the original parameters (Fig. 61 in p. 113).

The bulk phase rate equation (Eq. 11 in p. 43) was chosen to be optimised, because most of the delignification takes place in that phase. The original assumption was that almost same amount of lignin should be dissolved with both optimised and original parameters under the same, typical process conditions. Later this assumption was partly rejected and the amount of delignification is corrected with a yield factor, see Section 5.1.2. The model's temperature dependence can be affected by changing parameters B_1 and/or B_2 in the Arrhenius part of the rate equation. In order to keep the lignin yield at the same level, also parameters A_1 and/or A_2 have to be changed. Various combinations of parameters were tested and lignin yield was calculated with different temperatures in otherwise constant process conditions. The optimisation was based on the evaluation of the deviation between the modelled and measured blow-line Kappa numbers. The modelling results were analysed both qualitatively (visual observation) and quantitatively (ME and RMSE). The data from several production periods during different seasons was used in the optimisation of the parameters. The production periods occurred earlier than the example periods shown in this thesis.

The modified parameters are shown in Table 10. With the original softwood parameters, 1 K change of temperature produced about a 9% change in the lignin yield of both softwood and hardwood (slightly dependent on process conditions). With modified hardwood parameters, 1 K change of temperature produces only about a 3% change in the lignin yield in the case of conventional process. With the optimised parameters of Downflow Lo-SolidsTM cooking, 1 K change of temperature affects the lignin yield about 4.7% in softwood and about 2% in hardwood cooking.

The original parameters in the equations of initial and residual phase delignification were used. In Downflow Lo-SolidsTM hardwood cooking, and in the Kappa

Table 10. Original (Gullichsen 2000a) and optimised parameters of Arrhenius equations in bulk phase rate equation Eq. 11.

Parameter	Original value, SW	Optimised value, SW	Optimised value, HW
Conventional process			
A_1	35.5	-	28
B_1	17200	-	16800
A_2	29.4	-	6.39
B_2	14400	-	4575
Downflow Lo-Solids process			
A_1	35.5	29	28
B_1	17200	17000	16800
A_2	29.4	15.9	4.7
B_2	14400	8500	3800

number prediction of conventional cooking, the model's sensitivity to the changes in process conditions was also reduced by restricting the deviation between the modelled and target Kappa numbers. A certain relative difference between the modelled and target Kappa numbers was allowed.

The reaction rate of spruce is greater than the rate of pine (Kleinert 1966); (Wilder & Daleski 1965) and that difference could be taken into account by optimising the reaction rate parameters in the model (Gustafson *et al.* 1983). In the studied cases, the softwood mainly consists of pine and the original model parameters were not changed because of a minor portion of the spruce.

5.1.2 On-line updating

The original reaction rate parameters k_x (See Table 4 in p. 42.) in Eqs. 10, 11 and 14 were experimentally optimised (See Section 5.1.1.), and updated on-line so that the lignin yields approximately obeyed both the values reported in the literature and the measured blow-line Kappa numbers. Parameters k_x were multiplied with a 'yield factor', *Y factor*, which was updated on-line after each Kappa number measurement during normal production (*i.e.* excluding periods of grade changes and major process disturbances, and situations when earlier Kappa number measurements are not available from a reasonable time span). The procedure of the parameters updating is illustrated in Figs. 13 and 14, in Section 5.3, in which the structure of the simulation model is presented. By updating the yield factor on-line, long period changes in chip size distribution, in air removal and penetration of the chips, or changes in other process conditions can be taken into account. Yield factor is a function of grade, and the deviation between the mean values of modelled and measured blow-line Kappa numbers. The deviation was calculated from

the previous 80 minutes in conventional cooking, and residence time of zone D_3 in Downflow Lo-SolidsTM cooking. The present yield factor was changed by the product of Kappa number deviation and $Y_{correction}$. The yield factor was increased if the modelled Kappa numbers were higher than measured, and *vice versa*. Values for $Y_{correction}$ were empirically determined to minimise the deviation between measured and modelled blow-line Kappa numbers. In the case of conventional cooking, values of 0.0045 in SW, and 0.035 in HW were used for $Y_{correction}$. In the case of Downflow Lo-SolidsTM cooking, following values for $Y_{correction}$ were used: 0.0007 and 0.0035 (when D_3 was modelled in one or four parts) in SW, and 0.002 and 0.009 in HW, respectively. Examples of resulting yield factors are presented in Figs. 6 and 7.

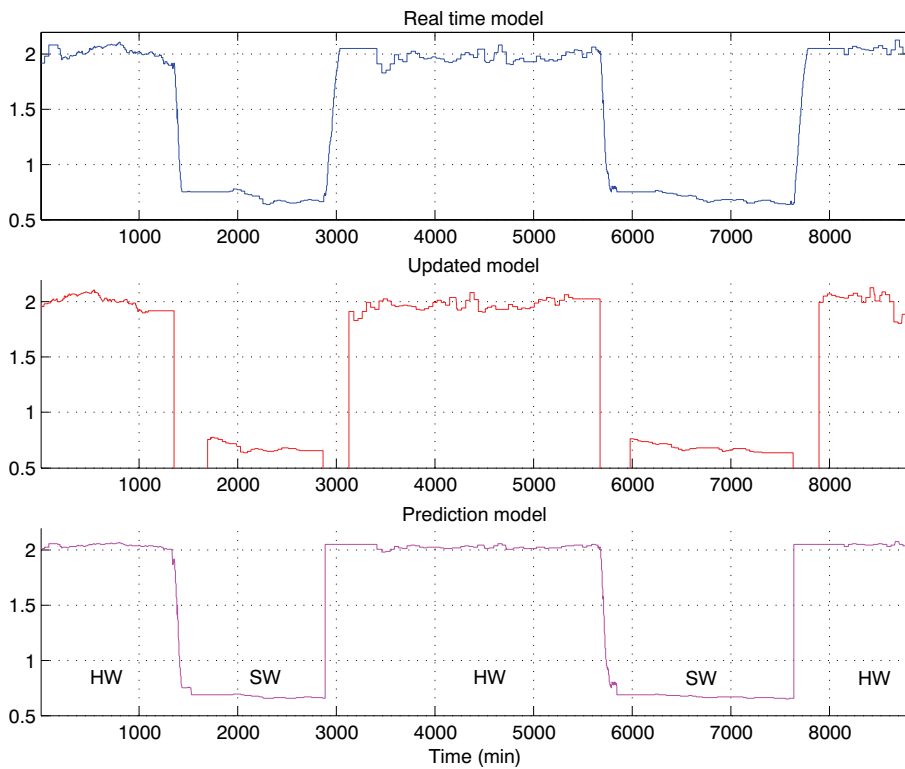


Fig. 6. Yield factors in conventional process.

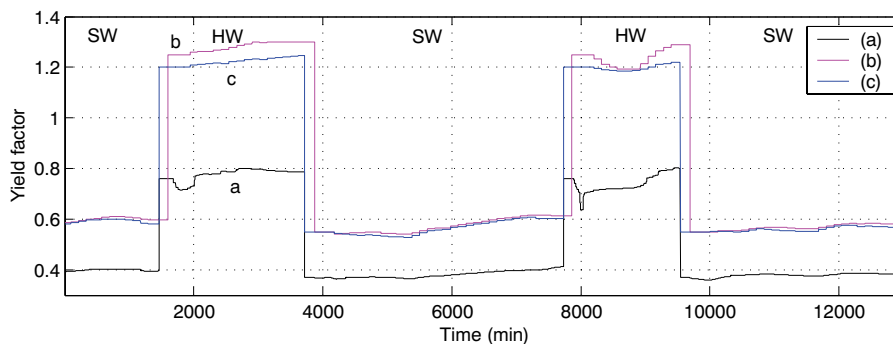


Fig. 7. Yield factors in zone D_3 in Downflow Lo-SolidsTM process; (a) calculation in one part, (b) calculation in four parts, (c) prediction model, calculation in four parts.

5.1.3 Modelling during grade transitions

Compared to the cooking of hardwood, higher temperatures and lower alkali concentrations are used in the softwood cooking. Thus, the process conditions are not ideal either for softwood or hardwood cooking during the grade transitions. The usual strategy is that adequate heating for softwood cooking is ensured. The grade transition may last from 100 to 300 minutes, and during that time improper pulp quality is achieved. In a real process, the physical boundary between different grades is relatively clear, and during the long residence time the temperatures and alkali concentrations become even. As a result, the difference between the measured and target blow-line Kappa numbers is not so large. If a clear boundary between the grades is assumed and parameters of normal production for both grades are used, a great difference between the modelled and measured Kappa numbers occurs.

The difference between the modelled and measured Kappa numbers was eliminated, especially when visualising the results for the operators, by two ways. First, the model parameters (Tables 4 in p. 42 and 10 in p. 55) were smoothly adjusted during the transition period as a function of the temperature change in each of the digester's zones (D_1 – D_4 in Fig. 1 and D_1 – D_3 in Fig. 2). The adjustment of the parameters in each zone was started when a new grade entered the zone and was done within the zone's temperature changing time. The parameters were restricted to between the modified values of softwood and hardwood. Similar approach has been used by Funkquist (1997) in order to get a smooth transition in reaction rate and consumption of alkali between the bulk and residual stages of the delignification. In Funkquist (1997), the Arrhenius factor and the stoichiometry coefficient have been changed linearly within an interval of lignin content in bulk and residual stages. The effect of parameter adjustment on the lignin yield in zone D_1 in the conventional process, during grade transition periods, is illustrated in Fig. 8.

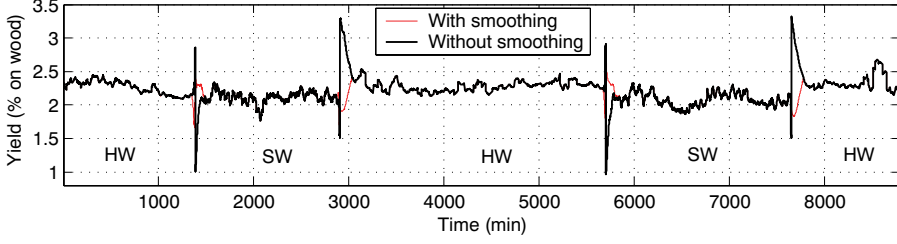


Fig. 8. Lignin yield in zone D_1 with and without smooth adjustment of model parameters (conventional process).

In Fig. 9, is illustrated how yield factor was smoothly adjusted as a function of temperature change over a grade transition period in each of the digesters's zones (D_1 – D_4 in Fig. 1). Temperature profiles in zones D_1 – D_4 during the grade transition are shown in Fig. 10. In Fig. 6, the yield factors over a longer production period of conventional cooking are presented. The value of adjusted yield factor during grade transition period was calculated as

$$Y factor_{gch} = Y factor_{old} \cdot (1 - factorchange) + Y factor_{new} \cdot factorchange \quad (31)$$

where $Y factor_{gch}$ is a yield factor during grade transition period, $Y factor_{old}$ is a yield factor of old grade, $Y factor_{new}$ is a yield factor of new grade and $factorchange$ determines the proportional amount of temperature change and is calculated as

$$factorchange = \frac{T_{Dx} - T_{Dx_{oldmean}}}{T_{Dx_{newmean}} - T_{Dx_{oldmean}}} \quad (32)$$

where T_{Dx} is present temperature in zone Dx, $T_{Dx_{oldmean}}$ is mean temperature of old grade and $T_{Dx_{newmean}}$ is mean temperature of new grade in zone D_x . $factorchange$ was restricted to stay between 0 and 1.

Another way to eliminate the Kappa number difference during grade transitions was the usage of the mean values of lignin yields instead of the calculated ones. Mean value of old grade was used until the new species achieved the calculation point. After that the mean value of new grade was used. The mean values of lignin yields in each zone were exploited during the temperature-changing time.

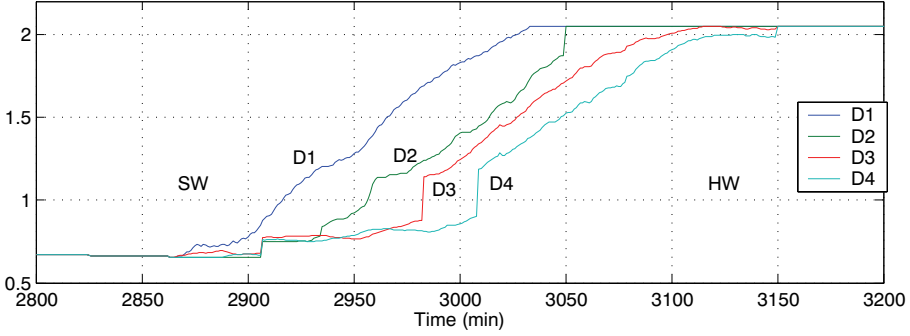


Fig. 9. Adjustment of yield factors during grade transition period (conventional process).

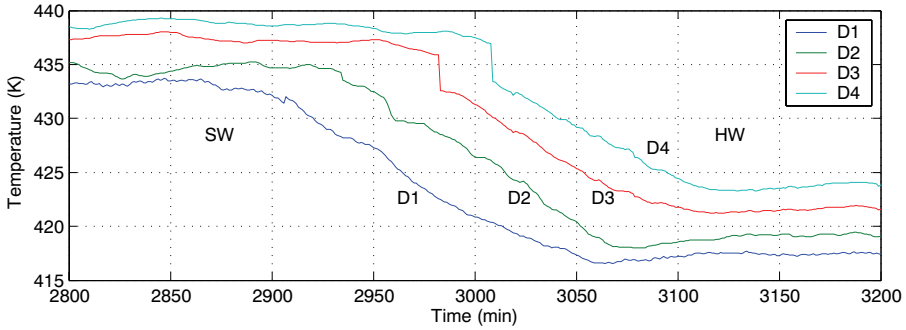


Fig. 10. Temperatures in zones D_1 – D_4 (Fig. 1) during grade transition period (conventional process).

5.1.4 Discussion

The parameters of the pulping model presented by Gustafson *et al.* (1983) are species specific. In Tables 4 and 10, the original softwood (Scandinavian Pine) parameters (k_x , A , B) (Gullichsen 2000a) are presented. By optimising the parameters, both the modelled amount of lignin dissolved and the temperature dependence of delignification model, were affected.

The modelled amount of dissolved lignin was affected by the on-line updating of the reaction rate parameters k by multiplying them with a yield factor. k parameters in all zones in the bulk phase were multiplied with the same yield factor. A yield factor was used in both conventional and Downflow Lo-SolidsTM cooking. Examples of yield factors in conventional cooking are presented in Fig. 6 and in Downflow Lo-SolidsTM cooking in Fig. 7.

When the yield factor has value 1, the original model parameters k_x are used. It can be seen, that in the case of hardwood cooking the updated k_x parameters are bigger than original parameters, and in the case of softwood cooking the updated k_x parameters are smaller than original parameters. The parameters behave the same way in both conventional and Downflow Lo-SolidsTM cooking. In the case of softwood cooking, this means that either the reaction rate dependence on temperature and/or alkali profile differ from the original case, or that the temperature and/or alkali profiles in the chips' scale differ from the used profiles. One reason most probably is the content of dissolved solids in the cooking liquor. This might be the explanation for dissimilar parameters in Downflow Lo-SolidsTM cooking compared to conventional cooking. The construction of temperature and alkali profiles are presented and discussed in Section 6.2. Temperature and alkali profiles were constructed to describe the chip scale conditions. However, these profiles were not verified by any measurement. In the case of hardwood cooking, one explanation for the need of yield factor is the different properties of hardwood compared with softwood chips.

The effect of the height of a calculation zone on the modelling results can be evaluated by analysing the data in Table 11. Zone D_3 of conventional cooking was modelled in 1, 5, 10 and 20 parts, of which heights were 11, 2.2, 1.1 and 0.55 m. The process conditions were normal, and the mean temperature was determined to be equal in each case. A yield factor, *Y factor*, was adjusted so that lignin content of chips decreased from 16.00 to about 8.56 % in wood in each case. Alkali consumption was slightly higher, and carbohydrates degradation was slightly lower, when the height of an individual calculation zone was reduced. While reducing the height of an individual calculation zone from 11 to 0.55 m, the value of yield factor had to be increased from 0.65 to 0.875. Thus, the adjusted reaction rate parameters k_x were closer the original values, when smaller calculation volumes were used. The use of smaller calculation volumes would be beneficial, especially, during grade change situations, or when process conditions are significantly changed.

The heights of the calculation zones D_1 – D_4 in the studied conventional process were not equal. Based on the above examination, the heights of the calculation zones should be equal if a same yield factor is used in all zones. Now, when zone D_3 was higher than other zones, too much lignin was dissolved in that zone compared to other zones. It can be assumed that the accuracy of the modelling results would improve with equal zones' heights. This assumption can be justified with the more stable process conditions in the upper part of the digester, as can be later seen *e.g.* of the predicted temperature profiles (See Section 6.2.1.).

The parameters A and B in the Arrhenius equation were optimised to decrease the temperature dependence of delignification. These parameters were not optimised to affect the amount of delignification. Thus, the updating of k parameters was required to ensure correct amount of delignification. If the delignification is not correctly modelled in Kappa number prediction and control, unrealistic temperature changes would be required. The model parameters were optimised based on data from earlier production periods than the ones presented in this thesis. The validity of the models might be improved by optimising the parameters with recent data.

Table 11. Effect of the height of a calculation zone on modelling results; as an example zone D_3 in conventional SW cooking.

D_x	t_x	T	$[OH^-]$	$[HS^-]$	L	C	H
Number of zones = 1 (Height of 1 zone = 11 m, $Y_{factor} = 0.650$)							
0					16.00	41.50	9.50
1	50	436.50	0.500	0.100	8.55	39.26	6.65
Number of zones = 5 (Height of 1 zone = 2.2 m, $Y_{factor} = 0.820$)							
0					16.00	41.50	9.50
1	10	435.90	0.593	0.100	14.03	41.00	8.85
2	10	436.20	0.540	0.100	12.35	40.55	8.33
3	10	436.50	0.495	0.100	10.90	40.16	7.91
4	10	436.80	0.456	0.100	9.66	39.80	7.57
5	10	437.10	0.423	0.100	8.57	39.47	7.29
mean		436.50	0.501				
Number of zones = 10 (Height of 1 zone = 1.1 m, $Y_{factor} = 0.855$)							
0					16.00	41.50	9.50
1	5	435.60	0.592	0.100	15.00	41.26	9.18
2	5	435.80	0.565	0.100	14.06	41.03	8.89
...							
5	5	436.40	0.497	0.100	11.64	40.40	8.19
6	5	436.60	0.477	0.100	10.94	40.21	7.99
...							
9	5	437.20	0.426	0.100	9.10	39.68	7.51
10	5	437.40	0.411	0.100	8.57	39.52	7.37
mean		436.50	0.493				
Number of zones = 20 (Height of 1 zone = 0.55 m, $Y_{factor} = 0.875$)							
0					16.00	41.50	9.50
1	2.5	435.55	0.592	0.100	15.49	41.38	9.34
2	2.5	436.65	0.578	0.100	15.00	41.26	9.19
...							
10	2.5	436.45	0.488	0.100	11.63	40.42	8.22
11	2.5	436.55	0.479	0.100	11.28	40.32	8.13
...							
19	2.5	437.35	0.413	0.100	8.81	39.63	7.47
20	2.5	437.45	0.405	0.100	8.55	39.55	7.41
mean		436.50	0.489				

t_x (min), T (K), $[OH^-]$ ($\frac{mol}{dm^3}$) and $[HS^-]$ ($\frac{mol}{dm^3}$) are mean values in zone;

L (% on wood), C (% on wood) and H (% on wood) are values after zone

Modelling during grade transitions is challenging, because of un-idealistic process conditions for either grades. There were two main aims in the modelling of grade transitions. One aim was to describe the delignification over grade transition and another aim was to visualise the results for the operators. The smooth adjusting

of parameters as a function of temperature change was found to be successful, especially in hardwood-softwood changes. It should be noted, that higher yields in earlier zones result in lower yields in later zones and *vice versa*.

Applicability of the optimised model parameters to other processes depends on the process conditions. Most probably some optimisation is required. The on-line updating of the reaction rate parameters k_x can easily be implemented into other processes. The values of $Y_{correction}$ are required to be determined case by case.

5.2 Identification of residence times

The modelling procedure of residence time in one calculation zone D_x is illustrated in Fig. 11. Residence times of chips in the digester were calculated as a function of the chips' volume fraction (ϵ_c in Eq. 25 in p. 47, optimised parameters are shown in Table 12 in p. 77) and volume flow.

Volume fraction of the chips is a function of the Kappa number (Fig. 38 in p. 91) and chip pressure (P_{chips} , Eqs. 24–26). Superficial velocity in Eq. 24 was calculated as a function of free liquor flow (Fig. 25 in p. 79). Chip and liquor densities in the digester are shown in Fig. 26 in p. 79. Densities were calculated in proportion to total yield. Measured chip and liquor levels in the digester are shown in Fig. 27 in p. 80. The chips' volume flow was calculated as a function of production rate target, density of chips and pulp yield.

5.3 Structure of the simulation model

The simulation model describes both real-time and future behaviour of continuous cooking process. The most important process conditions and cooking degree in the process are modelled. The process conditions include temperature and chemical profiles, and packing degree of the chip column. The packing degree depends, *e.g.*, on the cooking degree, and it is used in the modelling of the residence times of chips.

The structure of the cooking simulation model is described in the cases of conventional and Downflow Lo-SolidsTM cooking in Section 5.3.1. The cooking degree was described with the modelled lignin and carbohydrate contents of chips. The models presented by Gustafson *et al.* (1983) and Pu *et al.* (1991) were used in the modelling of the wood components' yields. Model parameters were optimised (See Section 5.1.1.), are updated on-line (See Section 5.1.2.) and smoothly adjusted over the grade changes (See Section 5.1.3.). Results concerning the real-time modelling of the cooking degree are presented in Sections 6.2.3, 6.2.4, 6.3.3 and 6.3.4. Results of the Kappa number prediction are presented in Section 6.4. A new Kappa number control strategy, based on the prediction models, is presented in Section 5.4 and applied in Section 6.5.

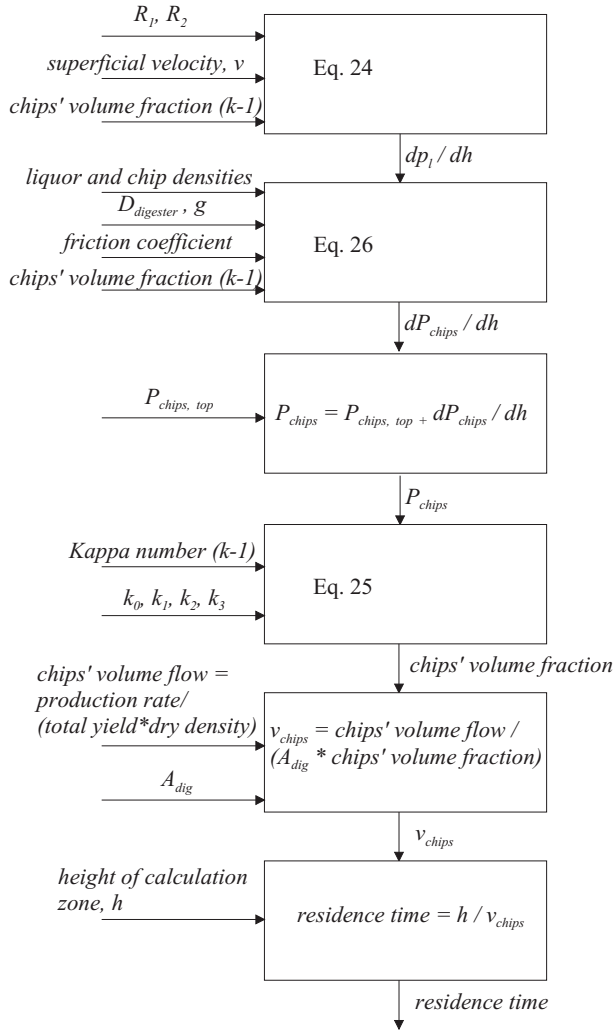


Fig. 11. Modelling of residence time in one calculation zone D_x .

The main process variables in the kraft cooking are grade, chemical concentrations, temperature and cooking time. Alkali concentrations in both processes were modelled using the model presented by Pu *et al.* (1991) (See Sections 6.2.2 and 6.3.2.). Real-time temperature profiles in both processes were based on measurements (See Sections 6.2.1 and 6.3.1.). In conventional cooking, the temperature profile was also predicted using linear models (See Section 6.2.1.). The cooking time in conventional process was modelled as a function of the chip column's packing degree as described in Section 5.2. The packing degree model presented by Härkönen (1987) was used with the parameters optimised by the present author. In

Downflow Lo-SolidsTM cooking, residence times given by equipment supplier were used.

Alkali and temperature profiles of the free liquor were used in the modelling. According to Bhartiya *et al.* (2003), EA concentration inside the chips and in the free liquor is almost the same in low production rate, and the difference is nearly constant in higher production rate. An increase of the blow-line flow in the high production rate case reduces available time for the diffusion of EA and HS^- from the free liquor to the entrapped phase. This leads to a drop in the concentrations of the entrapped phase. However, this drop increases the concentration's driving force, which in turn causes the free liquor concentrations to drop as well (Bhartiya *et al.* 2003). The assumption of the constant alkali concentration and temperature profiles within the chips was also made in Funkquist (1997) and Miyanishi & Shimada (2001). In Miyanishi & Shimada (2001), this assumption was made, because mill chips have a wide, not exactly known range of chip thickness.

The impregnation vessel and digester were divided into calculation zones I_x and D_x (See Figs. 1 and 2.). The modelling procedure in an individual calculation zone is shown in Fig. 12. Modelling of lignin (Kappa number) and carbohydrate yields and alkali consumption are presented in Sections 6.2 and 6.3.

5.3.1 Real-time model with on-line updating

The structure of the real-time simulation model is shown in Figs. 13 (conventional process) and 14 (Downflow Lo-SolidsTM process). The real-time simulation model consists of the following parts: the impregnation vessel's real-time model, digester's real-time model, updating of model parameters, and digester's recalculation model. After each Kappa number measurement during normal production (*i.e.* excluding periods of grade changes and major process disturbances, and situations when earlier Kappa number measurements are not available from a reasonable time span), bulk phase reaction rate parameters k_x are being updated by multiplying them with a yield factor (*Y factor*, see Section 5.1.2). After the updating of the parameters, profiles of the chip column in the cooking and washing zones, are being recalculated with digester's updated recalculation model. Recalculation is being made for zones D_1-D_6 in conventional cooking, and for zones D_3 and D_4 in Downflow Lo-SolidsTM cooking. The updated parameters are being used not only in the real-time simulation model, but also in the prediction and iteration models. Prediction and iteration models are presented in Section 5.3.2, and applied into the Kappa number control in Section 6.5.

Modelling in each calculation zone is being done, once a minute, at the end of the zone. There is one main difference between the conventional and Downflow Lo-SolidsTM processes in the handling of the modelling results between the calculation zones. In the conventional process, results at the calculation moment k are stored to row k in the result array. When these results are later used in the next calculation zone D_x , the values are picked up from an earlier row $k-t_x$ in the array. In the Downflow Lo-SolidsTM process, results at a calculation moment k are stored to

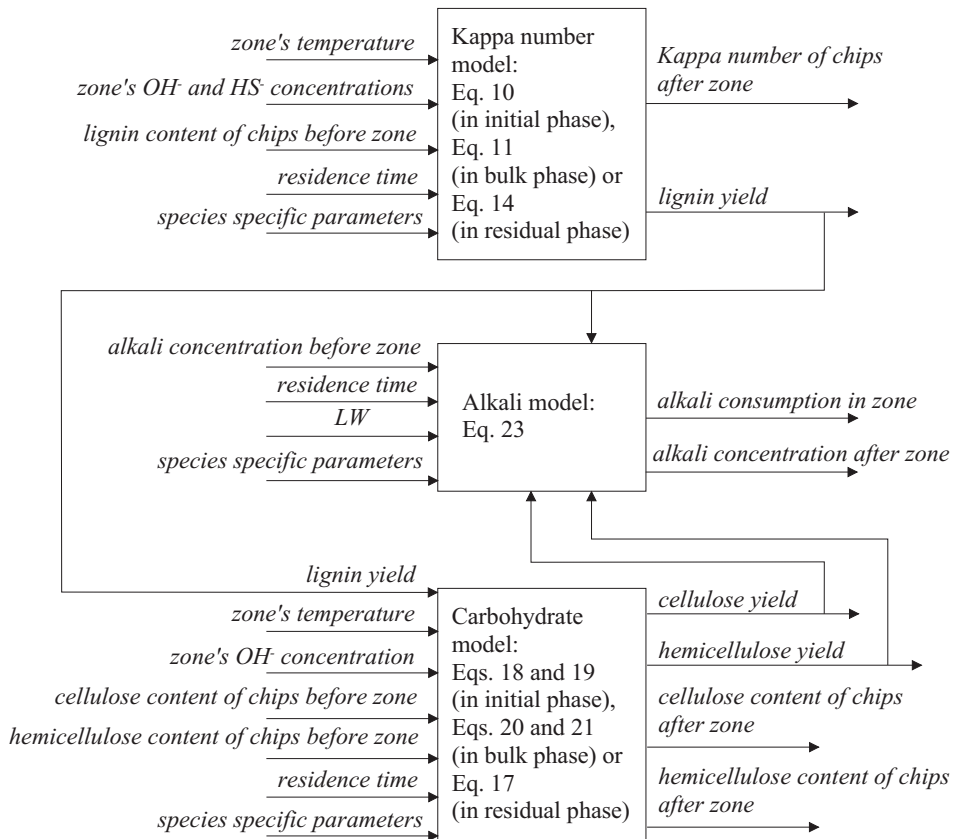


Fig. 12. Modelling of lignin and carbohydrates yields and alkali consumption in one calculation zone I_x or D_x .

rows $k+t_{x+1}:k+t_{x+1} + 100$. Thus, the previous results are ready to be used at a calculation moment k in each calculation zone. The extra 100 rows are filled to ensure that no empty rows exist in the result array, while the production rate is decreased.

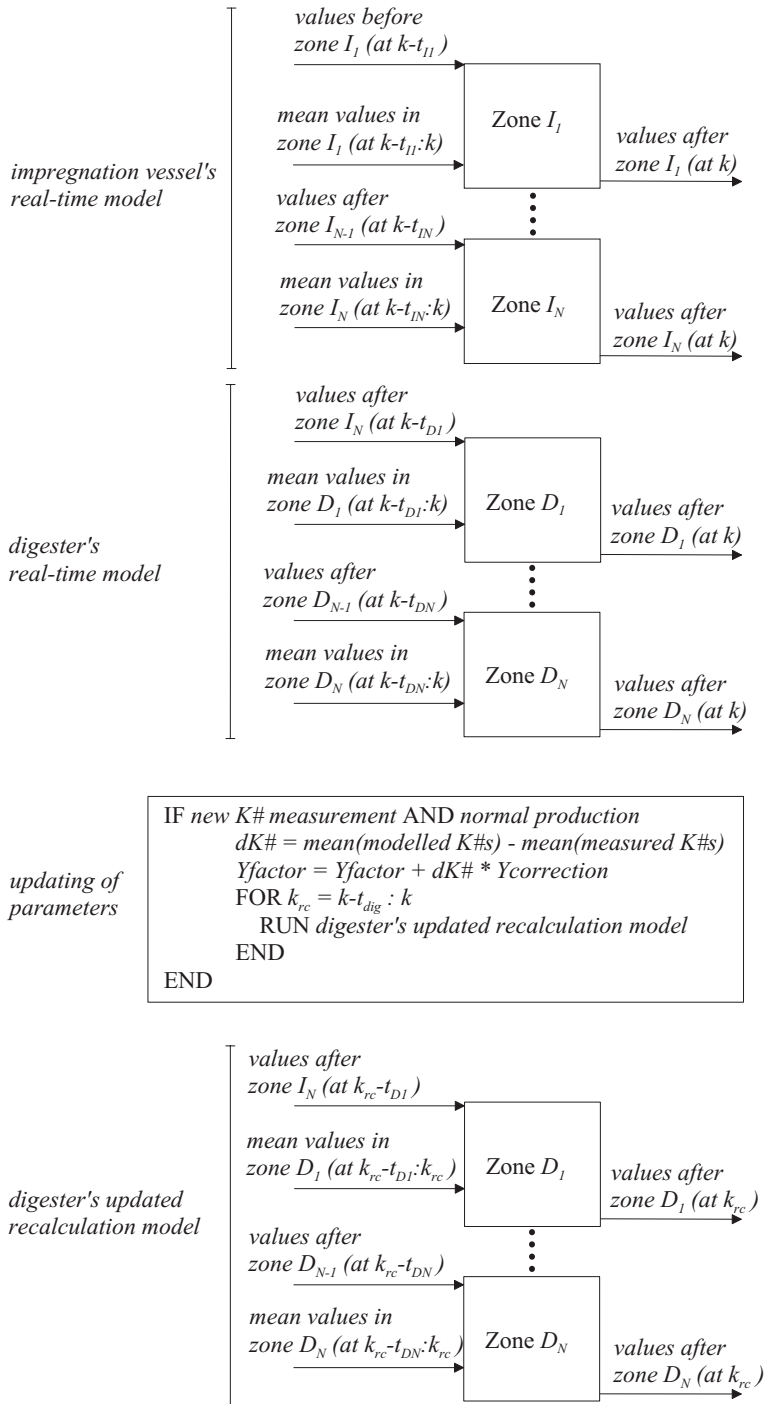


Fig. 13. Structure of the real-time simulation model with recalculation in conventional cooking; detailed structure of one calculation zone I_x or D_x is shown in Fig. 12.

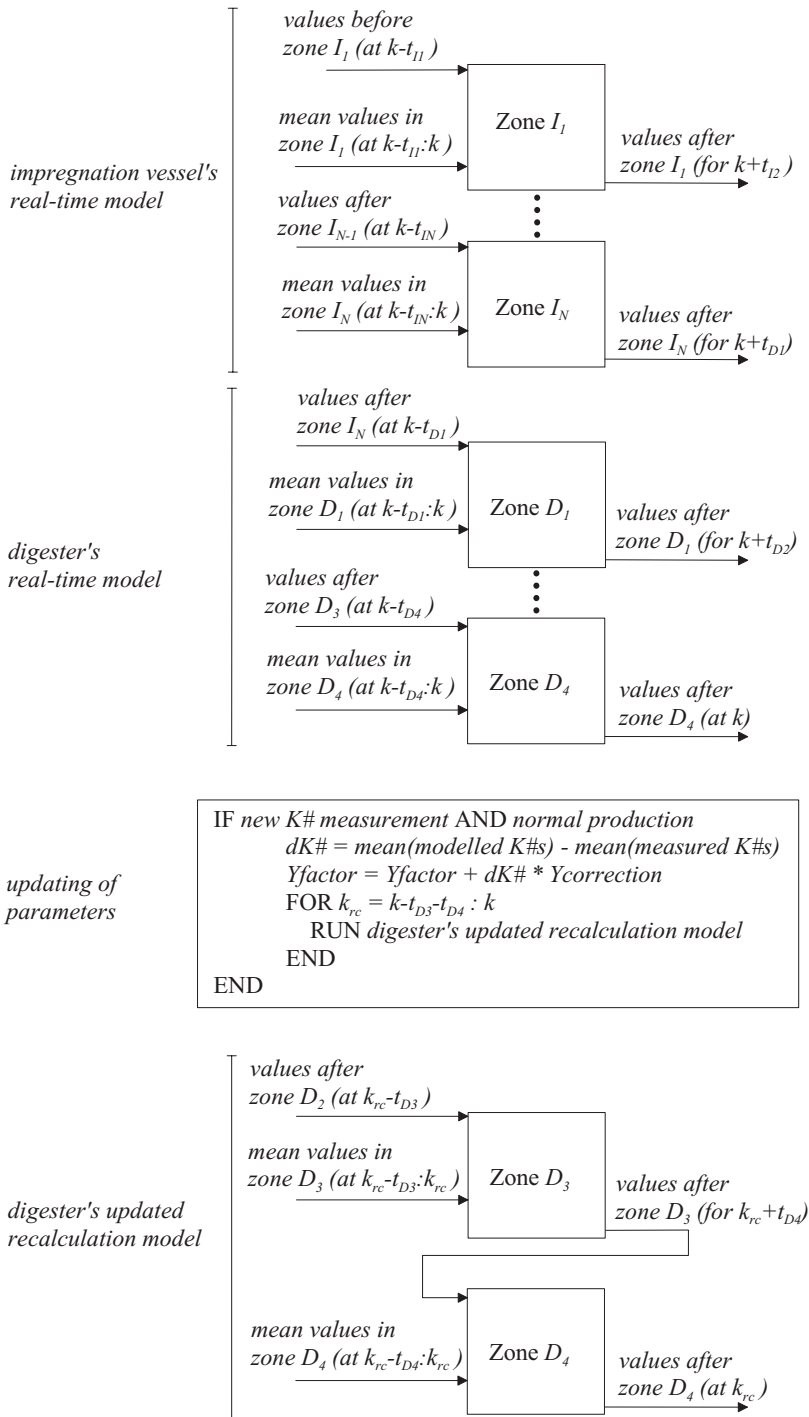


Fig. 14. Structure of the real-time simulation model with recalculation in Downflow Lo-SolidsTM cooking; detailed structure of one calculation zone I_x or D_x is shown in Fig. 12.

5.3.2 Prediction model

The structure of the prediction model in the conventional cooking is presented in Fig. 15. The model is based on the real-time simulation model, see Section 5.3.1. Inputs for the prediction model of the blow-line Kappa number are real-time Kappa numbers after the impregnation vessel, process data at the top of digester, and predicted temperature profile of the cooking zone. The residence times are calculated like in the real-time model. It is assumed that the production rate changes will not be made within the prediction horizon. The prediction of the temperature profile is presented in Section 6.2.1. The alkali profile is calculated as in the real-time Kappa number model, see Section 6.2.2.

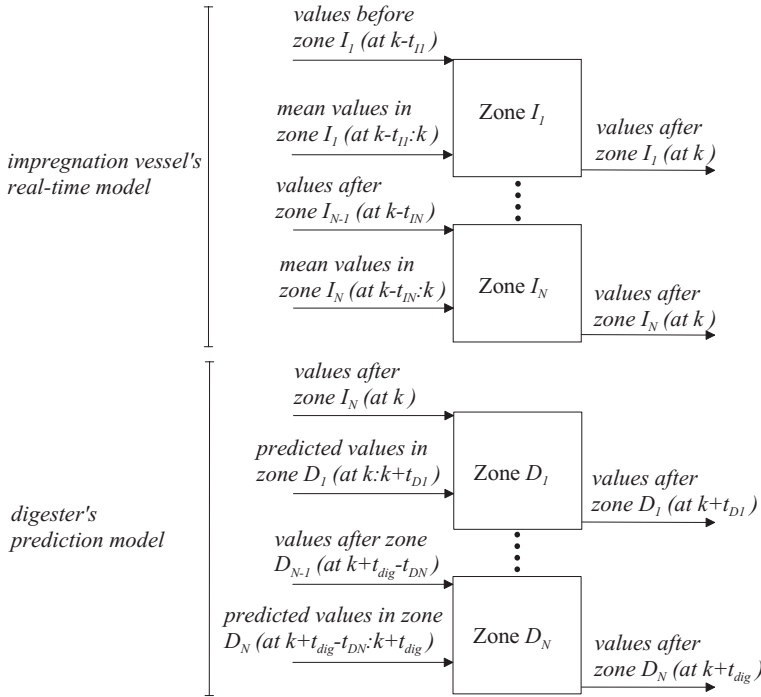


Fig. 15. Structure of the prediction model in conventional cooking; detailed structure of one calculation zone is shown in Fig. 12.

The structure of the prediction model in Downflow Lo-SolidsTM cooking is presented in Fig. 16. The model is based on a real-time simulation model, see Section 5.3.1. A real-time modelled Kappa number after zone D_2 is the input for the prediction model. The temperature profile of zone D_3 is predicted and the alkali profile is modelled as a function of the lignin and carbohydrates yields and liquor-to-wood ratio, as in the real-time modelling. As in the case of real-time modelling, cooking

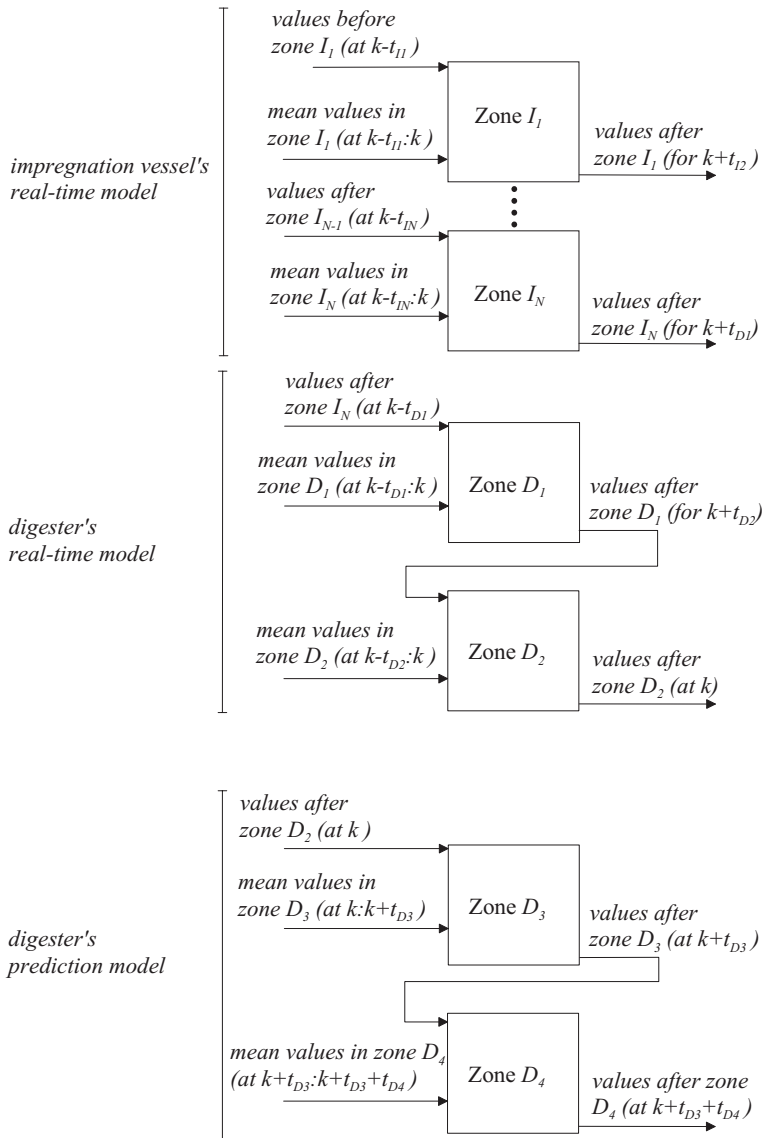


Fig. 16. Structure of the prediction model in Downflow Lo-SolidsTM cooking; detailed structure of one calculation zone is shown in Fig. 12.

zone D_3 is modelled in one part and in four parts. It is assumed that production rate changes will not be made within the prediction horizon.

5.4 New Kappa number control strategy

As was presented in Section 4.2, H -factor (Vroom 1957) and Hatton's Kappa number model (Hatton 1973) are widely used in the Kappa number control. The H -factor, which expresses the cooking temperature and time as a single variable, is used as an input of the Kappa number model. Other inputs of the Kappa number model are liquor-to-wood ratio and process specific model parameters. Despite of the wide usage, there are weaknesses in both H -factor and Hatton's Kappa number model. Main weakness in the Hatton's model is the inadequate consideration of the chemical concentrations. Sulfidity is not utilised in the model, and alkali concentration is based on the alkali charge, *i.e.*, the alkali profile is not properly considered. The alkali charge is controlled in proportion to the production rate without utilising on-line information about the Kappa number model. In practice, lignin is the only wood component that is modelled in the Hatton's model, although the model was proposed also for the modelling of pulping yield.

The H -factor describes the temperature dependence of reaction rate, but it does not describe any absolute reaction rate. Process operators are familiar with the H -factor, and sometimes it is manually controlled based on practical knowledge. In any case, H -factor is not informative as such without any other process information. In addition, both H -factor and Hatton's Kappa number require on-line updating because of unmeasurable disturbances, like variations in chip's quality.

In the new Kappa number control strategy, the control is based on the difference between predicted and target blow-line Kappa numbers. The blow-line Kappa number is predicted with the simulation model as shown in Section 5.3. The simulation model includes features to describe the profiles of temperatures, chemical concentrations and wood components within the cooking process. The calculation of the chip column's packing degree and residence times are also included. The main difference and advantage compared to the concept of Vroom's H -factor and Hatton's Kappa number model is, that process conditions are more comprehensively covered in the simulation model. The most important benefit is the better consideration of the alkali profile.

The iterative procedure of the determination of alkali and temperature set points is illustrated in Fig. 17. More detailed structures of the control strategy are shown in Figs. 18 (conventional cooking) and 19 (Downflow Lo-SolidsTM cooking). The blow-line Kappa number was predicted before the cooking zone, at the top of the digester in the conventional cooking (Fig. 1), and before zone D_3 in the Downflow Lo-SolidsTM cooking (Fig. 2). Inputs for the prediction model of the blow-line Kappa number were the outputs of the real-time simulation model and process data at the prediction point. The parameters of the real-time models were used in the prediction models.

In the control strategy, set points of cooking temperature and alkali charge, or only cooking temperature, are iteratively solved. In the first iteration round, new set points for both alkali charge and cooking temperature are calculated as a function of the difference between the predicted and target blow-line Kappa numbers. In the next iteration rounds, only the temperature set point is changed. Iteration is continued until the absolute value of the Kappa number difference is smaller than

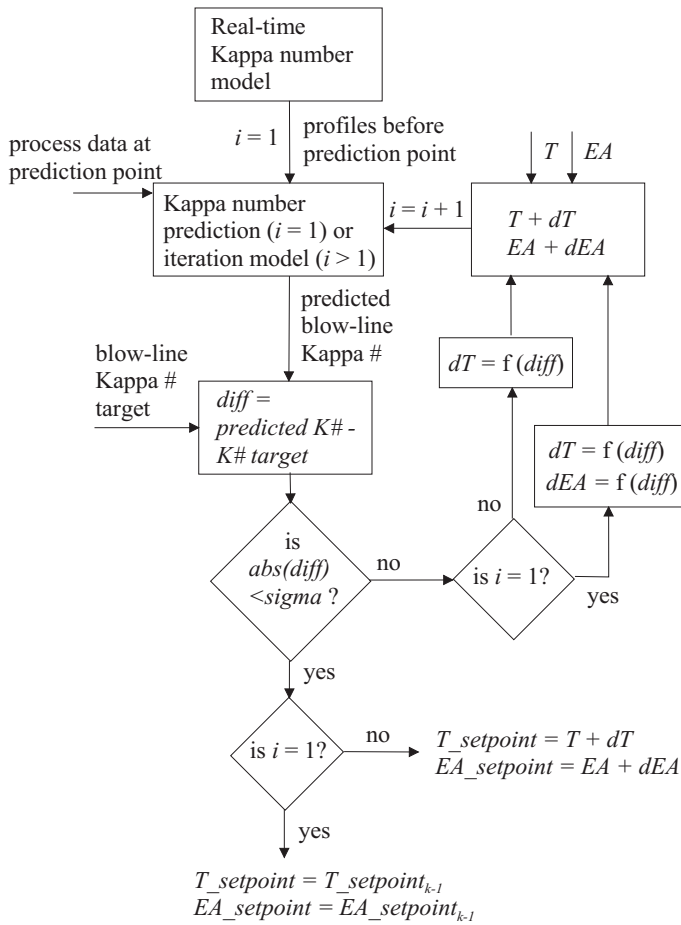


Fig. 17. Determination of temperature and alkali set points in the new Kappa number control strategy.

the iteration criteria σ . The relation between iteration's step size and the Kappa number difference σ , has to be experimentally determined based on process knowledge and the behaviour of the Kappa number model. To ensure that the iteration converges, the step size has to be restricted to a certain maximum. In case that iteration starts to diverge, the iteration step will be halved.

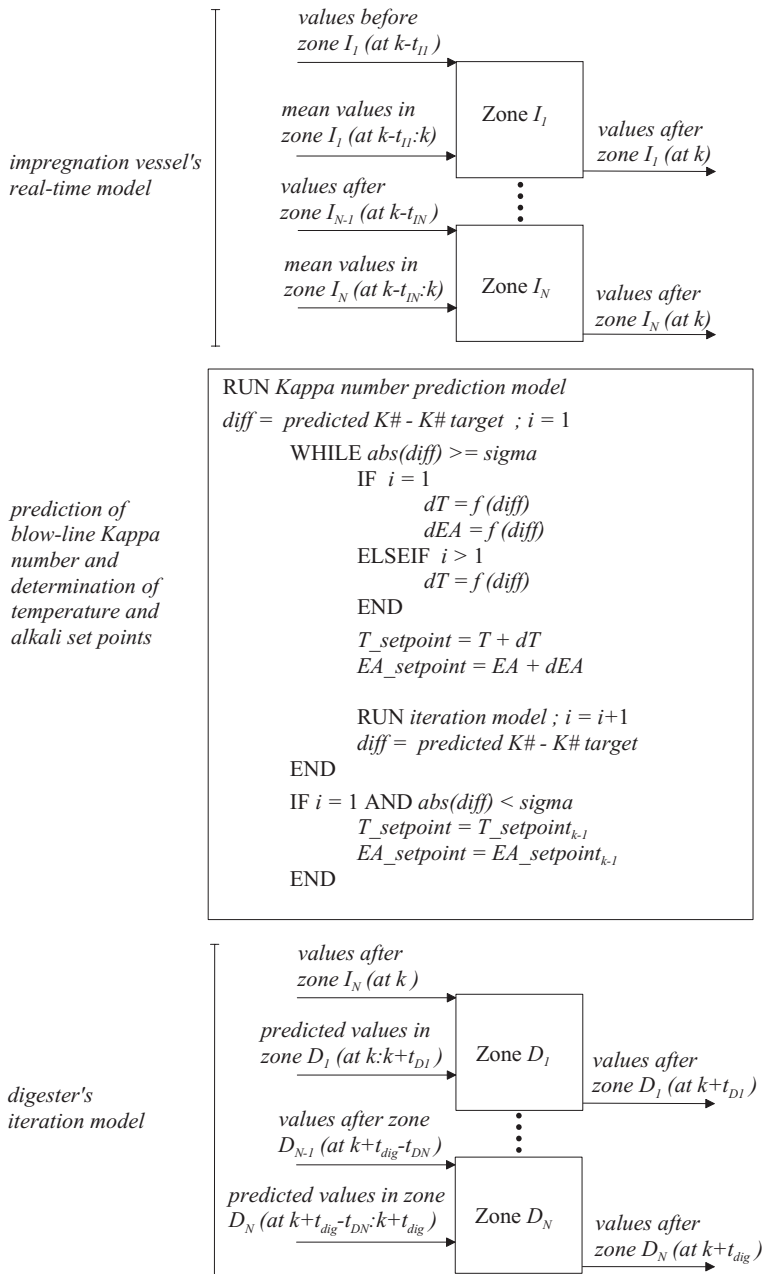


Fig. 18. Structure of the iteration model in conventional cooking; detailed structure of one calculation zone is shown in Fig. 12; see also Fig. 17.

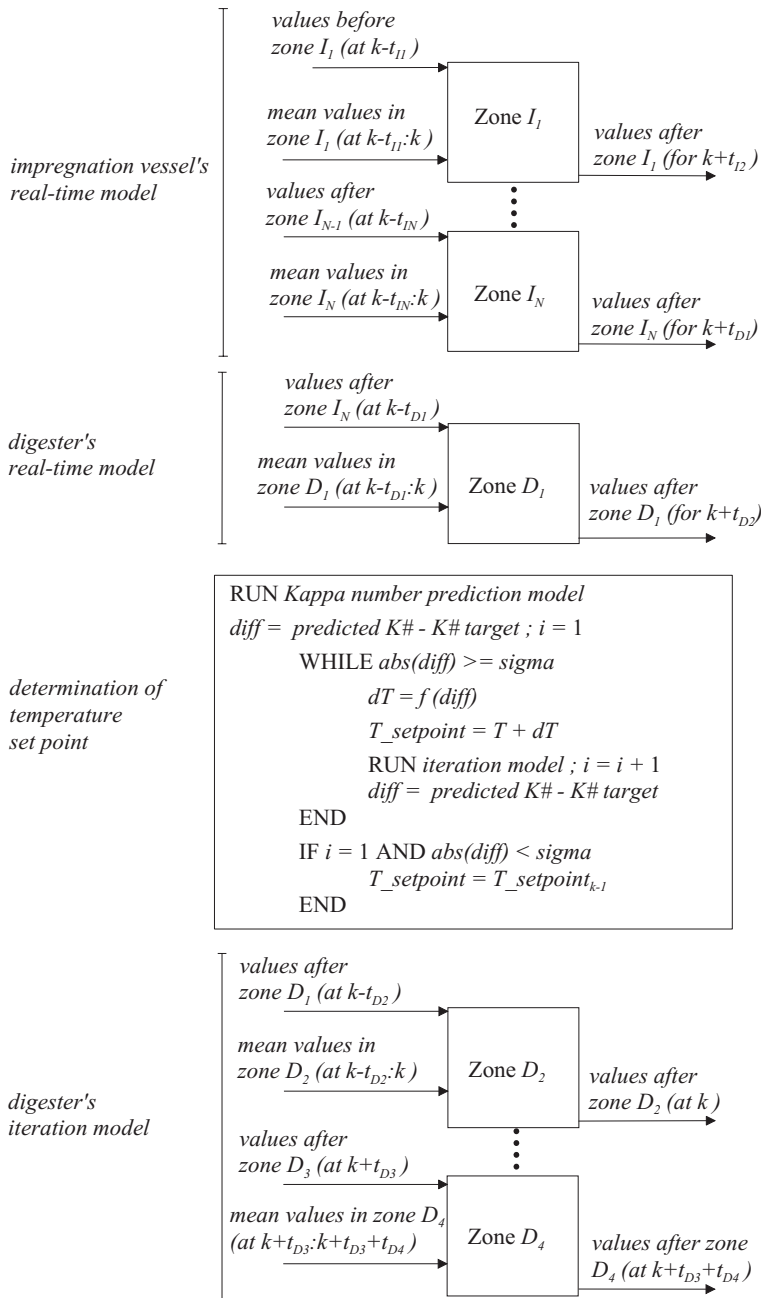


Fig. 19. Structure of the iteration model in Downflow Lo-SolidsTM cooking; detailed structure of one calculation zone is shown in Fig. 12; see also Fig. 17.

6 Simulation results and discussion

Results of the thesis were based on the investigations of two industrial continuous cooking processes. The modelling results of normal production periods without any major disturbances were chosen to be presented. However, because of the complex nature of the processes, minor disturbances occurred in the example periods. It should also be mentioned that data from Downflow Lo-SolidsTM process is of a period, when the process modifications from conventional to Downflow Lo-SolidsTM cooking were carried out, but operating practises were not yet totally established. Measurement data, averaged for one minute in the automation system, was available. Data was analysed, and single, clearly faulty measurements were replaced with earlier, proper measurements. Modelling was done using Matlab[®] software.

Accuracy and reliability of the modelling results was evaluated in several ways. Accuracy was measured by calculating the root mean squared error (RMSE). A benefit of RMSE is that it is measured in the same units as the original data. A drawback of RMSE is that large errors can dominate the value. Reliability was measured by calculating mean errors (ME), which represent the average deviation between the modelled and measured values. Simulation models were updated on-line using the mean errors. An assumption of normally distributed data was made and confidence intervals for the mean errors of modelled Kappa numbers were calculated. The assumption was tested with Matlab's *kstest* and *normplot* functions. An example of a normal probability plot is shown in Fig. 20. A linear line in the plot indicates that data is normally distributed. According to the tests the data was not normally distributed at a significance level of 95%. The assumption was further tested with Matlab's *bootstrp* function, which is based on the resampling of original data. A mean value was calculated using 10000 resampled data sets. Resulting mean values at 95% confidence intervals were almost equal to the values of the original data sets. Thus, the assumption of the normally distributed data can be said to be feasible. The non-linearities in the error data were mainly caused by grade and production rate changes, and unmeasurable variations in the quality of raw material.

A cross-correlation analysis was also considered when evaluating the accuracy of the Kappa number models. Analysis is problematic because of the different sampling intervals of the measured and modelled Kappa numbers. In addition, correlation

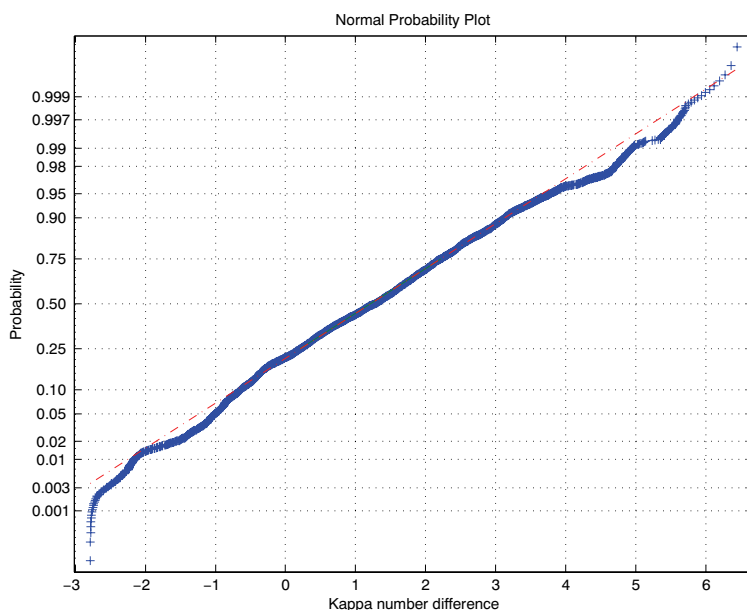


Fig. 20. A normal probability plot of the errors between predicted and measured blow-line Kappa numbers during all softwood periods in Downflow Lo-SolidsTM cooking (See Fig. 62 in p. 114.).

coefficients don't give reliable information if there exist only minor changes in the Kappa numbers. That is a usual case, especially in hardwood cooking, during steady production periods. Instead of the cross-correlation analysis, the accuracy of the Kappa number models was evaluated by comparing the measured and modelled Kappa numbers with the Kappa number target.

In the case of the conventional process, all modelling results are presented from a production period of 8800 minutes from winter. Three softwood and two hardwood periods are included. In the consideration of the results, the calculations of errors, differences *etc.*, are not made during the grade changes. In conventional cooking, calculations are made for the following periods: HW 0–1535, 3250–5740 and 8000–8800 minutes, SW 1700–2950 and 6050–7650 minutes.

In the case of the Downflow Lo-SolidsTM process, all results are presented from a production period of 13000 minutes from autumn. Two hardwood and three softwood periods are included. In the consideration of the results, calculations of errors, differences *etc.*, are not made during the grade changes. In Downflow Lo-SolidsTM cooking, calculation periods are: HW 1800–3800 and 8000–9600, SW 0–1500, 4200–7800 and 10000–13000 minutes.

6.1 Residence times

In the case of the Downflow Lo-SolidsTM process, residence times of the chips were calculated as a function of a production rate target and residence times given by the equipment supplier. In the case of the conventional process, residence times of the chips in the impregnation vessel were calculated as a function of production rate target and residence times given by the equipment supplier. The residence times in the digester in the conventional process were calculated based on the chip column's packing degree (See Section 3.3.), as is presented in this section.

An example of the chips' volume fractions in the digester is shown in Fig. 21. Chip pressures in the digester in the example case are shown in Fig. 22 and residence times of chips are shown in Fig. 23.

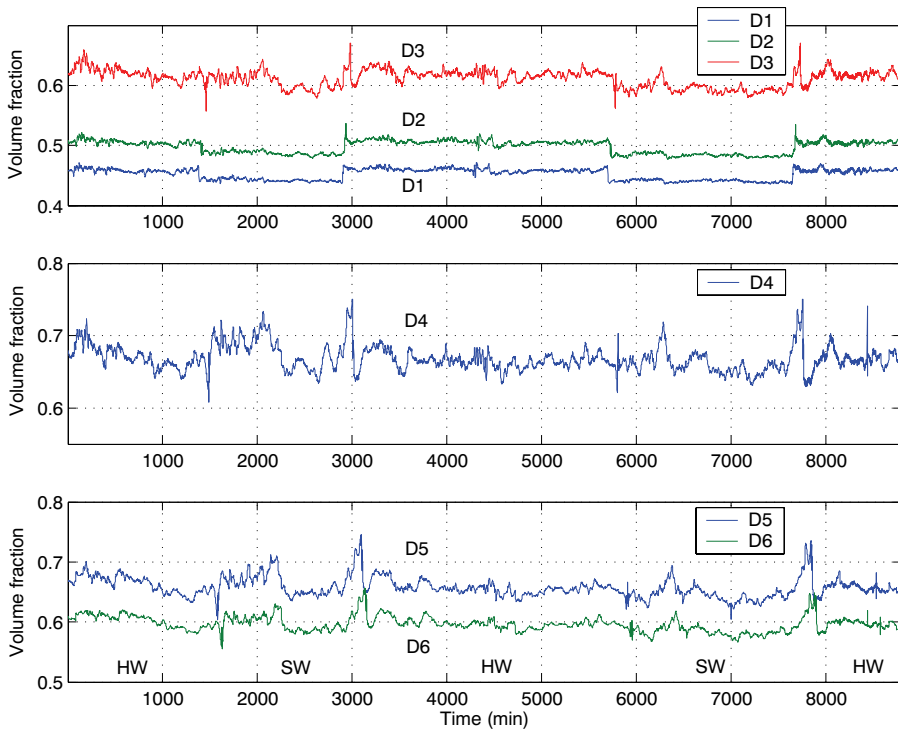


Fig. 21. Modelled chips' volume fractions in digester's zones D_1 – D_6 (conventional process).

In the case of the conventional process, the volume flow of chips was calculated from the production rate (calculated in automation system; $\frac{adt}{day}$). The production rate was converted to the volume flow of chips using yields (48% for SW and 53%

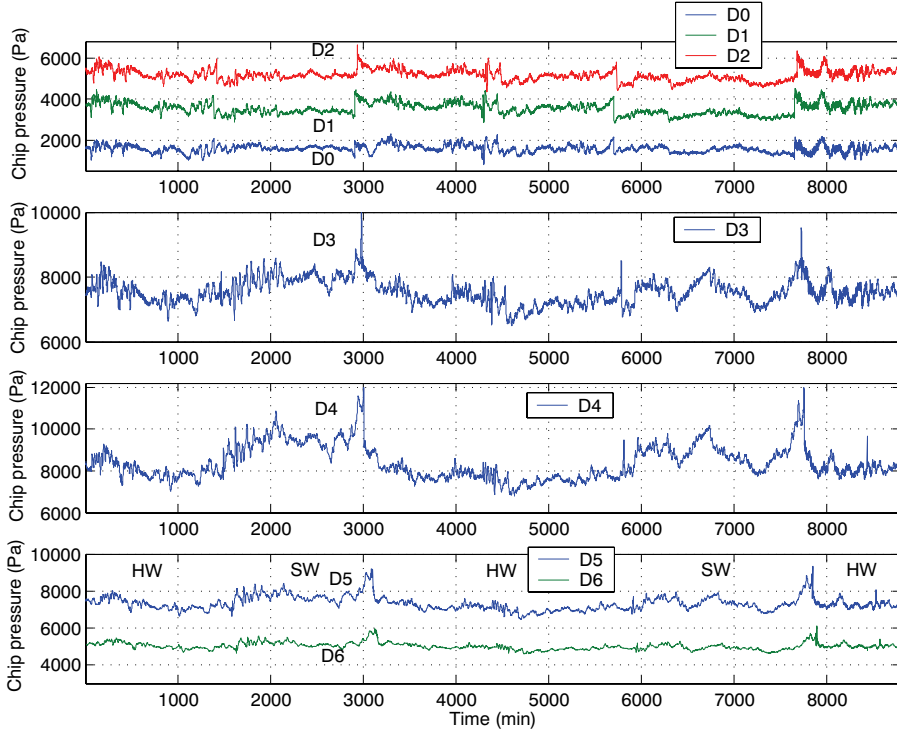


Fig. 22. Modelled chip pressures in digester's zones D_1 – D_6 (conventional process).

for HW) and approximated densities for chips ($490 \frac{kg}{m^3}$ for SW and $570 \frac{kg}{m^3}$ for HW) before being fed into the process. Residence times were calculated using volume flow of chips and calculated volume fractions.

The accuracy of residence times depends on the accuracy of the several models applied. The volume fraction of chips is a function of cooking degree and chip pressure (See Eq. 25.). The volume fraction is directly affected by parameters k_0 and k_3 . Parameter k_1 affects via chip pressure and parameter k_2 affects via Kappa number. Optimised combinations of original and optimised parameters of Eq. 25 are shown in Table 12.

Table 12. Optimised chips' void fraction constants of Eq. 25.

Wood species	k_0	k_1	k_2	k_3
Softwood	0.644	0.59	0.129	0.8
Hardwood	0.630	0.64	0.114	0.7

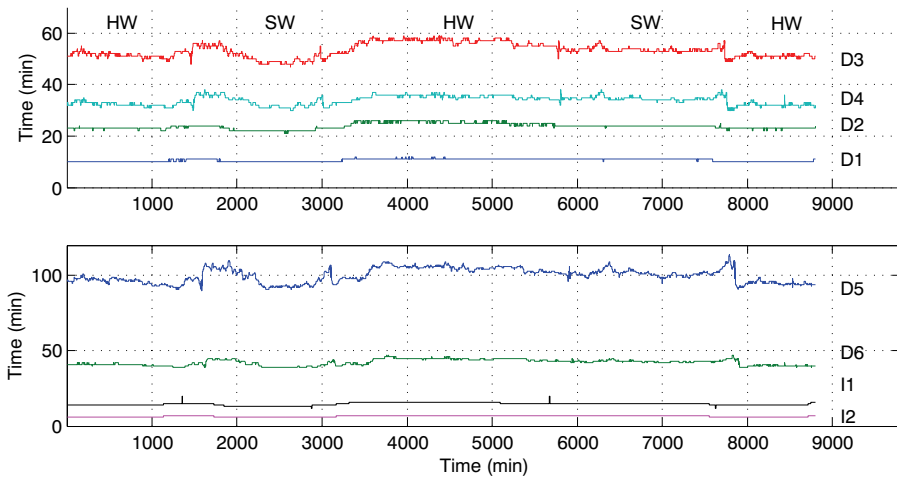


Fig. 23. Modelled chips' residence times in impregnation vessel and digester (conventional process).

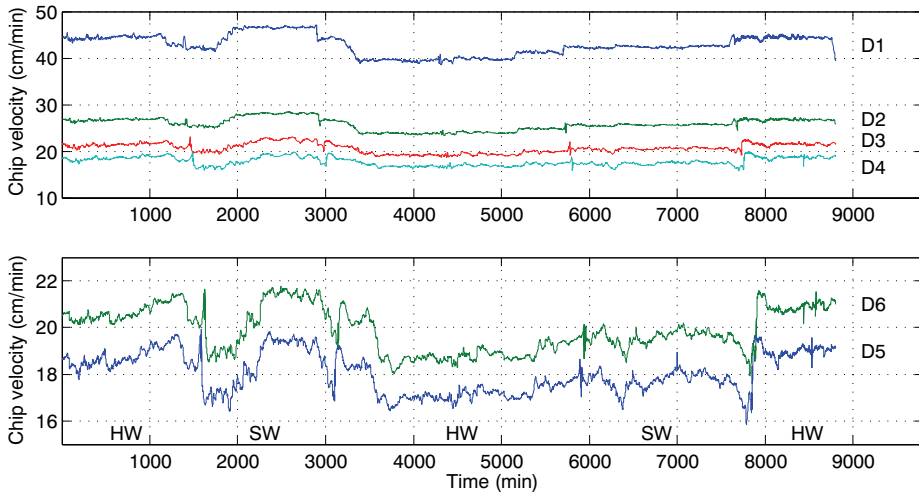


Fig. 24. Modelled chips' velocities in conventional process.

Chip pressure is a function of gravity force, friction force between chip column and digester wall, and friction force between chip column and liquor (See Eq. 26 in p. 47.). Gravity force, caused by density difference between chips and liquor, is the most important variable. As the densities of fully penetrated chips were used

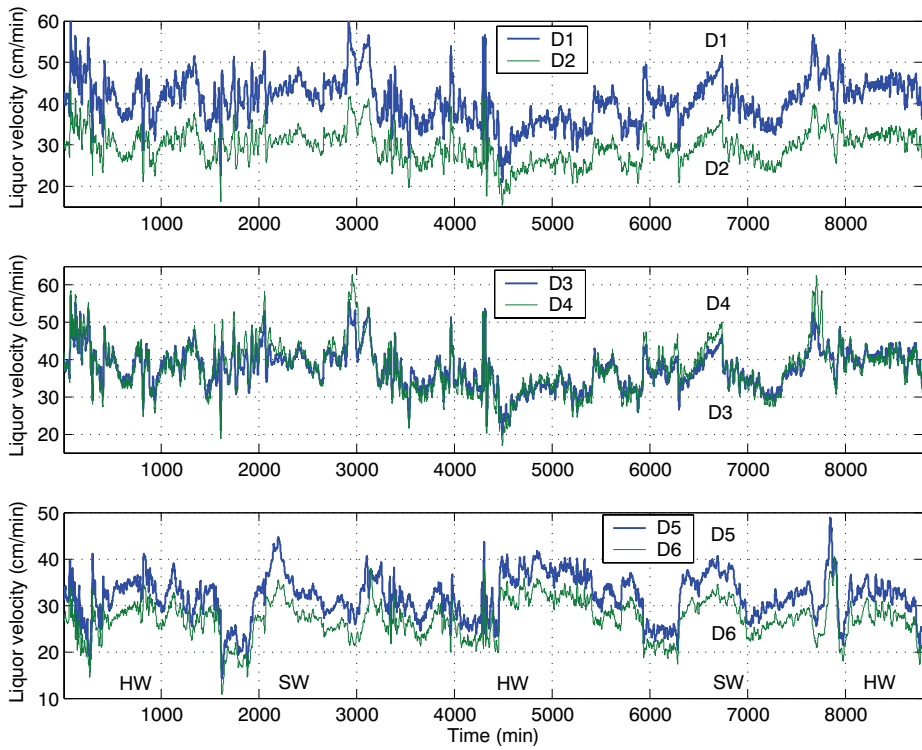


Fig. 25. Modelled liquor velocities in conventional process.

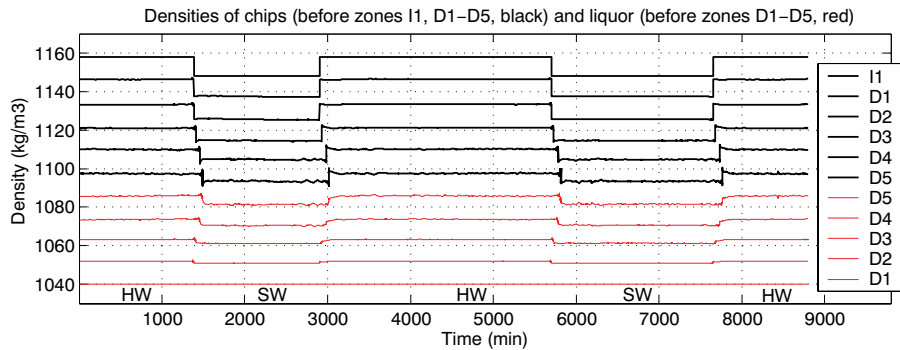


Fig. 26. Approximated chip and liquor densities in conventional process (before zones).

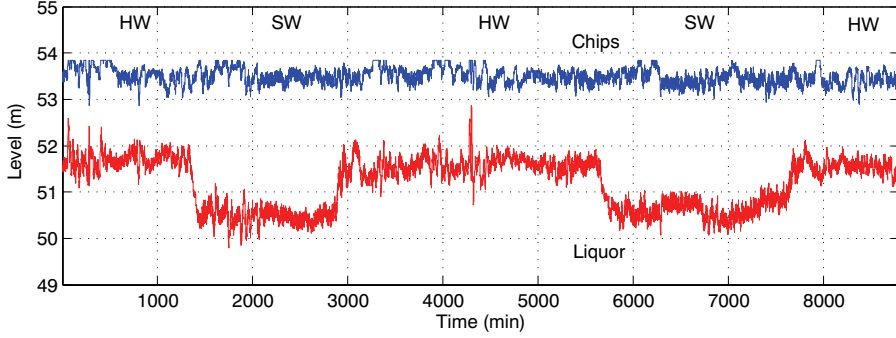


Fig. 27. Measured chip and liquor levels in conventional process.

1148 $\frac{kg}{m^3}$ for SW and 1158 $\frac{kg}{m^3}$ for HW. In Gullichsen (2000a) (p. A249), a value of 1150 $\frac{kg}{m^3}$ has been mentioned for SW. A constant value of 1040 $\frac{kg}{m^3}$ was used for the density of liquor at the top of digester. Density of 1050 $\frac{kg}{m^3}$ was mentioned in Holmlund & Parviainen (2000) (p. B39) for black liquor with a concentration of 16% of dry solids. In Lemmetti (1996), the concentration of dry solids in the transfer circulation flow of conventional cooking has been measured to be 13–19%. Density profiles of liquor and chips in the digester were calculated in proportion to total yield. Approximation of the densities was made because any measured density profiles were not available. Coefficient of static friction between wood and metal varies between 0.2–0.6, and friction between wet wood and metal is mentioned to be 0.2 (Engineers Edge 2006). A value of 0.3 was chosen, because of the higher friction at the areas of extraction screens.

Liquor flow resistance was calculated using Eq. 24. Parameters R_1 and R_2 for the equation were taken from Härkönen (1987) in the case of softwood and from Lammi (1996) in the case of hardwood (Scandinavian Birch); see Table 8 in p. 47. Ergun equation, presented by Härkönen (1987), differs from the original Ergun equation (Ergun (1952), Bird *et al.* (1960), Härkönen (1984)) because of a typing error in the exponent of ϵ_c in the latter term (Härkönen 2005).

Parameters presented by Härkönen (1987) (for Eqs. 24–26) have been defined for a special test digester, in which, *e.g.*, chip column did not move. Thus, parameters presented by Härkönen (1987) are only indicative, and should always be optimised case by case. In this study, the optimisation of the parameters was based on the overall consideration of chip pressures, chips' volume fractions and residence times. The results were verified by comparing the calculated residence times (using chips' velocities, Fig. 24) with residence times given by equipment supplier. The residence times were also evaluated by considering the correlation of modelling results and process data.

In washing zones D_5 and D_6 , the same density values for both softwood and hardwood grades were used. The density of the chips was 1080 $\frac{kg}{m^3}$ in D_5 and 1070 $\frac{kg}{m^3}$ in D_6 . The density of the liquor was 1020 $\frac{kg}{m^3}$ in D_5 and 1010 $\frac{kg}{m^3}$ in D_6 .

Parameters for Eqs. of chip pressure (Eq. 26), liquor flow resistance (Eq. 24) and chips' volume fraction (Eq. 25) were the same as in the cooking zones D_1 – D_4 . After the cooking zones, chip column consists more or less of chips and individual fibers. This means that the applicability of equations is questionable in the washing zone. It was also noticed that results were not usable without any modifications, because chip pressures, volume fractions and residence times were too high. The problem was solved by dividing the calculated chip pressures by two (resulting chip pressures used in the modelling are presented in subplot 4 in Fig. 22). However, it is clear that the models of washing zone require more detailed studies in the future.

The resulting volume fractions and chip pressures are shown in Figs. 21 and 22. Volume fraction profile is similar to the profiles presented by Härkönen (1987) and Saltin (1992). The values of volume fractions were restricted in each zone to eliminate very high chip pressures. However, chip pressure occasionally has relatively high values at the extraction screens (zones D_3 and D_4).

The chips' residence times are shown in Fig. 23. Residence times are shorter at the upper parts of digester (zones D_1 and D_2) and longer at zones D_3 and D_4 than the residence times given by equipment supplier. The total residence times are, however, almost equal. Chip and liquor velocities are shown in Figs. 24 and 25. Chips' velocity is highest at the top of the digester and decreases to the extraction screens because of the changes in the packing degree. At the same time, liquor velocity increases from top of the digester to the extractions. Liquor velocity in D_1 is higher than in D_2 , because the diameter of the digester vessel is smallest at the top. The chips' velocity increases slightly in the washing zone, when packing decreases.

In the case of the Downflow Lo-SolidsTM process, the amount of chips fed to the process is controlled by the chip screw speed. Actual production rate is calculated in the automation system utilising the screw speed and information given by the equipment supplier (filling rate vs. rpm of chip screw, packing degree of impregnation vessel and digester, yield *etc.*). Chip level in the impregnation vessel is also controlled with the chip screw speed. Thus, the production rate's target may differ from the actual production rate. The accuracy of the calculated production rate depends on the chip quality, moisture content, success of the air removal and impregnation, actual packing conditions vs. ideal packing conditions used by the equipment supplier *etc.* The use of the production rate target when calculating residence times causes some error. It may be estimated, however, that this error is not bigger than errors caused by unmeasurable factors if the chip screw speed would have been used. The accuracies of calculated residence times could be improved by considering the packing conditions in more detail, like in the case of conventional process.

6.2 Conventional cooking

In this section, modelling of temperatures, chemical concentrations, and profiles of Kappa number and carbohydrates in conventional cooking process are presented and discussed. The structure of the simulation model was presented in Section 5.3.

6.2.1 Temperatures

The temperature profile in the digester is affected by the heat of convection, heat of chemical reactions, and heat of conduction (vertical, between the two phases, and through the side walls of the vessel). Steady-state calculations have shown that with respect to the chip phase, the heat of reaction and the heat of conduction from the liquor are the most significant contributions. With respect to the liquor phase, the convection and the heat of conduction from the chips are the most significant. The vertical heat conduction and the heat of conduction through the walls are negligible for both phases. (Michelsen & Foss 1996a)

In this thesis, the temperature profile of chip column was assumed to be affected by the heat of reactions and conduction from free liquor.

The temperatures measured from the following points were available in the conventional process (See. Fig. 1.): impregnation vessel's feed circulation flow (T1, before zone I_1), middle part of impregnation vessel (T2, between zones I_1 and I_2), digester's feed circulation flow (T3, between zones I_2 and D_1), heated steam in digester (T4, before zone D_1), upper part of digester (T5, between zones D_1 and D_2), upper extraction flow (T6, between zones D_3 and D_4), lower extraction flow (T7, at the lower part of zone D_4), digester's washing zone (T8, between zones D_4 and D_5), digester's washing zone (T9, between zones D_5 and D_6) and blow-line flow (T10, after zone D_6).

Real-time temperature profile of conventional process, noticing the exothermic nature of delignification, was constructed weighting the temperatures as follows: I_1 (T2-3° 100%), I_2 (T2 50%, T3 50%), D_1 (T4-3° (SW) or T4-4° (HW) 80%, T5 20%), D_2 (T5+1° 100%), D_3 (T5+2° 40%, T6 60 %), D_4 (T6 100%), D_5 (T7 60%, T9 40%) and D_6 (T9 50%, T10 50%).

The measurements were filtered using exponentially weighted moving averages (EWMA). Weighting factor 0.4 was used. An example of a temperature profile is shown in Fig. 28.

Different modelling techniques have been used by the research group to predict the temperature profile of conventional cooking. Methods of fuzzy clustering (see *e.g.* Babuska (1998) and Bezdek *et al.* (1999)), see Ahvenlampi *et al.* (2004), and least squares regression (LS, see *e.g.* Basilevsky (1994)), see Tervaskanto *et al.* (2003, 2004), have been tested and validated. It was found, that the accuracies of linear models are adequate in the Kappa number modelling. Thus only LS method was used in the modelling of temperature profile in this thesis.

Temperatures after cooking zones D_1 – D_4 were predicted at the top of digester. Temperatures at the washing zone were not predicted, and real-time temperatures

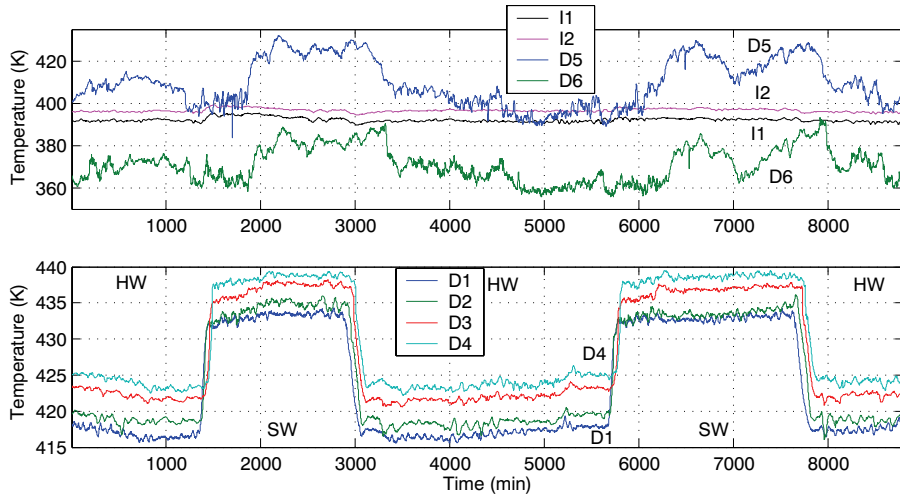


Fig. 28. Modelled real-time temperature profile of conventional process.

were utilised in zones D_5 and D_6 . The reason for this was, that it is not possible to know about the future operations in the washing zone, as early as at the prediction time. Separate models for softwood and hardwood cooking were utilised for temperatures after zones D_1 and D_2 . Temperatures at the upper and lower extraction were predicted using same model for both grades. Input variables of all models were the temperature of heated steam (T_4) and alkali concentration of digester's feed circulation flow (A_2). In the temperature model of lower extraction, the temperature of the upper part of washing zone (T_8) was also utilised.

The temperature profile in each zone was calculated by weighting the temperatures in the beginning and end of each zone as described in the case of real-time modelling, see Section 6.2.1. The predicted temperatures of zones were compared to the temperatures used in the real-time modelling. The prediction errors were eliminated by updating the prediction models on-line. The predicted values were corrected with the mean difference between predicted and measured temperatures from previous 150 minutes.

An example of predicted and real-time temperatures in zones D_1 – D_4 is shown in Fig. 29.

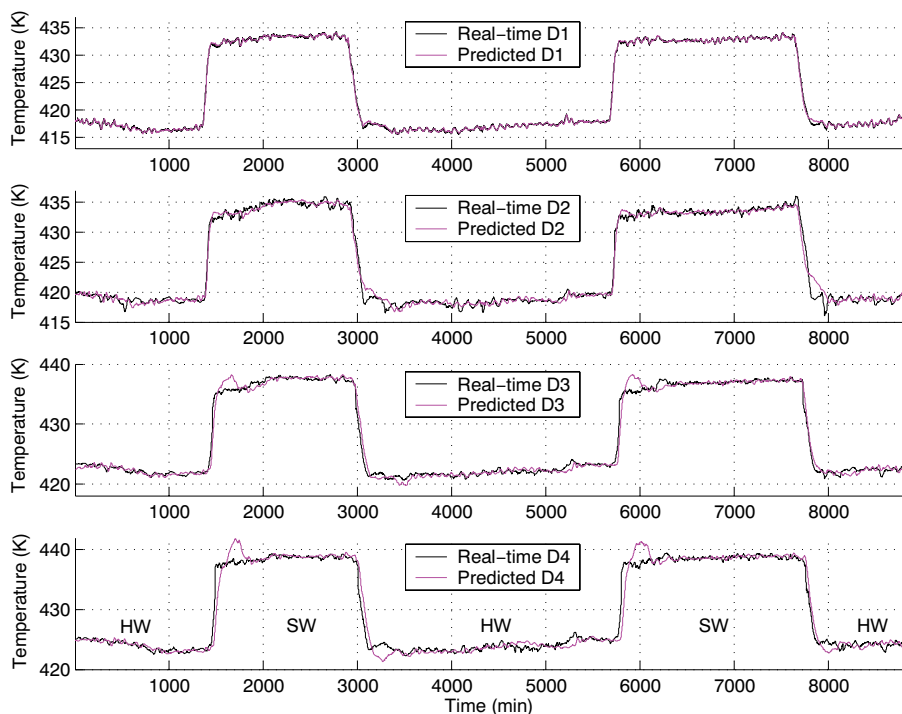


Fig. 29. Predicted and real-time modelled temperatures in zones D_1 – D_4 in conventional process.

6.2.2 Chemical concentrations

A chemical profile of the impregnation vessel and digester was constructed based on direct process measurements, flow balance based alkali concentrations, and calculated alkali consumption. The alkali profile inside chips was not calculated. Alkali concentrations inside chips and in free liquor were assumed to be equal.

A real-time alkali profile of the impregnation vessel was based on the impregnation vessel's feed circulation flow's effective alkali concentration, which was calculated from mass and flow balances. The alkali profile of the digester was based on the digester's feed circulation flow's measured effective alkali concentration (A_2 in Fig. 1). Alkali consumption, and further alkali profiles in the impregnation vessel and digester, were calculated as a function of lignin and carbohydrate yields and liquor-to-wood-ratio (Eq. 23). Examples of hydroxyl ion profiles in the impregnation vessel and digester are shown in Figs. 30 and 31.

The sulphidity of the white liquor was measured (Fig. 32) and it was assumed, that sulfide ions are not consumed in the reactions.

Different modelling techniques have been used by the research group to predict the alkali profile of the digester's cooking zone (Tervaskanto *et al.* 2004, 2003) and

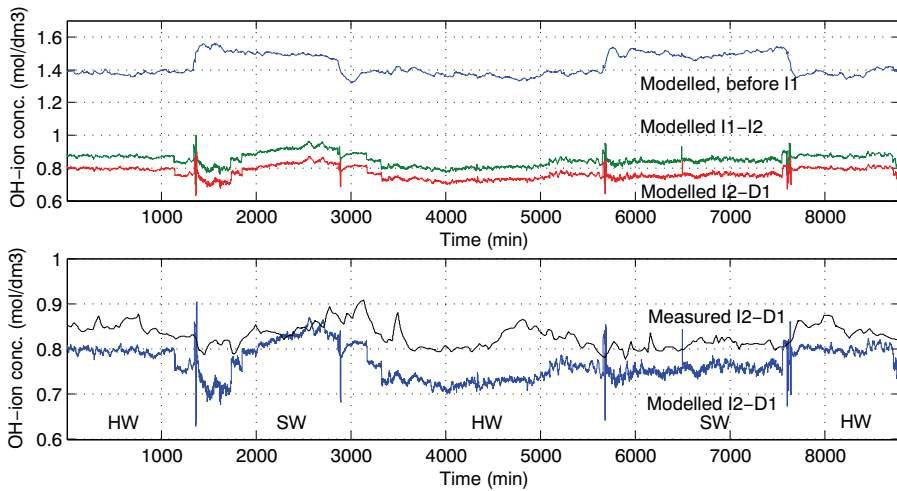


Fig. 30. Alkali profile of impregnation vessel in conventional process.

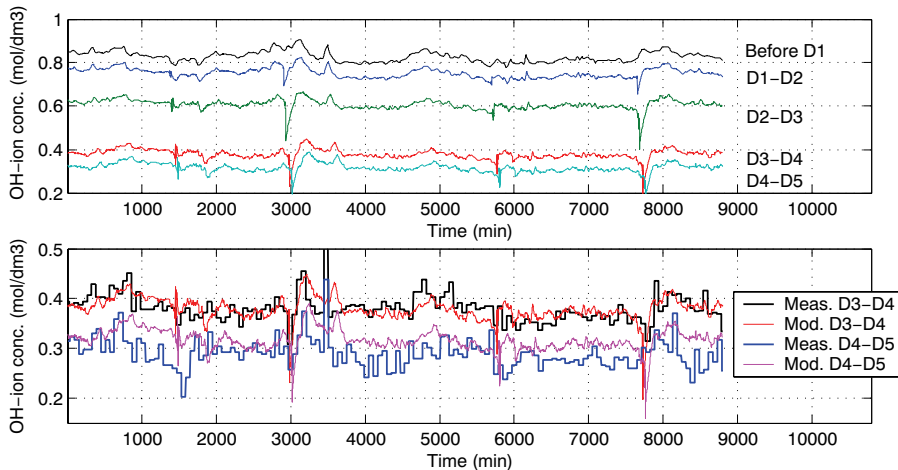


Fig. 31. Alkali profile of digester in conventional process.

(Ahvenlampi *et al.* 2004). Both linear and nonlinear modelling methods have been applied. Clustering methods (See *e.g.* Babuska (1998) and Bezdek *et al.* (1999)), neural networks (NN, See *e.g.* Haykin (1999) and Ikonen & Najim (2001).) and adaptive neuro-fuzzy inference systems (ANFIS, see *e.g.* Jang *et al.* (1997).) have been used in the non-linear modelling, and principal component analysis (PCA, See *e.g.* Basilevsky (1994) and Ikonen & Najim (2001).) and least squares re-

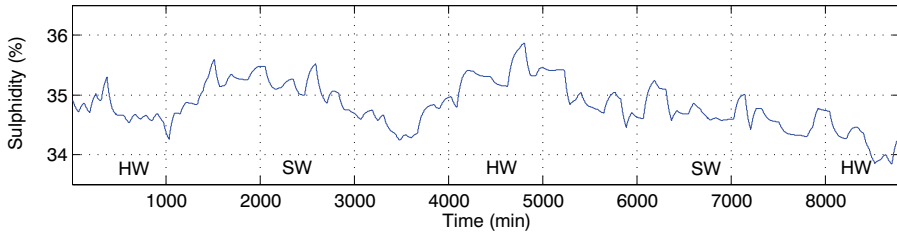


Fig. 32. Measured sulphidity of white liquor in conventional process.

gression have been used in the linear modelling (LS, See *e.g.* Basilevsky (1994) and Ikonen & Najim (2001).). The most accurate alkali model for extractions have been gained using NN models.

However, the real-time cooking model's alkali profile was found to follow the measured extraction flow concentrations well; see Fig. 31. The alkali profile also followed the profile of this type of process mentioned in the literature (Teder & Olm 1981). Thus, the alkali profile in the prediction model of Kappa number was calculated based on predicted lignin and carbohydrate yields and liquor-to-wood-ratio (Eq. 23).

An example of alkali concentrations after zones D_1 – D_4 , in the prediction model of Kappa number, is shown in Fig. 33.

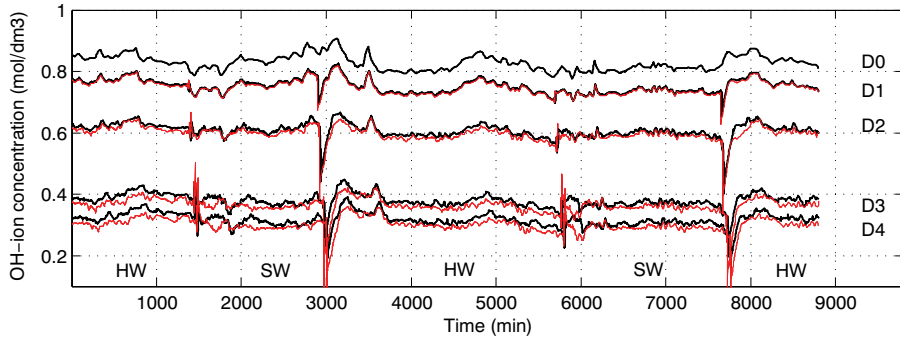


Fig. 33. Modelled alkali concentrations after zones D_1 – D_4 (red) in the prediction model of Kappa number, and the values of real-time model (black) at the top of digester (measured) and after zones D_1 – D_4 (modelled); conventional process.

6.2.3 Lignin

In the Kappa number modelling, Gustafson's model (See Section 3.2.) was utilised as described in Section 5.1. Input variables of the delignification model were grade, lignin content of chips, chemical concentrations, temperature and cooking time. Temperature and chemical concentration profiles were modelled as described in Sections 6.2.1 and 6.2.2. Cooking time was calculated as described in Section 6.1 as a function of the production rate target and chip column's compaction.

Lignin yields were calculated after each zone (I_1-D_6 in Fig. 1) once a minute, which is the sampling interval of process measurements in the automation system. Optimised hardwood parameters were used. After each measurement of blow-line Kappa number, the model parameters were adapted based on the difference between measured and modelled Kappa numbers. With the adapted parameters the lignin yields in digester (zones D_1-D_6) were recalculated. Recalculated real-time lignin yields are shown in red in Figs. 34 and 35. The original and recalculated real-time Kappa number profiles of the conventional digester are shown in Fig. 36. Recalculated real-time lignin profile and Kappa number profile are shown in Figs. 37 and 38. Recalculated real-time blow-line Kappa number, target and measured blow-line Kappa numbers and production rate are shown in Fig. 39.

In Figs. 40 and 41, the recalculated real-time Kappa number profile and blow-line Kappa numbers, when the original softwood parameters are used for hardwood, are shown.

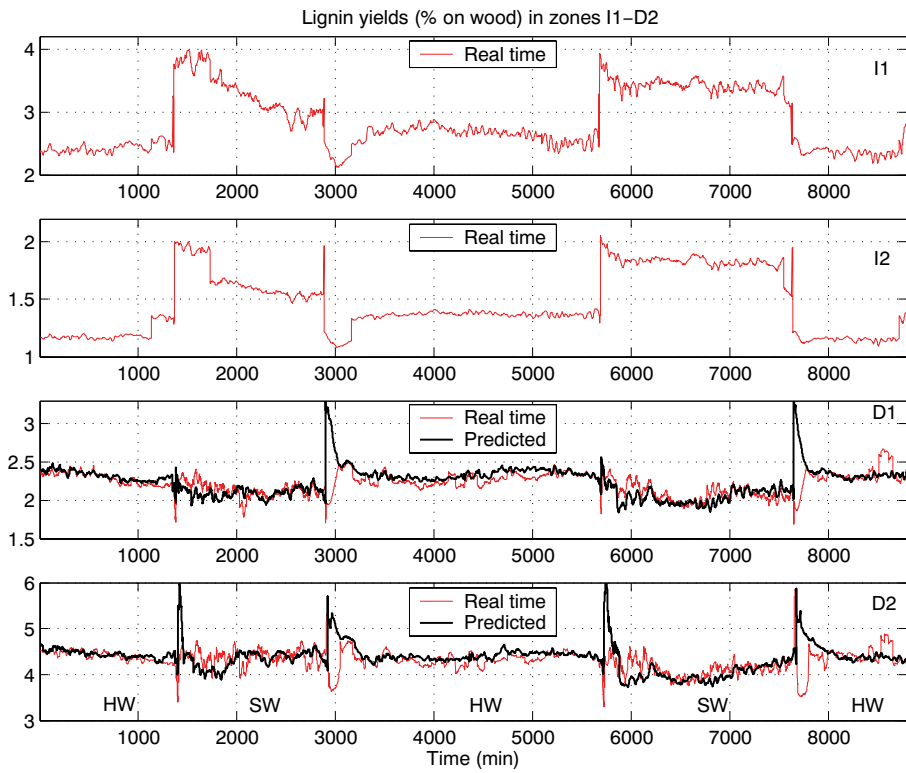


Fig. 34. Modelled real-time lignin yields in zones I_1 - I_2 ; recalculated real-time and predicted lignin yields in zones D_1 - D_2 ; conventional process.

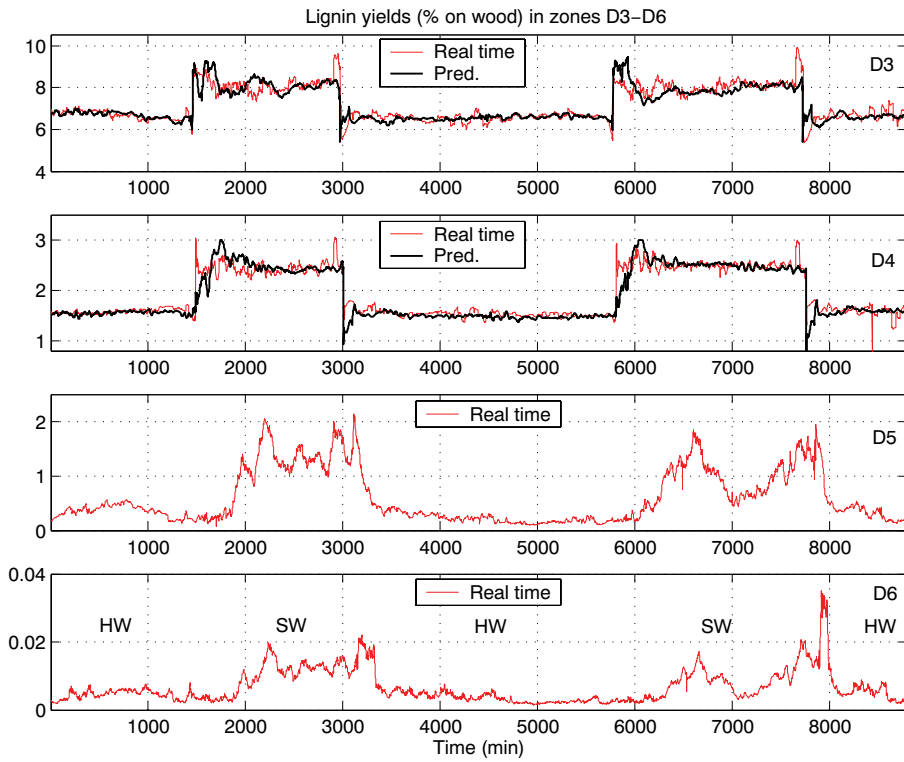


Fig. 35. Recalculated real-time and predicted lignin yields in zones D_3 – D_4 and modelled real-time lignin yields in zones D_5 – D_6 ; conventional process.

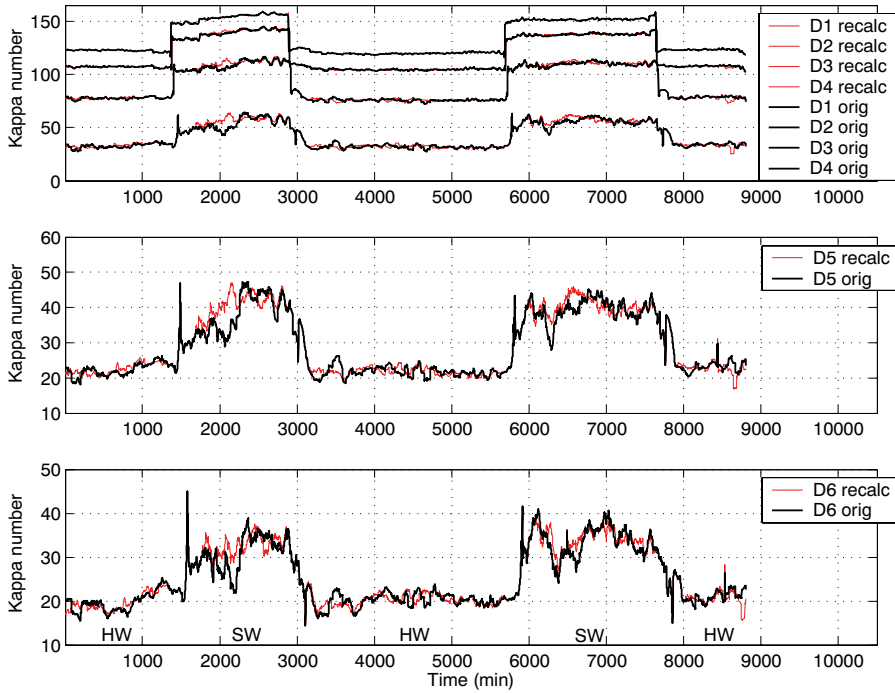


Fig. 36. Originally modelled and recalculated real-time Kappa number profiles in digester; conventional process (Kappa numbers before zones).

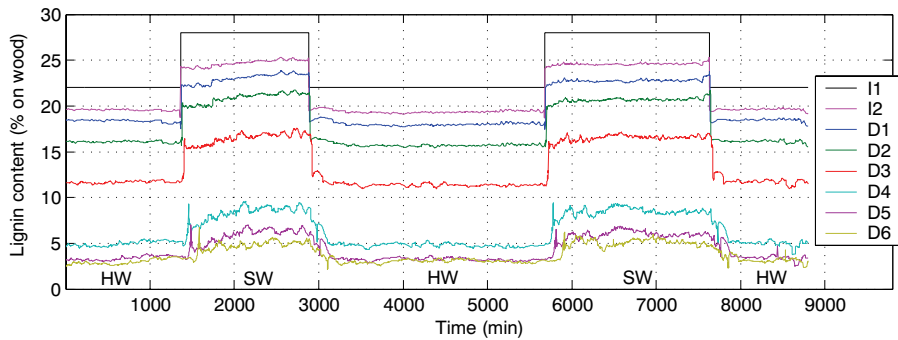


Fig. 37. Modelled real-time lignin content profile of chips in conventional process (lignin contents before zones I_1 – D_6).

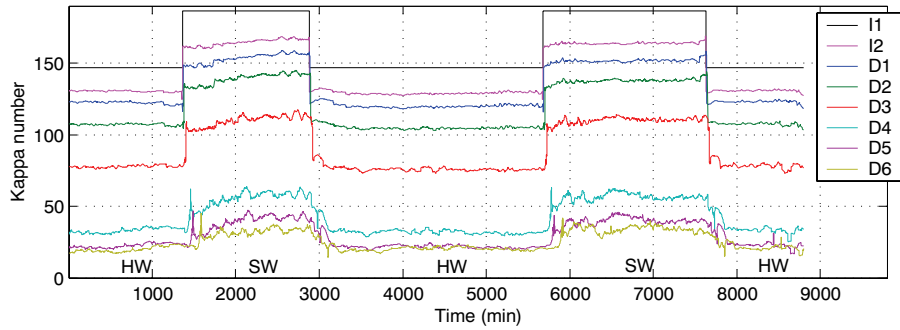


Fig. 38. Modelled real-time Kappa number profile of conventional process (Kappa numbers before zones).

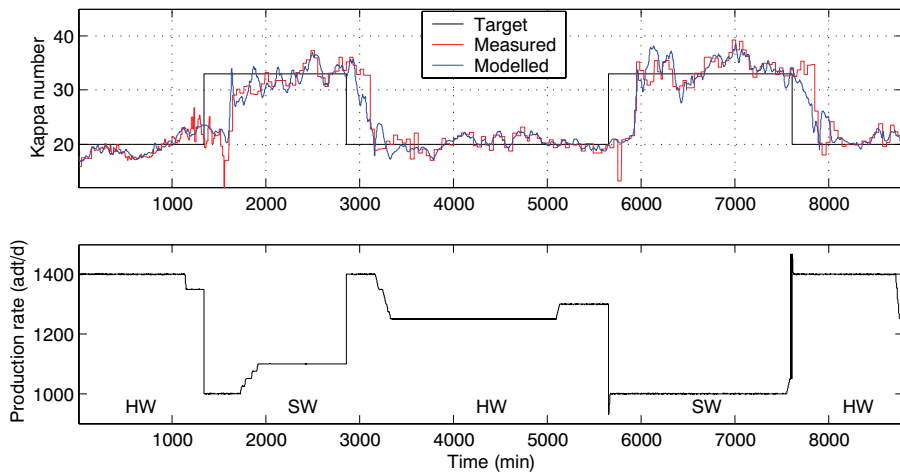


Fig. 39. Measured and modelled (recalculated) real-time blow-line Kappa numbers and production rate in conventional process.

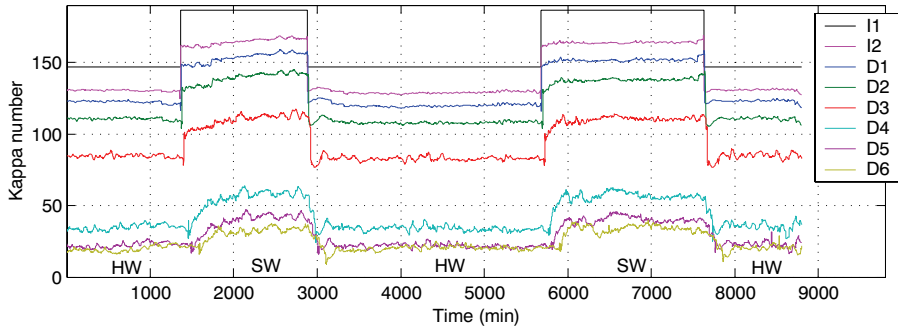


Fig. 40. Recalculated real-time Kappa number profile of conventional process (Kappa numbers before zones); original softwood parameters for hardwood.

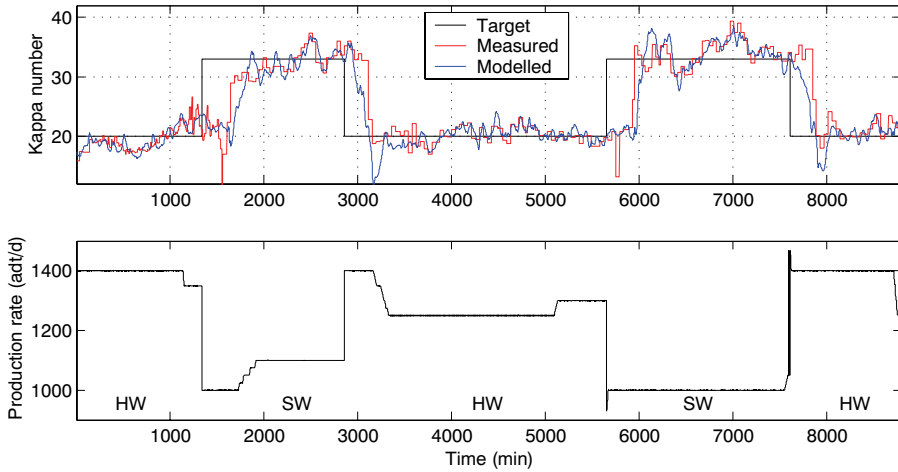


Fig. 41. Measured and modelled (recalculated) real-time blow-line Kappa numbers and production rate in conventional process; original softwood parameters for hardwood.

6.2.4 Carbohydrates

The carbohydrates were divided into cellulose and hemicellulose, and the yields were calculated according to Eqs. 18 and 19 in the initial phase, and Eqs. 20 and 21 in the bulk phase. The resulting yields were multiplied with factors shown in Table 13. The total yield of carbohydrates in the residual phase was modelled as a function of lignin yield using Eq. 17 (1.8 was used for q_r). Yields of cellulose and hemicellulose in the residual phase were calculated from the total carbohydrate yield in proportion to the contents of cellulose and hemicellulose after the bulk phase. Other wood components, *i.e.* extractives, were assumed to be removed during the steaming.

Real-time cellulose and hemicellulose profiles are shown in Figs. 42 and 43. The total yield profile (the amount of lignin and carbohydrates left in the wood) is shown in Fig. 44.

Table 13. Factors used to multiply resulting carbohydrate yields, when modelled with Eqs. 18–21.

Wood species	Eq. 18	Eq. 19	Eq. 20	Eq. 21
SW	0.6	0.5	1.8	1.7
HW	0.45	0.45	3.5	3.5

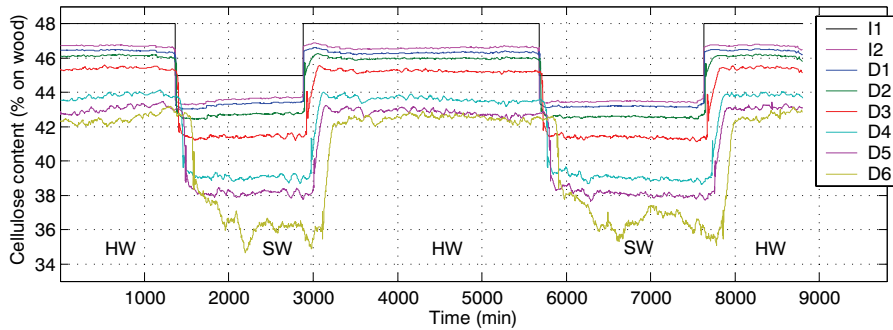


Fig. 42. Modelled real-time cellulose profile of conventional process (contents before zones).

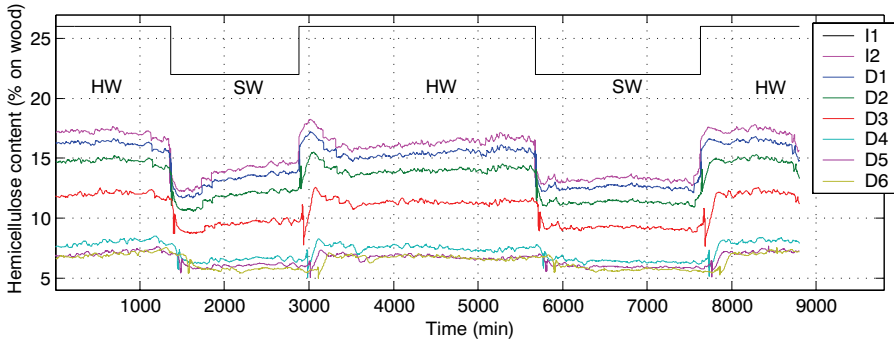


Fig. 43. Modelled real-time hemicellulose profile of conventional process (contents before zones).

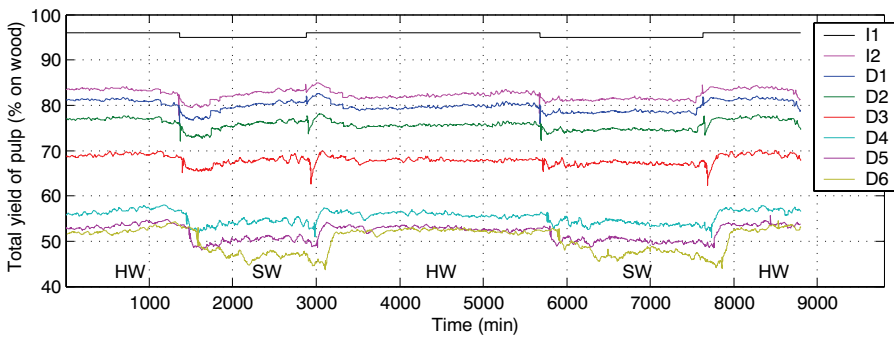


Fig. 44. Modelled real-time total yield profile of conventional process (yields before zones).

6.2.5 Discussion

Delignification and carbohydrate degradation in the impregnation vessel were modelled using real-time models. Because any predictions were not made, only T2 was required to describe the overall temperature in zone I_1 . T1 is measured from the return flow from the top of impregnation vessel after the addition of another circulation liquor. Thus, T1 does not give correct information about temperature profile in zone I_1 . T3 is measured from return flow from the top of digester. The liquor has not been in direct contact with heating steam and T3 gives good information about the conditions in zone I_2 , although some liquor is added to the bottom of impregnation vessel.

The alkali concentration at the top of impregnation vessel was calculated based on mass and flow balances. The accuracy of the alkali concentration can be eval-

uated by comparing the measured and calculated alkali concentrations after the impregnation vessel (See subplot 2 in Fig. 30.). The measured concentration is higher than modelled, because of white liquor addition. White liquor is added both before and after the impregnation vessel in proportion to production rate. Thus, differences between measured and modelled concentrations are mainly consequences of phenomena in the impregnation vessel. The accuracy of the modelled alkali concentrations is so good that modelling results in impregnation vessel can be said to be reliable.

Temperature profiles were not based on energy balances of the impregnation vessel or digester. The amount and temperature of heating steams and most liquor additions were known, but the amount of chips was not accurately known. Also, the amount of liquor at the top of digester was unclear, although an accurate liquor balance for the digester was possible to calculate. Few publications concerning reaction heats in conventional cooking exist. A temperature rise in cooking zones is known to be several degrees. When comparing temperatures T4, T5, T6 and T7, it looked obvious that the chips temperature at the top of digester is lower than steam temperature.

A constructed temperature profile in digester was mainly based on T4 and T6, which are the most reliable measurements. T5 is measured near the digester wall. However, a horizontal temperature profile at the upper part of digester is most likely even because the packing degree is low and risk of liquor channelling is small. T7 is measured from extraction flow, but washing liquor and channelling of liquor decreases the usability of T7. Because of the low packing degree, prediction models of temperatures are most accurate at the top of digester (See Fig. 29 and Tables 14 and 15.). In the case of softwood, predicted temperatures are too high, and in the case of hardwood too low during the example periods. The accuracy of the simulation results would probably be improved, if the prediction models of the temperature were determined from recent data.

Accuracy of a calculated alkali profile depends on the yields of wood components and *vice versa*. The predicted alkali profile is almost equal to the real-time profile (See Fig. 33 and Table 16.). Both models have a good accuracy with the measured alkali concentrations from the extraction (See Fig. 31 and errors in Table 17.). The alkali profile inside chips was not calculated, because on-line information about chip size distribution was not available. It has also been presented, that in steady-state the effective alkali concentrations in chips and free liquor are almost equal (Michelsen & Foss 1996a).

In softwood cooking, the modelled Kappa number after impregnation vessel is about 150 (See subplot 1 in Fig. 36.). This equals to lignin content about 22.5%, which is near the value of 22% mentioned by Olm & Tistad (1979). The Kappa number decreases from about 150 to about 35–40 in the bulk phase (zones D_1 – D_4). This means that in the bulk phase about 73–77% of lignin is dissolved (Bäckström *et al.* (1996) mentioned 80%). Thus, the amounts of lignin dissolved in the impregnation vessel and in the digester's zones D_1 – D_4 are similar as the values mentioned in the literature.

According to Marcoccia *et al.* (2000), about 20–30% of the wood dissolves during the impregnation phase. Total yield profile in Fig. 44 shows, that in both softwood

Table 14. Mean and root mean squared errors between predicted (updated on-line) and real-time temperatures in the case of Fig. 29; conventional process.

Error	Subplot 1	Subplot 2	Subplot 3	Subplot 4
Softwood				
ME	0.005	0.003	0.132	0.281
RMSE	0.134	0.511	0.772	1.110
Hardwood				
ME	0.020	0.091	-0.081	-0.148
RMSE	0.163	0.698	0.501	0.744

Table 15. Average corrections made to the predicted temperatures in the case of Fig. 29 (K); conventional process.

Zone	Period 1	Period 2	Period 3
Softwood			
D_1	-0.018	-0.155	
D_2	-0.435	-1.065	
D_3	-0.454	-0.483	
D_4	0.666	0.901	
Hardwood			
D_1	-0.156	-0.198	-0.182
D_2	-1.062	-1.404	-1.669
D_3	-0.989	-1.168	-1.487
D_4	0.101	0.145	-0.169

Table 16. Mean and root mean squared errors between predicted and real-time modelled alkali concentrations after zones D_1 - D_4 ; See Fig. 33; conventional process.

Error	After D_1	After D_2	After D_3	After D_4
Softwood				
ME	-0.003	-0.008	-0.013	-0.015
RMSE	0.004	0.010	0.019	0.020
Hardwood				
ME	-0.004	-0.011	-0.017	-0.018
RMSE	0.004	0.013	0.018	0.019

and hardwood cooking about 20% of wood is dissolved in the impregnation vessel. This represents about 40% of the total wood dissolution, when total yield after

Table 17. Mean and root mean squared errors between modelled and measured alkali concentrations after zones D_3 and D_4 ; See subplot 2 in Fig. 31; conventional process.

Error	After D_3 , SW	After D_4 , SW	After D_3 , HW	After D_4 , HW
ME	0.007	0.026	-0.006	0.030
RMSE	0.019	0.036	0.030	0.044

Table 18. Mean and root mean squared differences between recalculated and originally modelled real-time Kappa number profiles; See Fig. 36; conventional process.

Difference	After D_1	After D_2	After D_3	After D_4	After D_5
Softwood					
ME	0.358	0.976	1.825	1.520	0.888
RMSE	0.786	2.190	4.412	3.957	2.784
Hardwood					
ME	-0.001	-0.023	-0.055	-0.101	-0.122
RMSE	0.575	1.423	2.126	1.621	1.527

cooking is about 50%; see Fig. 44 (40–60% was mentioned by Marcoccia *et al.* (2000) and Olm & Tistad (1979)).

In hardwood cooking, the relative dissolution of different wood components in the impregnation vessel and digester is similar as in softwood cooking. The use of original softwood parameters instead of optimised parameters in hardwood cooking, has only minor affect on real-time modelling results (See Fig. 39 vs. Fig. 41, Table 19 and Fig. 45.). However, errors are smaller with optimised hardwood parameters.

Differences between the digester's original real-time Kappa number profile and recalculated Kappa number profile are presented in Table 18; profiles are presented in Fig. 36. It can be seen, that the original real-time Kappa number profile is updated reasonably in most situations. The difference is biggest after a production rate change made during the first softwood cooking period; at around 2000 minutes in the examples. Errors between modelled and measured blow-line Kappa numbers (See Figs. 39 and 41.) are presented in Table 19 and Fig. 45.

Carbohydrates dissolve fastest in the initial phase, and the dissolution slows down in the bulk phase (Gullichsen 2000b); see also Figs. 3 and 4. The same can be seen from the results in Figs. 42 and 43; especially in the case of hemicellulose when half of the total dissolution happens during the short initial phase. Total yield is about 45–48% in softwood and about 52–53% in hardwood cooking (See Fig. 44). These values are similar to the values calculated in the plant based on monthly information about wood consumption and the amount of pulp produced.

Table 19. Mean and root mean squared errors between real-time modelled and measured blow-line Kappa numbers in the cases of Figs. 39 (HW with optimised parameters) and 41 (HW with original SW parameters); conventional process.

Error	All periods	Period 1	Period 2	Period 3
Fig. 39, softwood				
ME	0.020 ± 0.066	-0.146 ± 0.097	0.145 ± 0.090	
RMSE	1.825	1.756	1.876	
Fig. 39, hardwood				
ME	-0.026 ± 0.032	0.208 ± 0.058	-0.069 ± 0.041	-0.339 ± 0.091
RMSE	1.143	1.179	1.040	1.356
Fig. 41, hardwood				
ME	-0.132 ± 0.039	-0.035 ± 0.073	-0.047 ± 0.050	-0.583 ± 0.100
RMSE	1.390	1.462	1.283	1.556

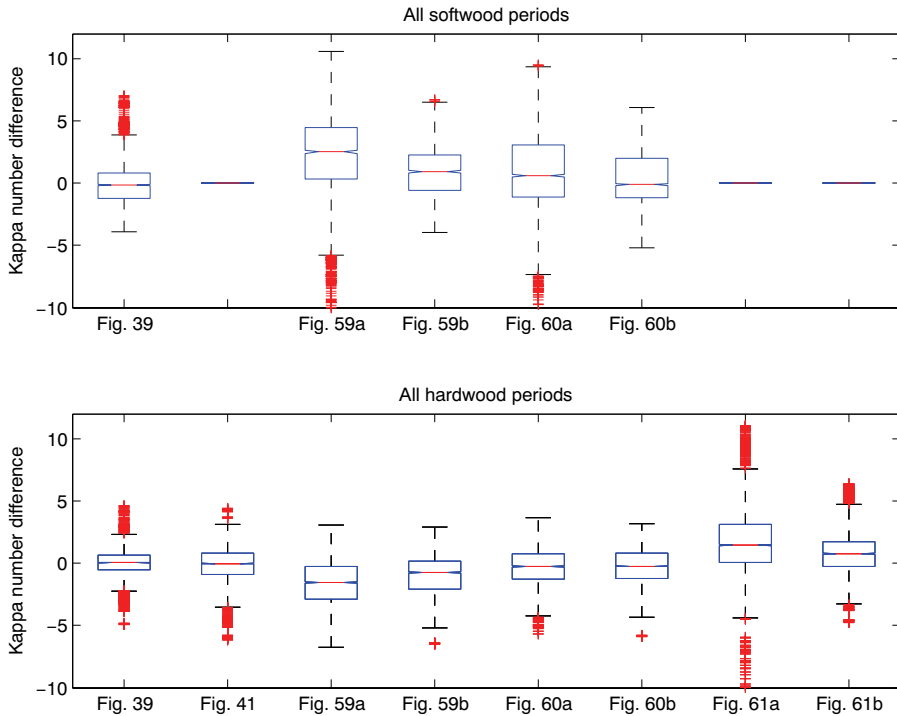


Fig. 45. Differences between modelled and measured blow-line Kappa numbers in conventional cooking. Each box has lines for lower and upper quartiles of data, red line for median, and notches for 95% confidence intervals about median. Whiskers extend the box 1.5 times the interquartile range away from the top or bottom of the box. Points beyond the whiskers are outliers.

6.3 Downflow Lo-SolidsTM cooking

In this section, construction of temperature and chemical profiles and modelling of real-time lignin and carbohydrate profiles of Downflow Lo-SolidsTM process are presented. The structure of the developed simulation model was presented in Section 5.3.

6.3.1 Temperatures

A real-time temperature profile of Downflow Lo-SolidsTM process was mainly based on the temperatures measured from the following points (See Fig. 2.): top of impregnation vessel (T1), middle part of impregnation vessel (T2), heated steam in digester (T4), upper extraction flow (T5), cooking circulation flow (T6) and lower extraction flow (T7). Also, temperatures of the digester's feed circulation flow (T3) and blow-line flow (T8) were considered. The values of temperatures were filtered either by calculating exponentially weighted moving averages (EWMA) or mean values of data. The mean values (calculation moment $\pm x$ minutes) were used in zones I_1 – D_1 and EWMA-filtered real-time measurements were used in zones D_2 – D_4 . Mean values were used to avoid the delay caused by filtering, so that the accuracy of the predicted blow-line Kappa number can be improved. In EWMA filtering, weighting factor 0.4 was used. The exothermic nature of delignification was considered and the filtered temperatures were suitably weighted to describe the chip column's temperature treatment when passing each zone. Temperatures in each zone were weighted as follows: I_1 (T1 50%, T2 50%), I_2 (T2 100%), D_1 (T4 100%), D_2 (T5 40%, T6 60%), D_3 (T6+4° 100%, when D_3 was modelled in one part) and D_4 (T7 100%). When zone D_3 was modelled in four parts, the temperature profile was supposed to be T6 plus 2 degrees raise of temperature in each of the subzones of D_3 . An example of the temperatures used in the modelling of each zone is shown in Fig. 46.

6.3.2 Chemical concentrations

A real-time alkali profile of the impregnation vessel was based on the impregnation vessel's feed circulation flow's effective alkali concentration, which was calculated from mass and flow balances. The alkali profile of the impregnation vessel was calculated as a function of alkali concentration before the impregnation vessel, and lignin and carbohydrate yields and liquor-to-wood ratio using Eq. 23. Impregnation of alkali from free liquor into the chips was also considered in the impregnation vessel's alkali profile. The alkali profile of the impregnation vessel was utilised in the modelling of carbohydrates degradation. Alkali concentrations before zones I_1 and I_2 were used in the modelling of carbohydrates degradation.

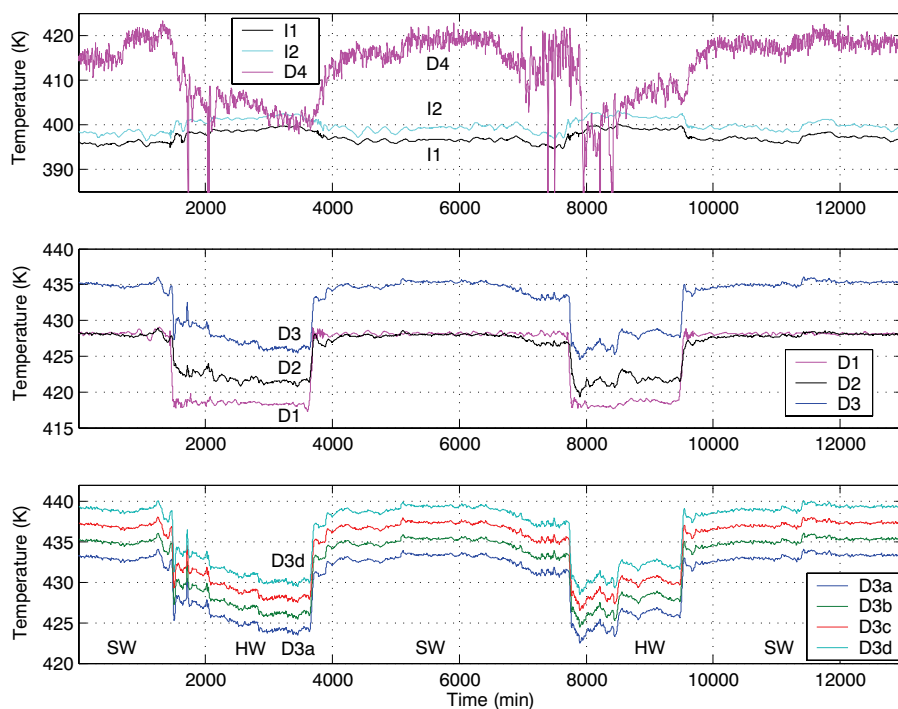


Fig. 46. Modelled temperature profile of Downflow Lo-SolidsTM process.

Digester's alkali profile was based on measured effective alkali concentrations of digester's feed circulation flow, upper extraction flow and cooking circulation flow. The alkali concentration of lower extraction flow was not used because of disturbances caused by the washing liquor. The average values of alkali concentrations before and after zones D_1 and D_2 were used in the modelling of these zones. The alkali profile in digester's zone D_3 was modelled as a function of lignin (See Fig. 51.) and carbohydrate (See Figs. 55 and 56.) yields and liquor-to-wood-ratio using Eq. 23, and measured alkali concentration of cooking circulation flow. The average values of alkali concentrations in D_3 and D_{3a} - D_{3d} were used in the modelling.

Examples of alkali profiles in the impregnation vessel and digester are shown in Figs. 47 and 48. In subplot 1 in Fig. 47, alkali concentrations before and after I_1 are calculated concentrations and alkali concentration after I_2 is measured. Alkali concentrations in D_1 and D_2 (subplot 2 in Fig. 47) are values used in the modelling. Fig. 48 presents measured alkali concentration before D_3 and calculated alkali concentrations after D_{3a} - D_{3d} .

The sulphidity of the white liquor is determined in laboratory once a week. Therefore, constant value 35% was used for sulphidity. It was assumed that sulfide ions are not consumed in the reactions.

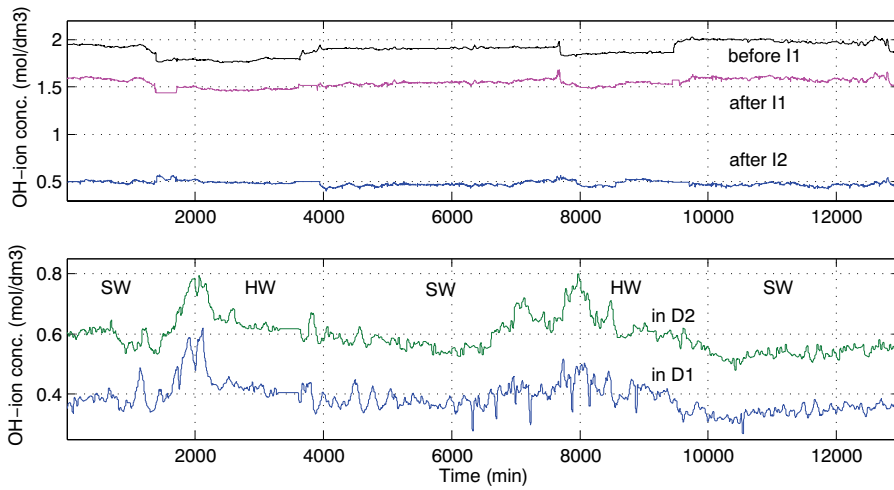


Fig. 47. Modelled alkali profile of Downflow Lo-SolidsTM process; zones I_1 – D_2 .

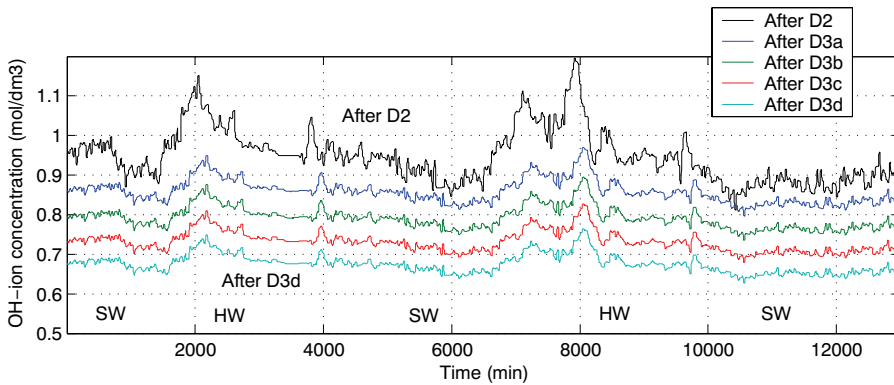


Fig. 48. Modelled alkali profile of zone D_3 in Downflow Lo-SolidsTM process.

6.3.3 Lignin

A real-time Kappa number profile of Downflow Lo-SolidsTM process was modelled using the optimised Gustafson's model as described in Sections 3.2 and 5.1. Temperature and alkali profiles were constructed as described in Sections 6.3.1 and 6.3.2. Residence times were calculated as a function of production rate target using residence times given by equipment supplier. Residence times in Downflow Lo-SolidsTM process during the example period are presented in Fig. 49.

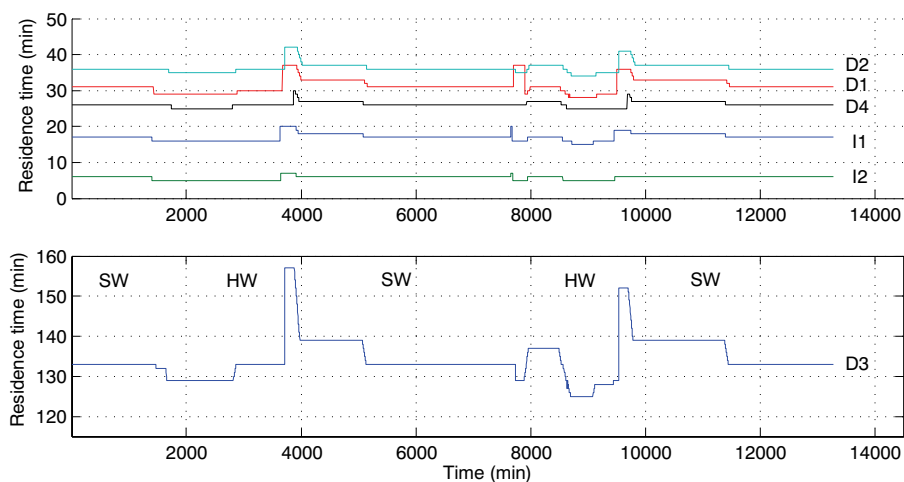


Fig. 49. Chips' residence times in impregnation vessel and digester (Downflow Lo-SolidsTM process).

For zones I_1 – D_2 , only real-time Kappa number profile was modelled. Model parameters of these zones were not updated on-line. Lignin yields in zones I_1 – D_2 are shown in Fig. 50.

The lignin yield in the cooking zone D_3 was modelled in two ways, namely considering D_3 as a one part or as four parts of same height (D_{3a} – D_{3d}). When calculating D_3 in one part, temperature and alkali values describing the average conditions in the whole zone were utilised. Both models of D_3 had separate yield factor parameters. Those parameters were updated on-line after each Kappa number measurement as a function of difference between measured and modelled Kappa numbers. The residence time of D_3 determined, how many previous Kappa numbers were compared. Delignification in zone D_3 was recalculated after each Kappa number measurement using 'one part' model with updated parameters. The residence time of D_3 determined how many previous calculations were recalculated.

Lignin yields, when modelling D_3 in one or four parts, are shown in subplot 1 in Fig. 51. The lignin yield in the washing zone D_4 was calculated when D_3 was modelled in one part. When D_3 was modelled in four parts, a constant value of lignin yield in D_4 was used. Calculated lignin yield in D_4 is shown in subplot 2 in Fig. 51. In the case of hardwood cooking, modelled blow-line Kappa number was restricted relative to Kappa number target, so that 25% of the deviation between predicted Kappa number and Kappa number target was allowed. Modelled and measured real-time blow-line Kappa numbers are shown in Figs. 52 (D_3 was modelled in one part) and 53 (D_3 was modelled in four parts). A real-time Kappa number profile in Downflow Lo-SolidsTM process, when D_3 was modelled in four parts, is presented in Fig. 54.

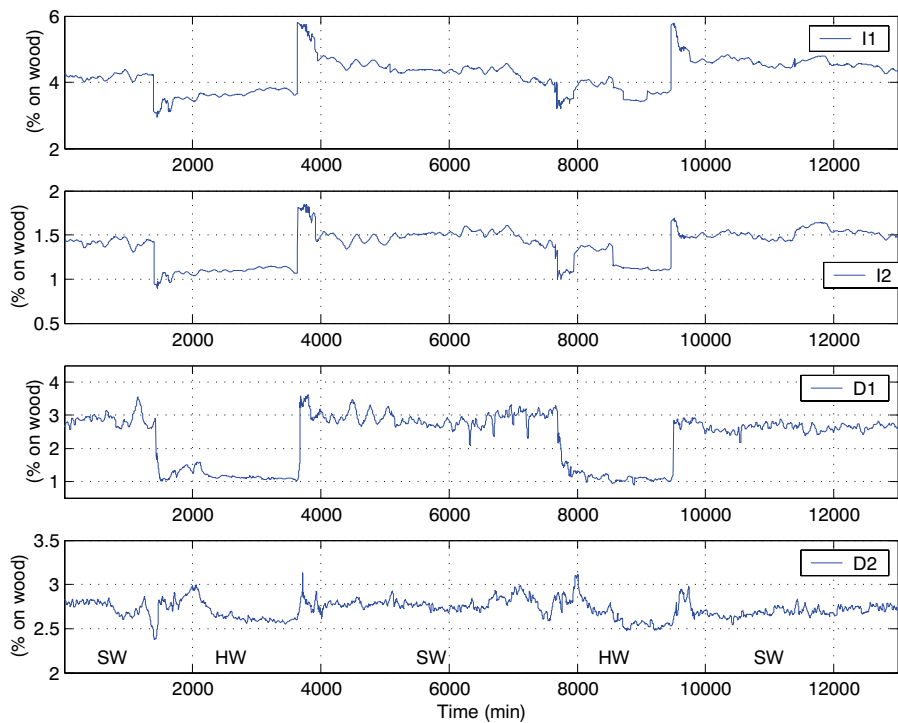


Fig. 50. Modelled lignin yields in zones I_1 – D_2 ; Downflow Lo-SolidsTM process.

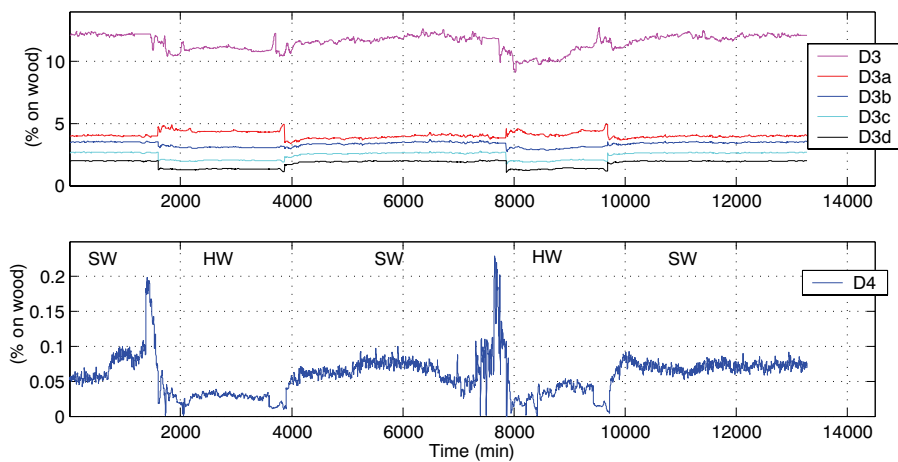


Fig. 51. Modelled lignin yields in zones D_3 and D_4 ; Downflow Lo-SolidsTM process.

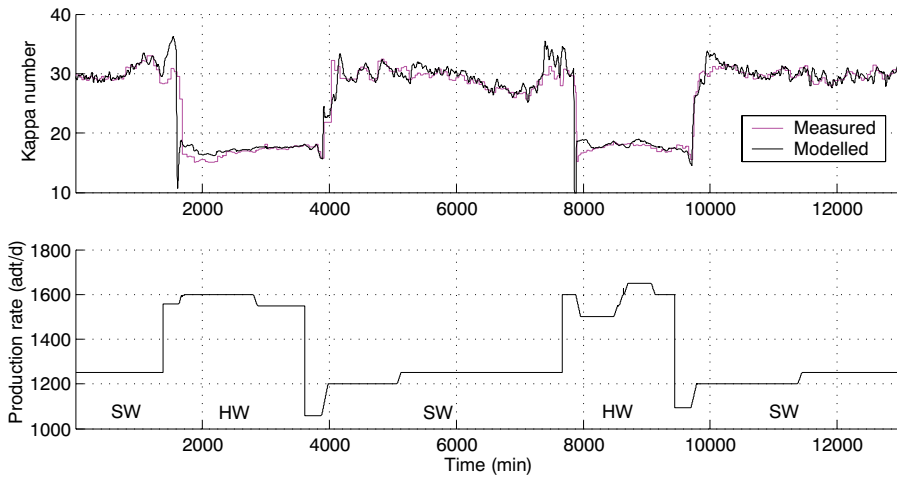


Fig. 52. Measured and modelled blow-line Kappa numbers and production rate, when D_3 is modelled in one part; Downflow Lo-SolidsTM process.

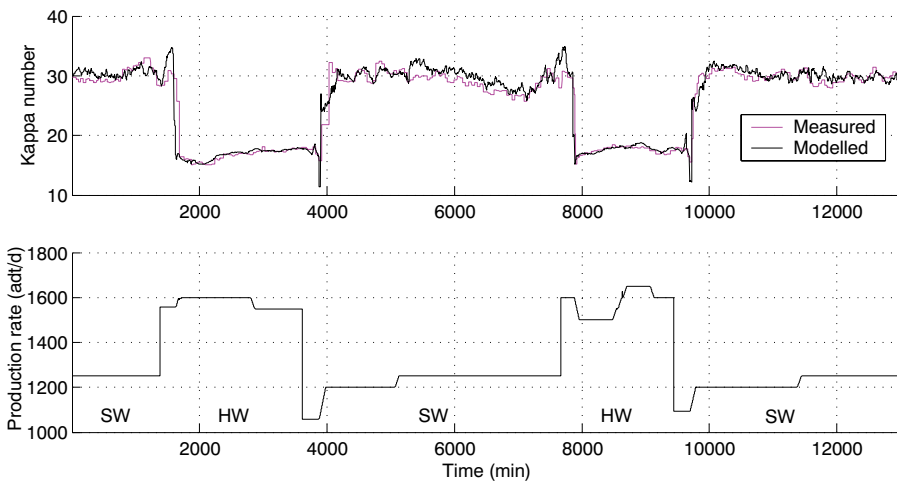


Fig. 53. Measured and modelled blow-line Kappa numbers and production rate, when D_3 is modelled in four parts; Downflow Lo-SolidsTM process.

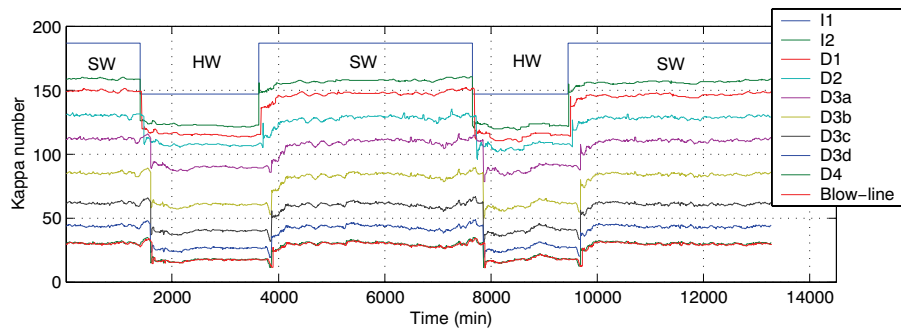


Fig. 54. Modelled real-time Kappa number profile in Downflow Lo-SolidsTM process; Kappa numbers before zones I_1 – D_4 and in blow-line.

6.3.4 Carbohydrates

In the impregnation vessel, total yield of carbohydrates (sum of cellulose and hemicellulose) was calculated using Eq. 15. In the digester, the yields of cellulose and hemicellulose were calculated separately using Eqs. 20 and 21. Other wood components, *i.e.* extractives, are usually removed during presteaming and early impregnation stages. In this study, they were assumed to be removed during the steaming.

Real-time cellulose and hemicellulose profiles are shown in Figs. 55 and 56. The total yield profile (the amount of lignin and carbohydrates left in the wood) is shown in Fig. 57.

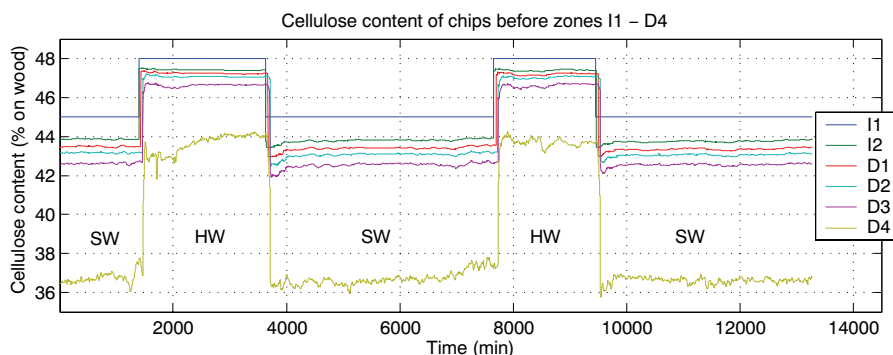


Fig. 55. Modelled real-time cellulose profile of Downflow Lo-SolidsTM process.

6.3.5 Discussion

The temperature profile of Downflow Lo-SolidsTM process was based on direct measurements from heated steam (T1, T4) and circulation flows (T3, T5, T6, T7, T8). Measurement T2 is near the wall of impregnation vessel. However, horizontal conditions in the impregnation vessel were considered to be steady enough, and it was important to have information also from the middle part of the impregnation vessel. Thus, T2 was also used. T3 was not used because of washing liquor addition to the bottom of impregnation vessel, and because T3 was measured from the return flow of digester's feed circulation. The temperature of cooking circulation flow (T6) was measured before a heat exchanger. Heated liquor flow was fed back to the digester via a central pipe. The temperature was raised on average 3 to 4 degrees in softwood cooking and less than 1 degree in hardwood cooking. Temperature T6 was used to describe the conditions in the cooking zone D_3 .

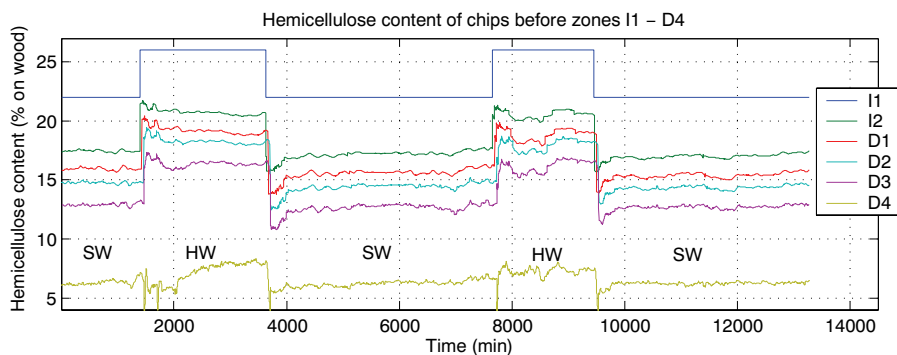


Fig. 56. Modelled real-time hemicellulose profile of Downflow Lo-SolidsTM process.

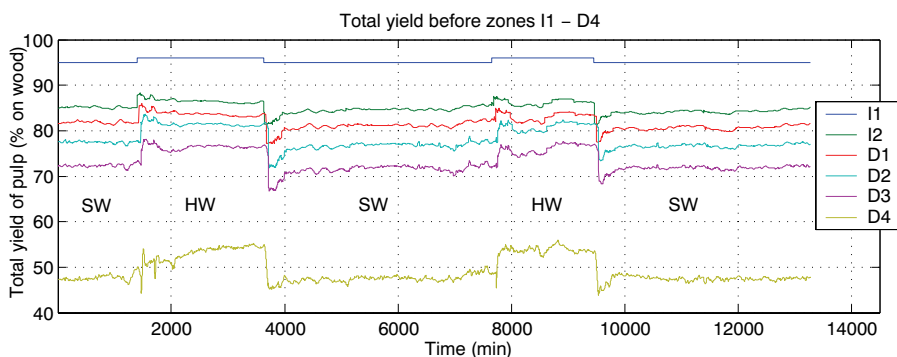


Fig. 57. Modelled real-time total yield profile of Downflow Lo-SolidsTM process.

Counter-current flow in the displacement heating zone (D_2) and liquor additions via central pipe, make the construction of temperature and alkali profiles challenging. Cooking circulation liquor, fed by a central pipe to the middle of digester above cooking circulation screens, heats up chip column before the cooking zone (D_3) and also upper extraction flow. The liquor added via the central pipe does not immediately distribute evenly in the chip column. Thus, the temperature and alkali concentration at the middle of chip column is higher than near digester wall. The extraction flow to the cooking circulation, taken via screens at the digester wall, evens the temperature and alkali profiles. It was assumed, that horizontal profiles even out at the upper part of cooking zone (D_3). However, it is likely that chips are not evenly cooked due to the liquor addition. In any case, it is difficult to say how much this affects the blow-line Kappa number distribution. Moreover, liquor addi-

tion via a central pipe is the only practical solution to carry out the temperature and alkali profiling in continuous cooking.

Alkali concentration of the impregnation vessel's feed circulation flow was not measured. Feed circulation flow's alkali concentration was calculated based on mass and flow balances and the white liquor's alkali concentration. Alkali concentration after the impregnation vessel was modelled based on alkali consumption caused by chemical reactions, and flow balance over the impregnation vessel. In this balance, also liquor impregnation from free liquor to the chips was estimated. Impregnation was estimated, because alkali consumption caused by chemical reactions did not explain the difference in alkali concentrations before and after the impregnation vessel. The idea in the modelling of impregnation was, that concentrations in the free liquor and inside the chips become even. The model based on alkali concentration after the impregnation vessel was found to be accurate.

To improve the chemical profiles, sulphidity of the white liquor should be measured continuously. Sulphidity of the white liquor was assumed to have constant value, and that sulfide ions are not consumed in the process. Liquor-to-wood ratios applied in the modelling of alkali consumption were calculated in the automation system. The accuracy of residence times could be improved by calculating them as a function of chip column's packing (like in the conventional process). Within this thesis that was not made.

In the modelling of Kappa number profile, the rate equation for the initial phase delignification (Eq. 10) was applied in the impregnation vessel. In the digester zones D_1 – D_3 , the rate equation for the bulk phase delignification (Eq. 11) was utilised. It is justified to use the bulk phase rate equation in the digester's zones D_1 (zone of extended co-current impregnation) and D_2 (counter-current washing zone), because the temperature at the top of digester is above 145°C (according to Marcoccia *et al.* (2000), bulk phase delignification starts when temperature is above 145–150°C). In softwood cooking, Kappa number, after the impregnation vessel is about 150 (equals to lignin content of about 22.5%), which is almost the transition content from the initial phase to bulk phase delignification (Olm & Tistad (1979); Rekunen *et al.* (1980)). Errors between modelled and measured blow-line Kappa numbers (See Figs. 52 and 53.) are presented in Table 20 and Fig. 58. The height of the calculation zone D_3 has only slight effect on the results. In softwood cooking, the modelling errors are smaller when D_3 is modelled in one part. In hardwood cooking, conversely results were achieved.

Models of cellulose and hemicellulose degradation were optimised, so that total yield was at a correct level. It was also ensured that unreactive contents of cellulose and hemicellulose were not degraded. The total yield of softwood cooking is known to be about 47% (information given from industry). A modelled yield of SW is in the range of 46–48% (See Fig. 57.). In hardwood cooking modelled total yield is about 51–56%.

Table 20. Mean and root mean squared errors between real-time modelled and measured blow-line Kappa numbers in the cases of Figs. 52 (D_3 is modelled in one part) and 53 (D_3 is modelled in four parts); Downflow Lo-Solids process.

Error	All periods	Period 1	Period 2	Period 3
Fig. 52, softwood				
ME	0.390 ± 0.026	0.463 ± 0.069	0.573 ± 0.042	0.134 ± 0.032
RMSE	1.252	1.440	1.408	0.905
Fig. 53, softwood				
ME	0.593 ± 0.025	0.450 ± 0.064	0.887 ± 0.040	0.311 ± 0.032
RMSE	1.295	1.341	1.512	0.941
Fig. 52, hardwood				
ME	0.396 ± 0.019	0.473 ± 0.025	0.299 ± 0.029	
RMSE	0.710	0.747	0.659	
Fig. 53, hardwood				
ME	0.117 ± 0.013	0.131 ± 0.016	0.099 ± 0.020	
RMSE	0.401	0.391	0.413	

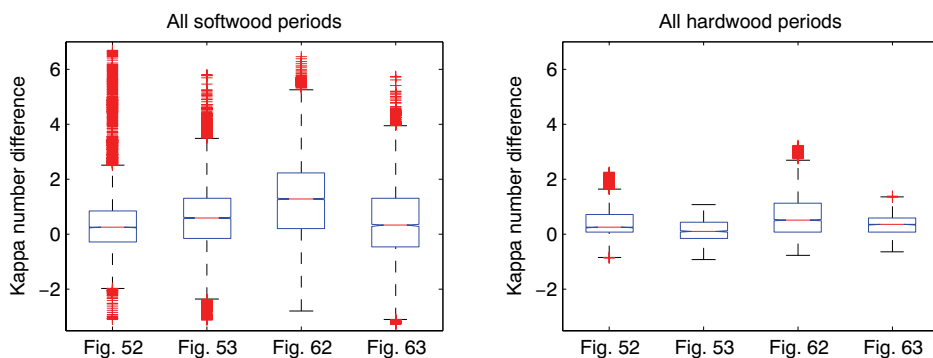


Fig. 58. Differences between modelled and measured blow-line Kappa numbers in Downflow Lo-SolidsTM cooking. Each box has lines for lower and upper quartiles of data, red line for median, and notches for 95% confidence intervals about median. Whiskers extend the box 1.5 times the interquartile range away from the top or bottom of the box. Red points beyond the whiskers are outliers.

6.4 Kappa number prediction

In this section, the Kappa number prediction in both conventional and Downflow Lo-SolidsTM cooking is presented and discussed. The structures of the prediction models were presented in Section 5.3.2.

6.4.1 Conventional cooking

In the case of the conventional cooking process, the blow-line Kappa number was predicted at the top of the digester, before the cooking zone. The structure of the prediction model is shown in Fig. 15 in p. 68. The main controlled variables in the process are the temperature of heated steam at the top of the digester ($T4$ in Fig. 1) and the alkali concentration of the digester's feed circulation flow ($A2$ in Fig. 1). These variables, $T4$ and $A2$, were also used as inputs for the prediction model. Thus, the predicted blow-line Kappa number can be utilised in the Kappa number control.

Lignin yields in zones D_1 – D_4 (See Figs. 34 in p. 88 (D_1 and D_2) and 35 in p. 89 (D_3 and D_4)) were calculated based on the predicted temperature (See Fig. 29 in p. 84.) and modelled alkali (See Fig. 33 in p. 86.) profile of the digester. Lignin yields in washing zones D_5 and D_6 were assumed to be constant, because in practice it is not possible to know process conditions in the washing zone in advance, *i.e.* before the cooking time. The predicted blow-line Kappa numbers were restricted relative to the Kappa number target, so that 50% of the deviation between the predicted Kappa number and the Kappa number target was allowed. The predicted and measured blow-line Kappa numbers are presented in Fig. 59.

From the predicted lignin content after zone D_4 was subtracted a mean value of the previous real-time lignin yields in the washing zone. The resulting Kappa number was compared with the measured blow-line Kappa number, to evaluate the accuracy of the prediction model (See Fig. 60.). In Fig. 61, the predicted blow-line Kappa number is presented, when original softwood parameters are used for hardwood. In the prediction models of Kappa number, the updated parameters of the real-time Kappa number model were used.

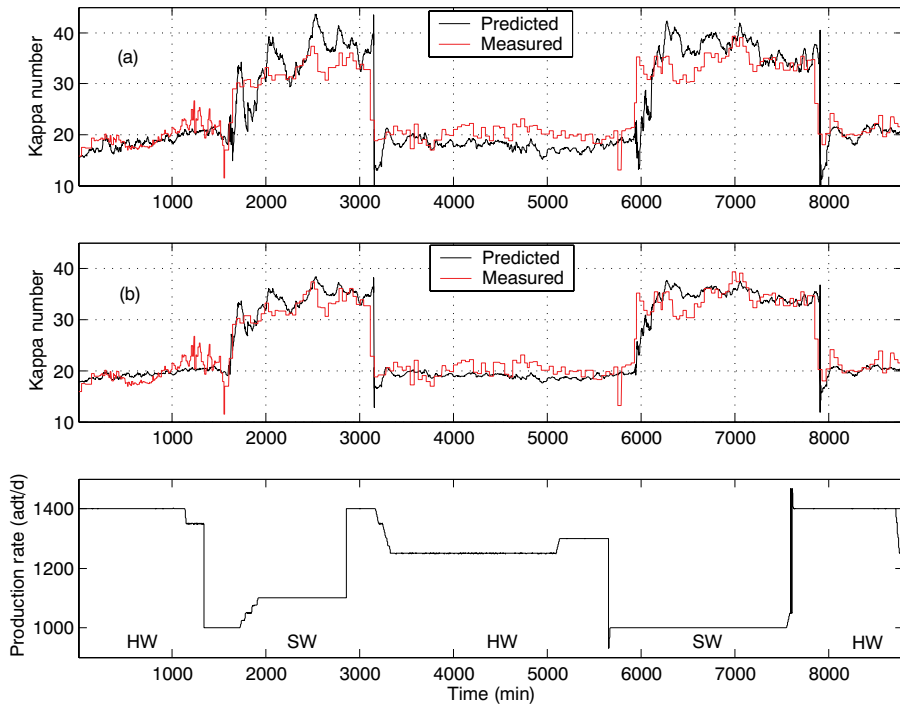


Fig. 59. Measured and predicted (constant lignin yield in washing zone) blow-line Kappa numbers (up: unrestricted; middle: restricted) and production rate in conventional cooking.

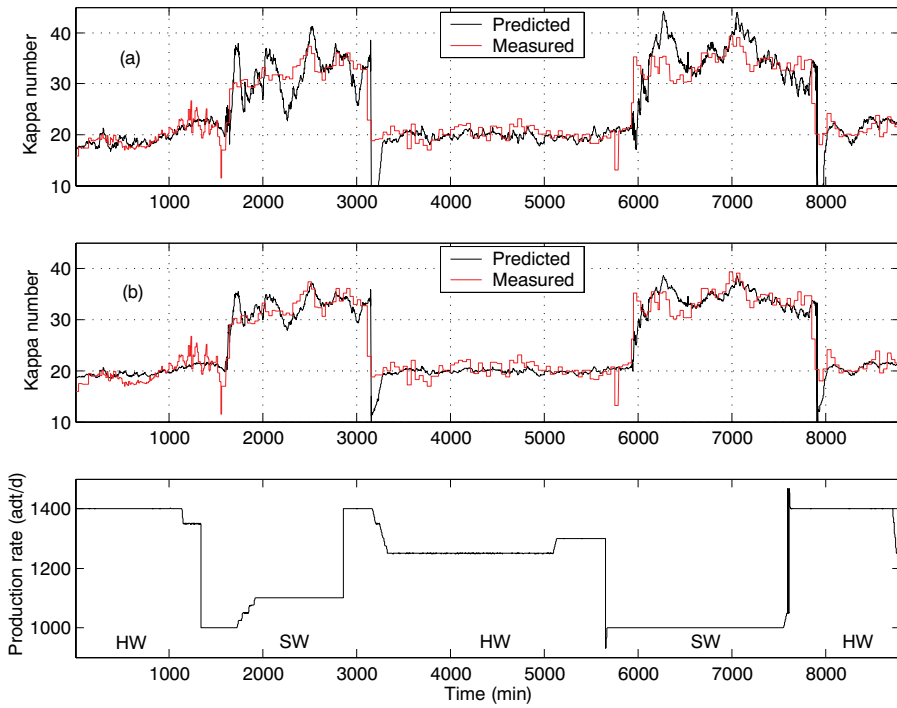


Fig. 60. Measured and predicted (real-time lignin yield in washing zone) blow-line Kappa numbers (up: unrestricted; middle: restricted) and production rate in conventional cooking.

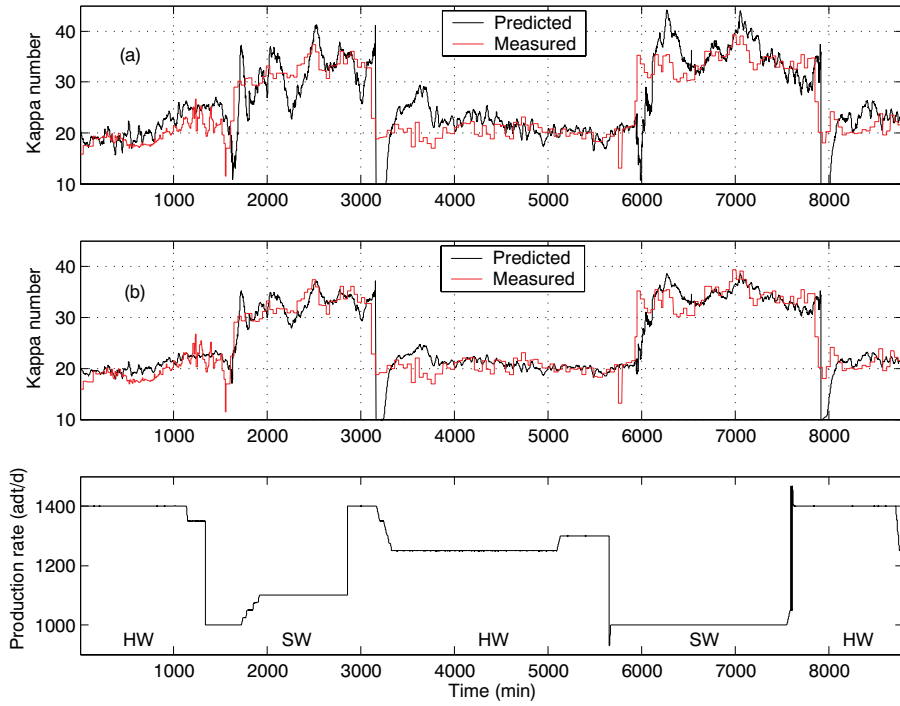


Fig. 61. Measured and predicted (real-time lignin yield in washing zone) blow-line Kappa numbers (up: unrestricted; middle: restricted) and production rate in conventional cooking; original softwood parameters for hardwood.

6.4.2 Downflow Lo-SolidsTM cooking

The structure of the prediction model in the case of the Downflow Lo-SolidsTM process, is presented in Fig. 16 in p. 69. The blow-line Kappa number was predicted at the beginning of the cooking zone D_3 . The beginning of zone D_3 is a logical prediction point, because the cooking is mainly controlled by adjusting the temperature and alkali concentration of the cooking circulation flow (T_6 and A_4 in Fig. 2). Thus, the predicted blow-line Kappa number can be utilised in the Kappa number control. Temperature and alkali profiles of zone D_3 , utilised in the prediction model, can be seen in Figs. 46 in p. 100 and 48 in p. 101.

The lignin yield in the washing zone D_4 was assumed to be constant (mean value of real-time modelling was utilised). When D_3 was modelled in one part, the updated parameters of the real-time model were used. When D_3 was modelled in four parts, the model parameters were updated as a function of difference between predicted and measured blow-line Kappa numbers. In the case of hardwood cooking, the predicted Kappa number was restricted relative to the Kappa number target, so that 25% of the deviation between the predicted Kappa number and the Kappa number target was allowed.

The results of the Kappa number prediction are shown in Figs. 62 (D_3 was calculated in one part) and 63 (D_3 was calculated in four parts).

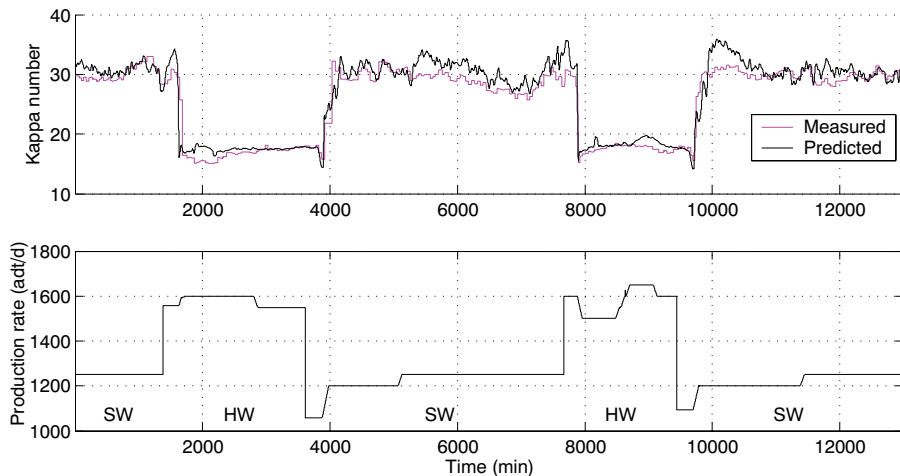


Fig. 62. Measured and predicted blow-line Kappa numbers and production rate in Downflow Lo-SolidsTM cooking, when D_3 is modelled in one part.

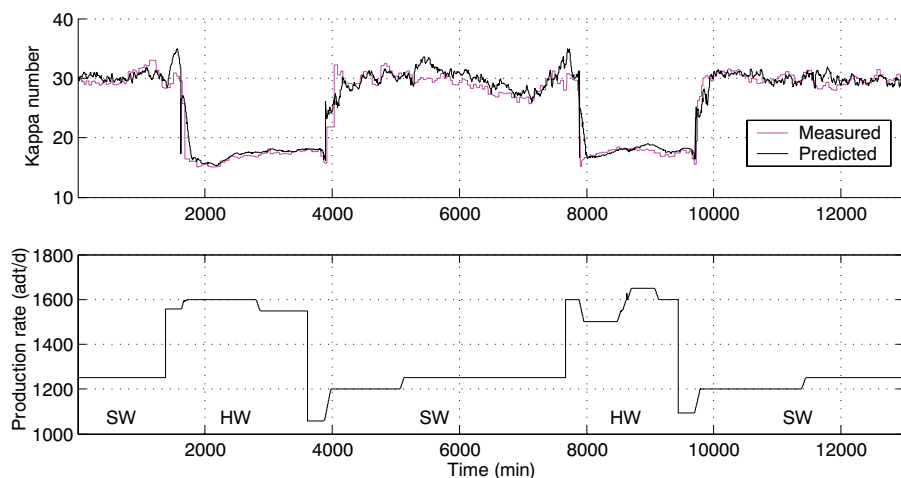


Fig. 63. Measured and predicted blow-line Kappa numbers and production rate in Downflow Lo-SolidsTM cooking, when D_3 is modelled in four parts.

6.4.3 Discussion

One purpose of the study has been to utilise the predicted blow-line Kappa numbers in the new control strategy (See Section 5.4.). This requires that the Kappa number prediction gives reliable and feasible information about the future blow-line Kappa numbers. There are several requirements for the feasibility of the prediction. The predicted and measured Kappa numbers must have good correlation, and no bias should be between them. It is also essential, how the Kappa number model functions over and during various disturbance situations, like grade and production rate changes.

In the conventional cooking, the predicted blow-line Kappa number has a relatively good correlation with the measured Kappa number in most situations. Changes in the Kappa number at the top of the digester can be seen in the Kappa number profile through the whole digester. This is obvious, because conditions of the chip column in the cooking zones are relatively steady, when the chip column moves downwards with co-current cooking liquor flow. The packing degree is low in the upper parts of the cooking zone, when chips are uncooked. The packing degree may change substantially in the lower parts of the cooking zone at the area of extractions.

Differences between predicted and real-time lignin yields in conventional process are presented in Table 21. Predicted and measured Kappa numbers are compared with Kappa number targets in Tables 23–28. The proportion of time, when both measured and predicted blow-line Kappa numbers are either bigger or smaller than the Kappa number target at the same time, gives information about prediction's

feasibility. Feasibility can be improved by updating the model, to ensure that the predicted Kappa number is at a same level than measured Kappa number.

Channelling of the cooking liquor may be a problem mainly in zones with counter-current flows. This may occur especially in continuous cooking with long counter-current washing zone (D_5 and D_6). The effects of the washing zone's phenomena on blow-line Kappa number can be seen by comparing modelling results in Figs. 59 and 60, see Table 22 and Fig. 45 (in p. 98). Errors are smaller in both grades, when the real-time lignin yields of zones D_5 and D_6 are used. In hardwood cooking, as was in the case of the real-time modelling, the modelling errors are bigger when original softwood parameters are used. In all cases, as was expected, the modelling errors are smaller when predicted blow-line Kappa numbers are restricted. When considering the Kappa number prediction, steady conditions in the washing zone should be ensured. The amount of the counter-current washing liquor is not considered in the modelling of the lignin yield in zones D_5 and D_6 . This information should be utilised, because the chip column's temperature profile depends on the washing factor.

In Downflow Lo-SolidsTM cooking, the washing zone is short and does not have a big effect on the Kappa number distribution of blow-line pulp. Other pulp properties are affected by the washing efficiency, however. In the counter-current displacement heating zone (D_2) of Downflow Lo-SolidsTM cooking, channelling is not a significant problem because uncooked chips have their original shape and packing degree of chip column is low. The modelling errors between predicted and measured low-line Kappa numbers in Downflow Lo-SolidsTM cooking are presented in Table 29 and Fig. 58 (in p. 109). The errors are smaller, in both softwood and hardwood cooking, when D_3 is modelled in four parts.

In the studied processes, Kappa numbers are measured on-line from the blow-line pulp. Pulp samples are analysed with kajaaniKAPPATM analysers based on optical measurement techniques. Values measured on-line may differ from the values determined in laboratories. In the laboratories, TAPPI T236 standard method, based on titration with potassium permanganate ($KMnO_4$), is used. Kappa number is calculated from the amount of $KMnO_4$ solution consumed by 1 gram of a dry pulp at 25°C during ten minutes. The relationship between the lignin content (% on wood) and Kappa number is expressed with a coefficient. Value 0.15 is used for the coefficient in this study, so that 1 Kappa number unit equals to a lignin content of 0.15% in wood (Ålander *et al.* 1963). The consumption of $KMnO_4$ depends on the reaction area of the pulp sample. Thus, a sample including uncooked material, *i.e.* having higher Kappa number, consumes less $KMnO_4$ than a sample of well disintegrated fibers. The values measured in the laboratory may be even several units lower than the on-line analyser's measurements. This may cause problems if an on-line analyser is calibrated based on the laboratory analyses. The on-line analyser's standard deviation per month, in a Kappa number range 20–40, is specified to be $1.0 + \sigma_L$ % of the maximum of the measuring range (σ_L is standard deviation of laboratory measurements). According to the information given from industry, σ_L can be assumed to vary between 1 and 2 Kappa number units. Thereby, the standard deviation of the measured Kappa number can be estimated to be about one unit in the example measurement range. In that case, 95% confidence intervals of the

measured Kappa number would be ± 1.96 units. In the most cases of real-time and prediction models, the modelling errors of the blow-line Kappa number are within these intervals.

Table 21. Mean and root mean squared differences between predicted and recalculated real-time modelled lignin yields after zones D_1 – D_4 ; See Figs. 34 in p. 88 and 35 in p. 89; conventional cooking.

Error	After D_1	After D_2	After D_3	After D_4
Softwood				
ME	-0.038	-0.071	-0.047	0.035
RMSE	0.109	0.229	0.423	0.158
Hardwood				
ME	0.035	0.046	-0.043	-0.035
RMSE	0.084	0.138	0.021	0.085

Table 22. Mean and root mean squared errors between predicted and measured blow-line Kappa numbers in the cases of Figs. 59 (constant yield in D_5 and D_6 , HW with optimised parameters), 60 (real-time yield in D_5 and D_6 , HW with optimised parameters) and 61 (real-time yield in D_5 and D_6 , HW with original SW parameters); conventional cooking; see also Fig. 45 in p. 98.

Error	Subplot 1, SW	Subplot 2, SW	Subplot 1, HW	Subplot 2, HW
Fig. 59				
ME	2.165 \pm 0.139	0.900 \pm 0.081	-1.549 \pm 0.048	-0.874 \pm 0.043
RMSE	4.400	2.402	2.299	1.763
Fig. 60				
ME	0.888 \pm 0.129	0.261 \pm 0.077	-0.217 \pm 0.042	-0.208 \pm 0.040
RMSE	3.642	2.128	1.516	1.437
Fig. 61				
ME	0.613 \pm 0.142	0.124 \pm 0.080	1.509 \pm 0.083	0.655 \pm 0.054
RMSE	3.942	2.207	3.318	2.0068

Table 23. Proportion of time when both measured and modelled Kappa numbers, at the same time, are bigger or smaller than Kappa number target in the case of Fig. 59 (constant yield in D_5 and D_6 , HW with optimised parameters), subplot 2; conventional cooking.

Grade	All periods	Period 1	Period 2	Period 3
Softwood	70.50%	70.26%	70.68%	
Hardwood	62.86%	86.07%	42.83%	80.65%

Table 24. Measured and predicted blow-line Kappa numbers vs. Kappa number target in the case of Fig. 59 (constant yield in D_5 and D_6 , HW with optimised parameters); mean values of differences; subplot 2; conventional cooking.

Comparison	All periods	Period 1	Period 2	Period 3
Measured-target, SW	0.427	-0.303	0.941	
Predicted-target, SW	1.184	0.831	1.453	
Measured-target, HW	0.301	-0.319	0.100	1.405
Predicted-target, HW	-0.532	-0.628	-0.979	0.575

Table 25. Proportion of time when both measured and modelled Kappa numbers, at the same time, are bigger or smaller than Kappa number target in the case of Fig. 60 (real-time yield in D_5 and D_6 , HW with optimised parameters), subplot 2; conventional cooking.

Grade	All periods	Period 1	Period 2	Period 3
Softwood	68.95%	65.55%	71.53%	
Hardwood	63.96%	88.74%	45.36%	74.28%

Table 26. Measured and predicted blow-line Kappa numbers vs. Kappa number target in the case of Fig. 60 (real-time yield in D_5 and D_6 , HW with optimised parameters); mean values of differences; subplot 2; conventional cooking.

Comparison	All periods	Period 1	Period 2	Period 3
Measured-target, SW	0.420	-0.325	0.945	
Predicted-target, SW	0.614	-0.164	1.205	
Measured-target, HW	0.293	-0.336	0.096	1.402
Predicted-target, HW	0.092	-0.159	-0.207	0.949

Table 27. Proportion of time when both measured and modelled Kappa numbers, at the same time, are bigger or smaller than Kappa number target in the case of Fig. 61 (real-time yield in D_5 and D_6 , HW with original SW parameters), subplot 2; conventional cooking.

Grade	All periods	Period 1	Period 2	Period 3
Softwood	69.54%	66.75%	71.65%	
Hardwood	63.40%	78.91%	53.59%	64.17%

Table 28. Measured and predicted blow-line Kappa numbers vs. Kappa number target in the case of Fig. 61 (real-time yield in D_5 and D_6 , HW with original SW parameters); mean values of differences; subplot 2; conventional cooking.

Comparison	All periods	Period 1	Period 2	Period 3
Measured-target, SW	0.420	-0.325	0.945	
Predicted-target, SW	0.492	-0.279	1.077	
Measured-target, HW	0.293	-0.336	0.096	1.402
Predicted-target, HW	0.901	0.724	0.694	1.291

Table 29. Mean and root mean squared errors between predicted and measured blow-line Kappa numbers in the cases of Figs. 62 (D_3 is modelled in one part) and 63 (D_3 is modelled in four parts); Downflow Lo-SolidsTM cooking; see also Fig. 58 in p. 109.

Error	All periods	Period 1	Period 2	Period 3
Fig. 62, softwood				
ME	1.260 ± 0.033	0.065 ± 0.069	1.630 ± 0.047	1.061 ± 0.056
RMSE	1.954	1.487	2.164	1.894
Fig. 63, softwood				
ME	0.465 ± 0.028	0.279 ± 0.067	0.840 ± 0.044	0.108 ± 0.035
RMSE	1.352	1.344	1.595	0.991
Fig. 62, hardwood				
ME	0.700 ± 0.026	0.703 ± 0.039	0.697 ± 0.033	
RMSE	1.064	1.140	0.962	
Fig. 63, hardwood				
ME	0.307 ± 0.014	0.374 ± 0.012	0.223 ± 0.027	
RMSE	0.531	0.466	0.603	

6.5 Kappa number control

In this section, the Kappa number control strategy, presented in Section 5.4, is applied to the conventional and Downflow Lo-SolidsTM cooking processes. The determination of the temperature and alkali set points in the conventional cooking, using digester's iteration model, is presented in Fig. 18 in p. 72. In Fig. 19 in p. 73, the determination of the temperature set point in the case of Downflow Lo-SolidsTM cooking is presented.

6.5.1 Conventional cooking

The main controlled variables in the studied conventional cooking process are the temperature of heated steam at the top of digester and alkali concentration of the digester's feed circulation flow. Either both temperature and alkali concentration, or only temperature was controlled by the new control strategy (See Section 5.4.). If only temperature was controlled, an alkali concentration of the digester's feed circulation flow was assumed to be as measured. The alkali charge would be controlled with liquor-to-wood and alkali-to-wood ratios, like in the present control strategy. Approach, when only temperature was controlled, does not differ from the usual case in the industry, when the temperature control strategy is based on the use of the H -factor. The digester's alkali profile was, however, modelled as a function of the controlled temperature. Thus, both the temperature and alkali profiles differed from the measured profiles also in that case.

In the new control strategy, cooking temperature and alkali concentration were controlled based on the difference between predicted (See Section 6.4.1.) and measured blow-line Kappa numbers. In Figs. 64 and 65, the original and controlled temperatures and alkali concentrations in zones D_1 – D_4 , when both temperature and alkali concentration are controlled, are shown. In Fig. 66, the target, predicted and controlled blow-line Kappa numbers, and the original and controlled temperatures of heated steam at the top of digester are shown, when both alkali and temperature are controlled. Predicted blow-line Kappa number in Fig. 66 is the same as in subplot 1 in Fig. 59. Number of required iteration rounds is shown in Fig. 67.

The results, when only temperature was controlled, are presented in Figs. 68–71. Results, when both alkali and temperature were controlled with original softwood parameters for hardwood, are presented in Figs. 72–75.

In the studied case, alkali concentration was changed $0.01 \frac{\text{mol}}{\text{dm}^3}$ ($0.310 \frac{\text{g}}{\text{dm}^3}$) in hardwood and $0.005 \frac{\text{mol}}{\text{dm}^3}$ ($0.155 \frac{\text{g}}{\text{dm}^3}$) in softwood for the difference of one Kappa number unit. Temperature was changed 0.27 K in hardwood and 0.22 K in softwood for the difference of one Kappa number unit.

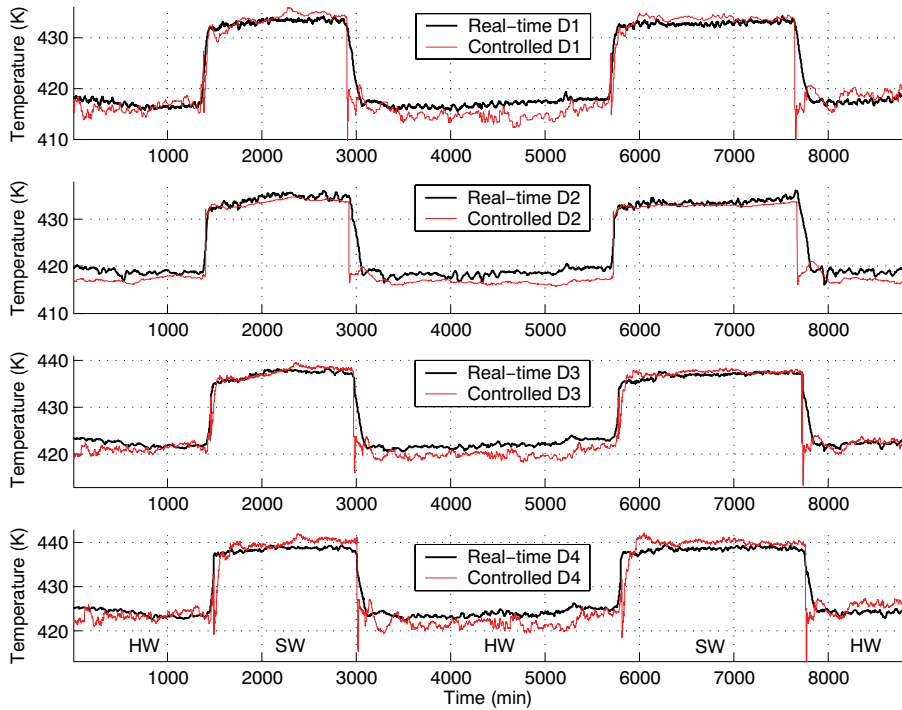


Fig. 64. Controlled and real-time modelled temperatures in zones D_1 – D_4 in conventional cooking, when both temperature and alkali concentration are controlled.

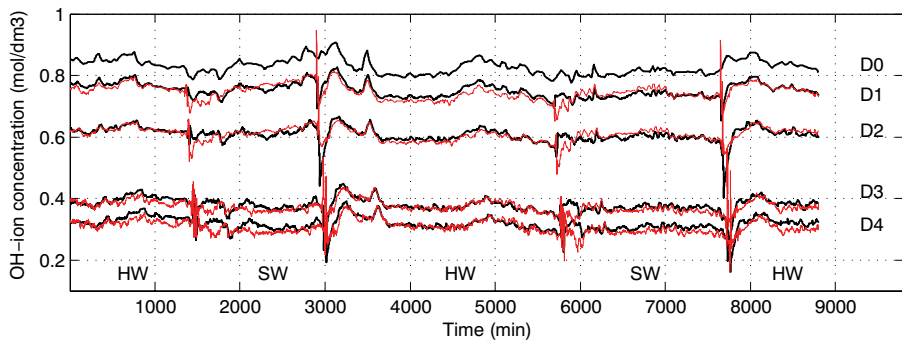


Fig. 65. Modelled alkali concentrations after zones D_1 – D_4 when both temperature and alkali concentration are controlled (red), compared with values of real-time modelling in the cooking zone (black); conventional cooking.

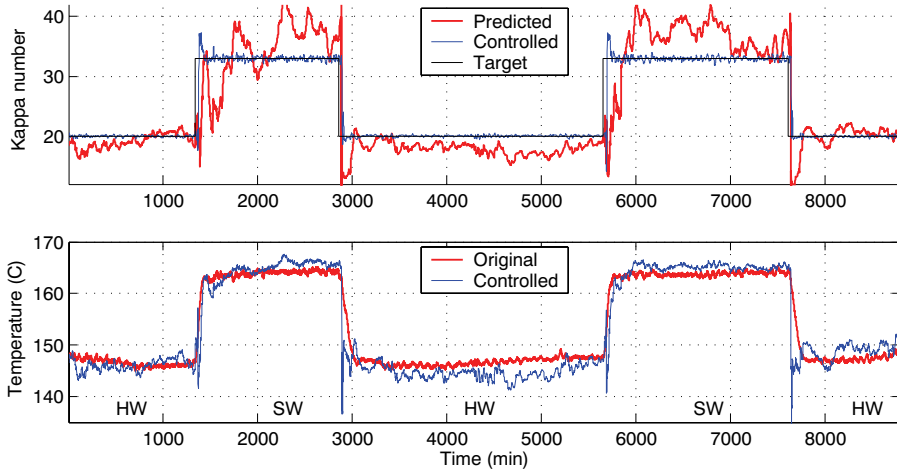


Fig. 66. Predicted, controlled and target blow-line Kappa numbers, and measured and controlled temperatures of heated steam in conventional cooking (both temperature and alkali concentration are controlled).

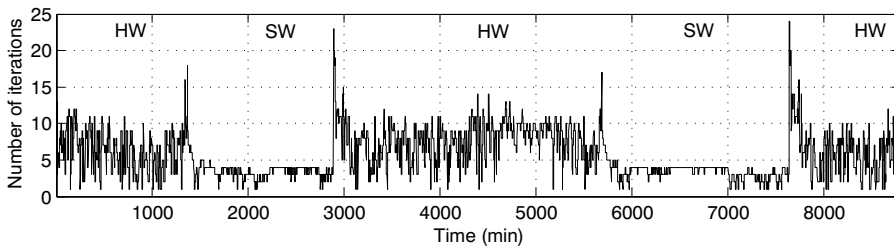


Fig. 67. Iteration rounds required in the case of conventional cooking in Fig. 66, on average 3.34 in SW and 6.82 in HW.

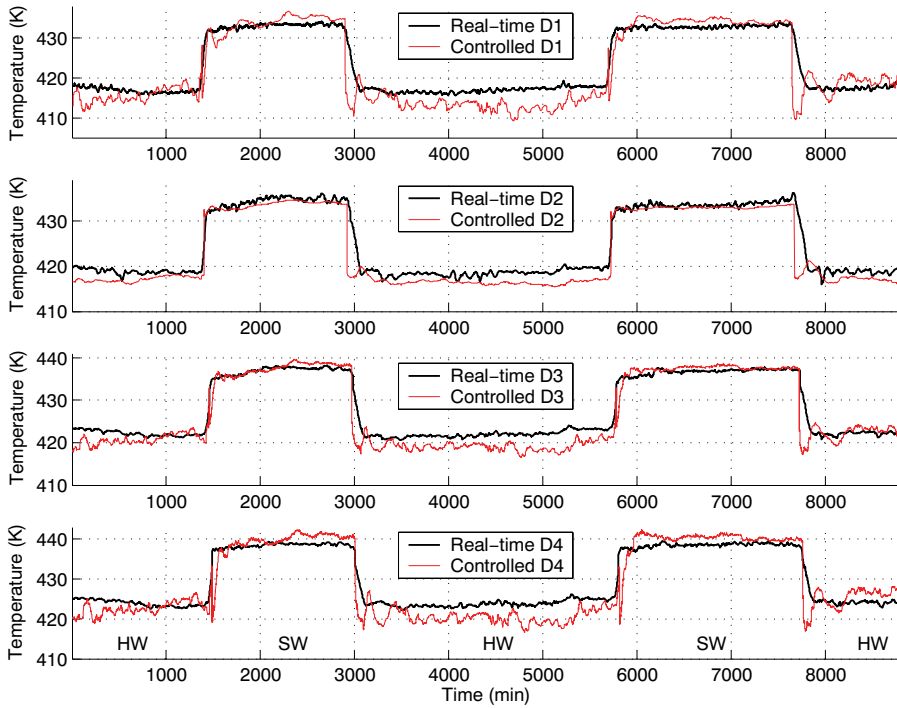


Fig. 68. Controlled and real-time modelled temperatures in zones D_1 – D_4 in conventional process (only temperature is controlled).

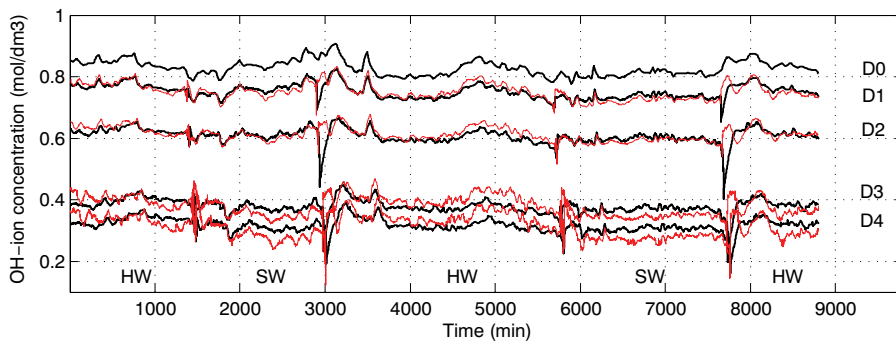


Fig. 69. Modelled alkali concentrations after zones D_1 – D_4 when only temperature is controlled (red), compared with values of real-time modelling in the cooking zone (black); conventional cooking.

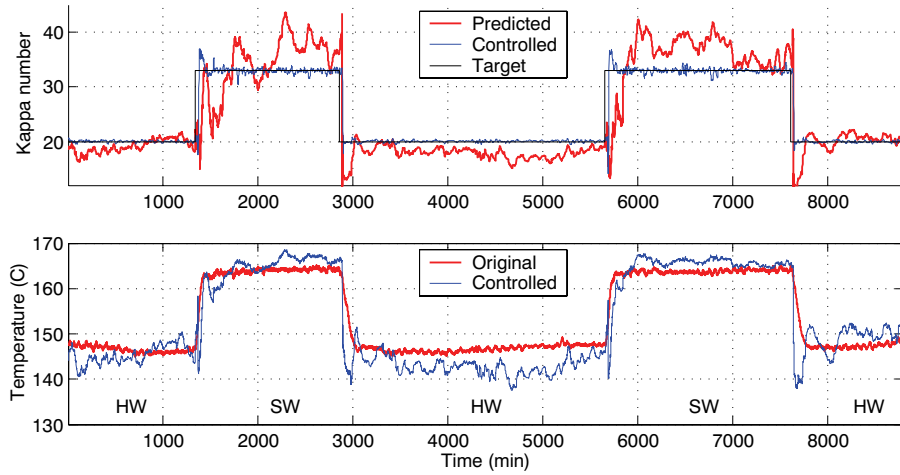


Fig. 70. Predicted, controlled and target blow-line Kappa numbers and measured and controlled temperatures of heated steam in conventional cooking (only temperature is controlled).

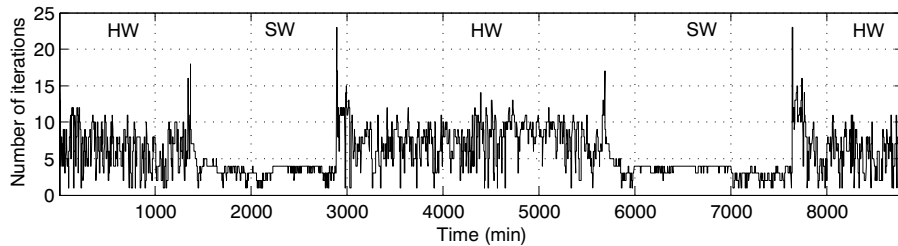


Fig. 71. Iteration rounds required in the case of conventional cooking in Fig. 70, on average 3.34 in SW and 6.84 in HW.

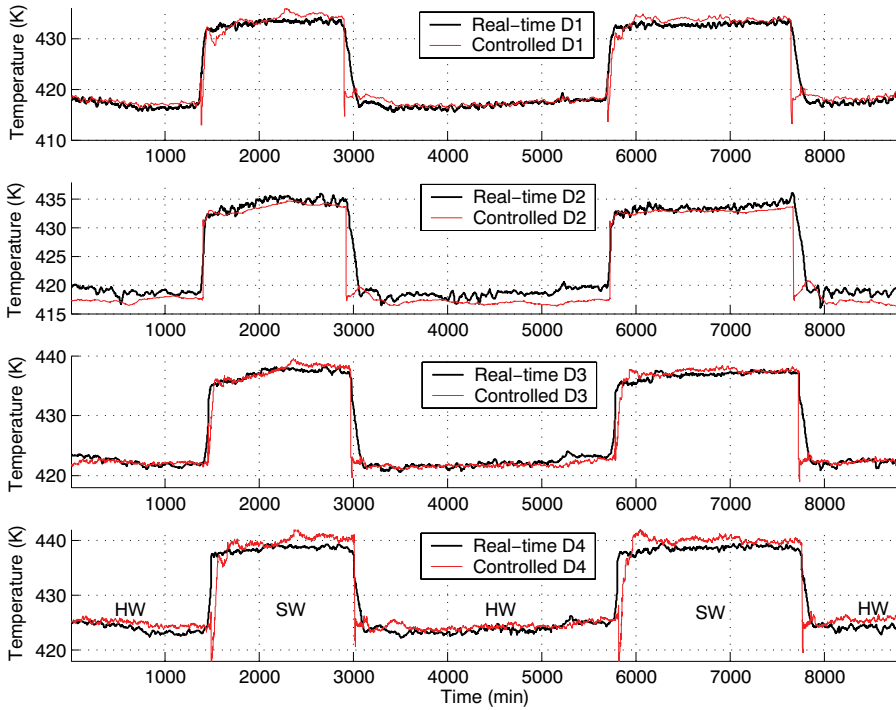


Fig. 72. Controlled and real-time modelled temperatures in zones D_1 – D_4 in conventional process (both temperature and alkali concentration are controlled); original softwood parameters for hardwood.

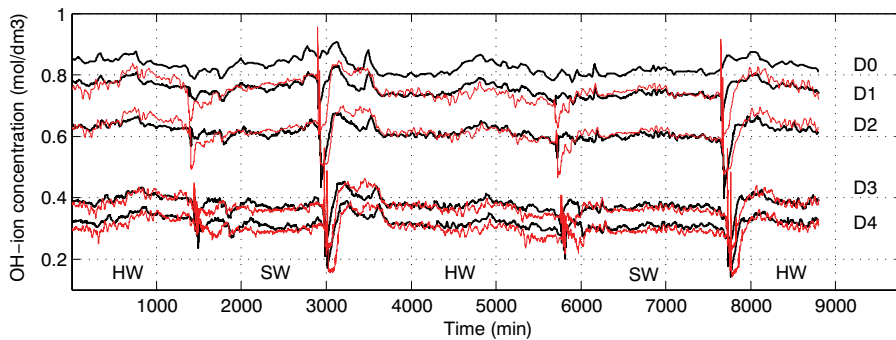


Fig. 73. Modelled alkali concentrations after zones D_1 – D_4 when both temperature and alkali concentration are controlled (red), compared with values of real-time modelling in the cooking zone (black); conventional cooking, original softwood parameters for hardwood.

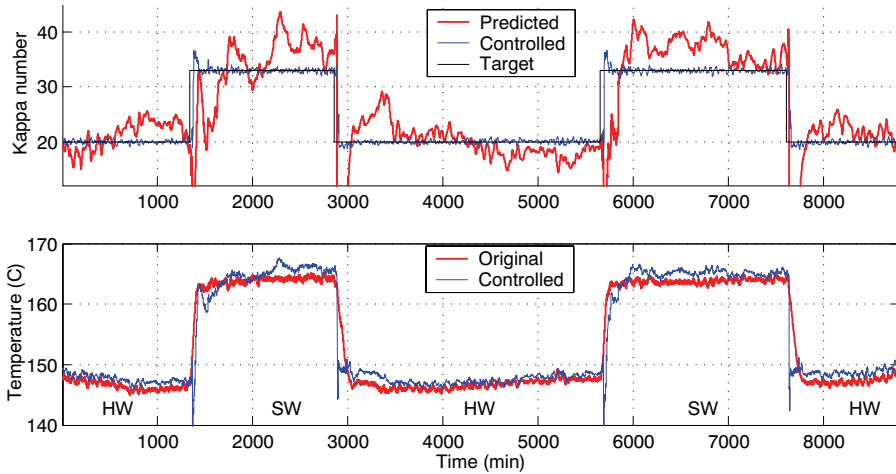


Fig. 74. Predicted, controlled and target blow-line Kappa numbers and measured and controlled temperatures of heated steam in conventional cooking (both temperature and alkali concentration are controlled); original softwood parameters for hardwood.

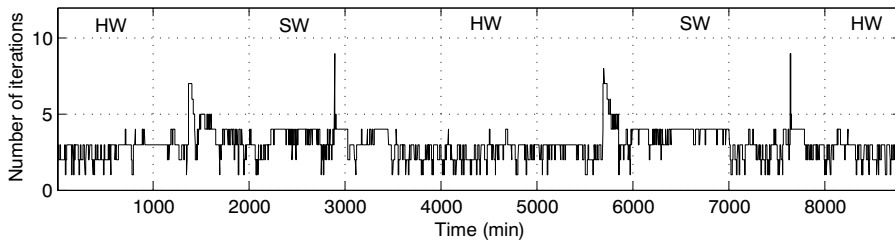


Fig. 75. Iteration rounds required in the case of Fig. 74 (on average 3.38 in SW and 2.68 in HW); conventional cooking, original softwood parameters for hardwood.

6.5.2 Downflow Lo-SolidsTM cooking

The cooking circulation flow's temperature and alkali concentration are the main controlled variables in the Kappa number control of the studied Downflow Lo-SolidsTM cooking process (See Fig. 2). The new control strategy was applied in the Downflow Lo-SolidsTM process by controlling the temperature of the cooking circulation flow. The lignin yields in zones D_2 and D_3 were modelled as a function of controlled temperature profile. Alkali concentration was modelled as a function of the controlled temperature and modelled alkali profiles in zones D_2 and D_3 . In Fig. 76, the predicted and controlled blow-line Kappa numbers and the correction of cooking circulation temperature, during an example period, are shown. The number of required iteration rounds is shown in Fig. 77.

The temperature was changed by 0.20 K for the difference of one Kappa number unit, when difference was smaller than 1 unit, otherwise temperature was changed by 0.16 K for the difference of one Kappa number unit.

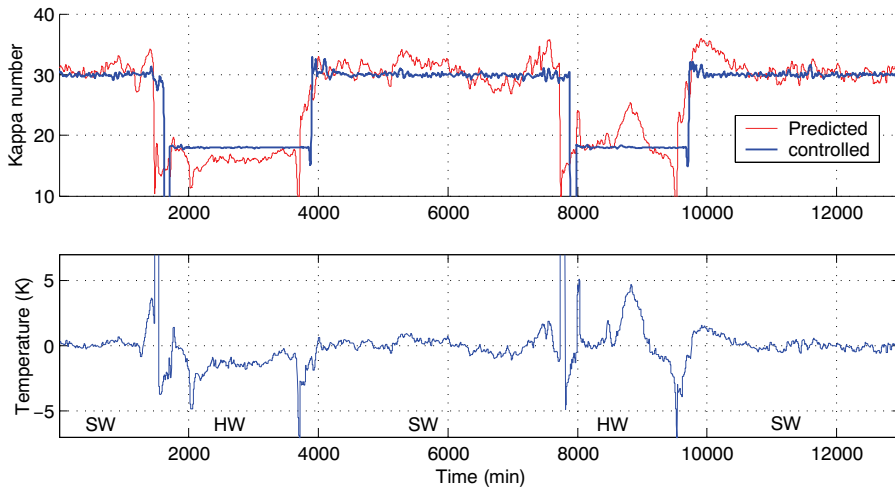


Fig. 76. Predicted, controlled and target blow-line Kappa numbers and correction of cooking circulation temperature in Downflow Lo-SolidsTM cooking.

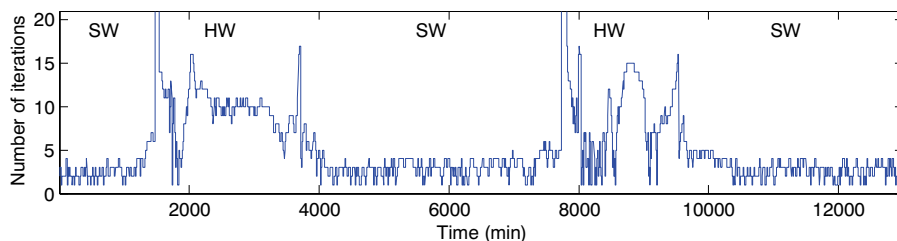


Fig. 77. Iteration rounds required in each iteration time in Downflow Lo-SolidsTM cooking (on average 4.22 in SW and 13.39 in HW).

6.5.3 Discussion

In the case of conventional cooking, the new control strategy was applied with and without the control of alkali charge. Determination of new set points for controlled variables was made in every 5 minutes, based on unrestricted blow-line Kappa numbers. Between iterations, constant corrections to the measured temperature and alkali concentration were made. Reason for the use of unrestricted Kappa numbers was, that the iteration model was not configured for the use of restricted Kappa numbers. Mean and root mean squared differences between real-time and controlled temperatures in zones D_1 – D_4 in the cases of Figs. 64, 68 and 72 are shown in Table 30. Mean and root mean squared differences between controlled and real-time modelled alkali concentrations after zones D_1 – D_4 in the cases of Figs. 65, 69 and 73 are shown in Table 31. Mean and root mean squared errors between controlled and target blow-line Kappa numbers in the cases of Figs. 66, 70, 74 and 76 are shown in Table 32.

Results, when both temperature and alkali concentration are controlled, are presented in Figs. 64–67. Required temperature set point changes are reasonable, when also alkali charge was controlled. Results, when only temperature was controlled, are presented in Figs. 68–71.

Results, when original softwood parameters were used for hardwood cooking (both temperature and alkali concentration were controlled), are presented in Figs. 72–75. The blow-line Kappa number was predicted and controlled with original softwood parameters for hardwood also, because unrestricted blow-line Kappa numbers were used, and because temperature dependence with optimised hardwood parameters was decreased. If optimised hardwood parameters were used, the required temperature set point changes had been unrealistically great (See especially subplot 2 in Fig. 70, but also subplot 2 in Fig. 66.). This also indicates, that it is essentially important to have the predicted blow-line Kappa number at a correct level by updating the prediction model.

In conventional cooking, usually less than 15 iteration rounds were needed, to achieve smaller than 0.1 difference between the predicted and target blow-line Kappa numbers. Over the example period, including grade changes, on average 5.62 iter-

ation rounds were needed, when both temperature and alkali were controlled (See Fig. 66; iteration rounds required at each iteration time are shown in Fig. 67). More iteration rounds were required in hardwood cooking, because temperature dependence is smaller, and the risk of divergence in the iteration was minimised.

In Downflow Lo-SolidsTM cooking, there are several possibilities to make corrections to the Kappa number profile, when prediction point of blow-line Kappa number is before cooking zone D_3 . The control strategy, used in the industry, is to control the alkali concentration and temperature of cooking circulation flow. The disadvantage of this control strategy is that it also affects the conditions of chip column in zone D_2 . However, residence time in zone D_3 is much longer than residence time in zone D_2 (120–150 vs. 35–40 minutes, see Fig. 49 in p. 102), and lignin is mainly dissolved in zone D_3 . In any case, Kappa number profiles of both zones D_2 and D_3 should be considered, when making control actions to the cooking circulation flow.

Another possibility to affect on the blow-line Kappa number is to change the residence time in zone D_3 . This could be handled by controlling the rate of blow-line flow. The rate of blow-line flow is, however, changed only in exceptional situations, like if the digester's packing degree is too high. Namely, it is essentially important to maintain as steady movement of chip column as possible to ensure uniform quality of the pulp. This strategy would also change the residence times in other zones, and the changes in the packing degree profile of the whole digester would be difficult to predict and handle.

Conditions in the washing zone D_4 also affect on the quality of blow-line pulp. Pulp is washed and cooking reactions are stopped by adding cold washing liquor to the bottom of digester. The washing liquor is fed via a central pipe slightly above the lower extraction screens. The counter-current washing liquor is fed via nozzles at the bottom of digester. Because the temperatures of washing liquors are low (under 100°C), and residence time in D_4 is only 20–30 minutes, in practice the blow-line Kappa number has to be affected before the chip column reaches the washing zone.

The results of the Kappa number control in Downflow Lo-SolidsTM cooking, are presented in Figs. 76 and 77.

In the case of Downflow Lo-SolidsTM cooking, the new control strategy was applied so that only the temperature of cooking circulation flow was controlled. Determination of a new temperature set point was made in every 10 minutes. Between iterations, constant correction to the measured temperature was made. In Downflow Lo-SolidsTM cooking, usually less than 10 iteration rounds were needed in softwood, and less than 20 in hardwood, when smaller than 0.1 difference between the predicted and target Kappa numbers was achieved in the iteration. Iteration rounds required in each iteration time are shown in Fig. 77. In the case of Fig. 76, an average 4.22 iterations were required in softwood cooking and 13.39 iteration rounds were required in hardwood cooking. Over the whole example period, including grade changes, an average 6.7 iteration rounds were needed.

Table 30. Mean and root mean squared differences between controlled and real-time modelled temperatures in zones D_1 – D_4 in the cases of Figs. 64 (temperature and alkali control, HW with optimised parameters), 68 (only temperature control, HW with optimised parameters) and 72 (temperature and alkali control, HW with original SW parameters); conventional process.

Difference	Subplot 1	Subplot 2	Subplot 3	Subplot 4
Fig. 64, softwood				
ME	0.696	-0.625	0.454	1.126
RMSE	1.154	0.843	0.787	1.837
Fig. 68, softwood				
ME	1.007	-0.643	0.575	1.305
RMSE	1.751	0.858	0.920	2.183
Fig. 64, hardwood				
ME	-1.046	-1.736	-1.305	-0.998
RMSE	1.936	1.960	1.858	2.139
Fig. 68, hardwood				
ME	-1.927	-1.852	-1.828	-1.803
RMSE	3.245	2.098	2.630	3.275
Fig. 72, hardwood				
ME	0.530	-1.453	-0.141	0.761
RMSE	0.733	1.687	0.624	1.098

Table 31. Mean and root mean squared differences between controlled and real-time modelled alkali concentrations after zones D_1 - D_4 in the cases of Figs. 65 (temperature and alkali control, HW with optimised parameters), 69 (only temperature control, HW with optimised parameters) and 73 (temperature and alkali control, HW with original SW parameters); conventional process.

Difference	After D_1	After D_2	After D_3	After D_4
Fig. 65, softwood				
ME	0.004	0.005	-0.009	-0.015
RMSE	0.015	0.016	0.017	0.021
Fig. 69, softwood				
ME	-0.009	-0.006	-0.019	-0.024
RMSE	0.012	0.009	0.023	0.030
Fig. 65, hardwood				
ME	-0.011	-0.004	-0.005	-0.009
RMSE	0.013	0.009	0.011	0.015
Fig. 69, hardwood				
ME	0.003	0.009	0.009	0.006
RMSE	0.011	0.015	0.026	0.028
Fig. 73, hardwood				
ME	0.001	0.009	-0.006	-0.014
RMSE	0.024	0.022	0.020	0.023

Table 32. Mean and root mean squared errors between controlled and target blow-line Kappa numbers in the cases of Figs. 66 (temperature and alkali control, HW with optimised parameters), 70 (only temperature control, HW with optimised parameters) and 74 (temperature and alkali control, HW with original SW parameters) in conventional cooking and Fig. 76 in Downflow Lo-SolidsTM cooking.

Error	Fig. 66	Fig. 70	Fig. 74	Fig. 76
Softwood				
ME, SW	-0.025±0.013	-0.022±0.019	-0.025±0.014	-0.023±0.007
RMSE, SW	0.369	0.515	0.380	0.320
Hardwood				
ME, HW	-0.001±0.019	0.047±0.006	-0.072±0.022	0.005±0.010
RMSE, HW	0.669	0.203	0.765	0.094

7 Conclusions and future work

Conventional and Downflow Lo-SolidsTM cooking processes have been under investigation in this study. Motivation for the research arised from the industry's interest and needs concerning continuous kraft pulping. The purpose of the research has been to improve the knowledge of the processes, and to achieve supporting tools for the process operators and control purposes. It was assumed, that the cooking conditions could be described with relatively simple kinetic models, by optimising and on-line updating the model parameters with the information gained from the process data.

Real-time simulation models for temperature, chemical concentrations, and chips' lignin and carbohydrates contents were developed. A Kappa number profile, giving information about digester's packing conditions, was utilised in the modelling of residence times. Based on the real-time models, the Kappa number profile and blow-line Kappa number were predicted in both conventional and Downflow Lo-SolidsTM cooking processes. The predicted blow-line Kappa numbers were used as inputs to a new Kappa number control strategy. In the developed strategy, set points for alkali charge and cooking temperature were iteratively solved. Based on the developed simulation model and control strategy, a cooking control application is being constructed and experimented on in a pulping plant.

In the investigated cooking processes, both softwood and hardwood pulp were produced. Grade change situations, occuring frequently, have a significant economical importance. Non-idealistic process conditions during the grade changes set demands for the process control and modelling. In this study, both grades were modelled using basicly the same models, so that only model parameters were adjusted. Temperature is perhaps the most critical variable in the modelling of the delignification and carbohydrates degradation. Thus, the adjustment of the model parameters was accomplished as a function of a temperature change.

The findings and developed models presented in this thesis can provide new, valuable information about the conditions and phenomena in the continuous cooking processes. The models of cooking degree and chemical consumption were established and proven feasible, and they can thus support the operators' work. The results indicate, that the usability of the kinetic models can be improved by on-line updating the model parameters. Immeasurable disturbances, for example in the

quality of raw materials, can be successfully considered in this way. The results suggest, that the modelling over grade change situations can be accomplished by smoothly adjusting the model parameters as a function of the temperature change profiles. The main advantage of the new control strategy is that the chemical profile of the cooking can be more precisely taken into account. That is the main difference compared to the H -factor based control strategies widely used in the industry.

In most situations, reasonable updating of the model parameters was required. However, the updated parameters may considerably differ from the optimised ones. Therefore, it would be essentially important to have more information about the front-end process conditions and the properties of the raw material. The on-line updating and smooth adjusting of the parameters seemed to be feasible, when the optimised, species specific parameters were utilised. However, changes to the model structures should be considered, especially in hardwood cooking, to enhance the validity of the real-time models and the predictability of the Kappa number. Also, more information should be available about the heats of reactions. The presented modelling procedures could be relatively easily applied to other continuous pulping processes. Nevertheless, the optimisation of the species and process specific model parameters, would apparently be necessary. Within this thesis, functionality of the new Kappa number control strategy was tested off-line. According to the simulations, the structure of the control strategy seemed to be feasible. However, further improvements and changes to the prediction models would be beneficial.

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