Simulation of flat falling film evaporator system for concentration of black liquor

R. Bhargava[a], S. Khanam[b,]*, B. Mohanty[a] and A. K. Ray[c]

[a] Department of Chemical Engineering, Indian Institute of Technology Roorkee,

Roorkee – 247 667, India

[b] Department of Chemical Engineering, National Institute of Technology Rourkela,

Rourkela – 769 008, India

[c] Department of Paper Technology, Indian Institute of Technology Roorkee,

Roorkee – 247 667, India

---

* Corresponding author:
E-mail address: shabinahai@gmail.com, skhanam@nitrkl.ac.in
Phone No. +91-9938185505, +91-661-2462267

Abstract

In the present investigation, a nonlinear mathematical model is developed for the analysis of Septuple effect flat falling film evaporator (SEFFFE) system used for concentrating weak black liquor in a nearby paper mill. This model is capable of simulating process of evaporation considering variations in boiling point rise (τ), overall heat transfer coefficient (U), heat loss from evaporator (Q_{loss}), flow sequences, liquor/steam splitting, feed, product and condensate flashing, vapor bleeding and physico-thermal properties of the liquor. Based on mass and energy balance around an effect a cubic polynomial is developed, which is solved repeatedly in a predetermined sequence using generalized cascade algorithm.

For development of empirical correlations for τ, U of flat falling film evaporators and Q_{loss}, plant data have been collected from SEFFFE system. These correlations compute τ, U and Q_{loss} within average absolute errors of 2.4%, 10% and 33%, respectively, when their results are compared with the plant data.

Keywords: Nonlinear model, Flat falling film evaporator, Empirical correlations, Boiling point rise, Overall heat transfer coefficient, Heat Loss

1. Introduction

Evaporators are integral part of a number of process industries namely Pulp and Paper, Chlor-alkali, Sugar, Pharmaceuticals, Desalination, Dairy and Food processing, etc. The Pulp and Paper industry, which is the focus of the present investigation, predominantly uses the Kraft Process in which black liquor is generated as spent liquor. This liquor is concentrated in multiple effect evaporator (MEE) house for further processing. Earlier, long tube vertical (LTV) type of evaporators were employed in India (Bhargava, 2004). However, with development of flat falling film evaporators (FFFE), which claim many benefits over its counter parts LTV evaporators, most Indian Paper Mills have already switched to FFFE systems. In fact, it operates under low temperature drop (about 5°C) across the film and thus, more evaporators can be accommodated within the total temperature difference available (T_S-T_{Le}) for evaporation to offer higher steam economy.

Rao and Kumar (1985) pointed out that the MEE house of Indian Paper mills alone consumes around 24-30% of the total steam required in a large paper mill. Therefore, it calls for a thorough investigation into its analysis and various energy reduction schemes.

For the analysis of MEE system mathematical models have been reported in the literature since last seven decades. A few of these were developed by Kern (1950), Itahara and Stiel (1966), Holland (1975), Radovic et al. (1979), Nishitani and Kunugita (1979), Lambert et al. (1987), Mathur (1992) and El-Dessouky et al. (1998, 2000), Costa and Enrique (2002), Agarwal et al. (2004), Miranda and Simpson (2005). These models are generally based on a set of linear or non-linear equations and can accommodate effects of varying physical properties of vapor/steam and liquor with change in temperature and concentration.

These models offer limited flexibility as far as handling of operating strategies is concerned. For example, if feed sequence has to be changed or any flash term (Product, Feed, condensate, etc.) is to be added or deleted or the streams are to be splitted or joined the whole set of equations of the model needs to be reframed. This offers considerable rigidity for use of the model, especially when one is exploring an optimum operating strategy from a number of feasible ones (Mathur, 1992).

To overcome this difficulty, Stewart & Beveridge (1977) developed cascade algorithm in which model equations of an effect is solved repeatedly in a predetermined sequence to simulate different operating strategies of a MEE system. The cascade simulation based model of Stewart and Beveridge (1977) was improved by Ayangbile et al. (1984). Their algorithm was capable of handling any number of feed splitting/joining operations. However, it has limitation, as it did not account operating strategies like reheating, flashing, etc. Bremford and Muller-Steinhagen (1994) proposed an iterative method for the simulation of MEE system but did not include the provision of vapor bleeding and also considered constant value of U.

Under the above background the present work has been planned to provide a model which has the flexibility of model of Ayangbile et al. (1984) but do not have the limitations. Thus, the model of above authors has been modified and improved in the present work. It accounts for different operating strategies such as steam and liquor splitting, feed sequencing, condensate, feed and product flashing, vapor bleeding for re-heaters, etc. In this paper the model for an effect is represented by single cubic polynomial, which utilizes the value of U supplied to it through an empirical correlations developed from the plant data. The model also accounts for Q_{loss} from effects and τ. It will be validated against plant data and used to study the effect of variations of different operating parameters such as T_S, x_F, T_{Le}, T_F and F on steam consumption (SC), steam economy (SE) and product concentration (x_p).

2. Problem statement

The MEE system selected for above investigation is a Septuple Effect Flat Falling Film Evaporator (SEFFFE) system operating in a nearby Indian Kraft Pulp and Paper Mill for concentration of non-wood (straw) black liquor. Black liquor is a mixture of organic and inorganic chemicals. The proportion of organic compounds in the liquor ranges from 50 to 70%. Table 1 shows the inorganic constituents of Kraft black liquor found in Indian paper mills.

Table 1

Weak Kraft Black Liquor Constituents

S. No.	Organic Compounds	
1	Alkali lignin and thiolignins	
2	Iso-saccharinic acid	
3	Low molecular weight polysaccharides	
4	Resin and fatty acid soaps	
5	Sugars	
	Inorganic Compounds	**gpl**
1	Sodium hydroxide	4-8
2	Sodium sulphide	6-12
3	Sodium carbonate	6-15
4	Sodium thiosulphate	1-2
5	Sodium polysulphides	Small
6	Sodium sulphate	0.5-1
7	Elemental sulphur	Small
8	Sodium sulphite	small

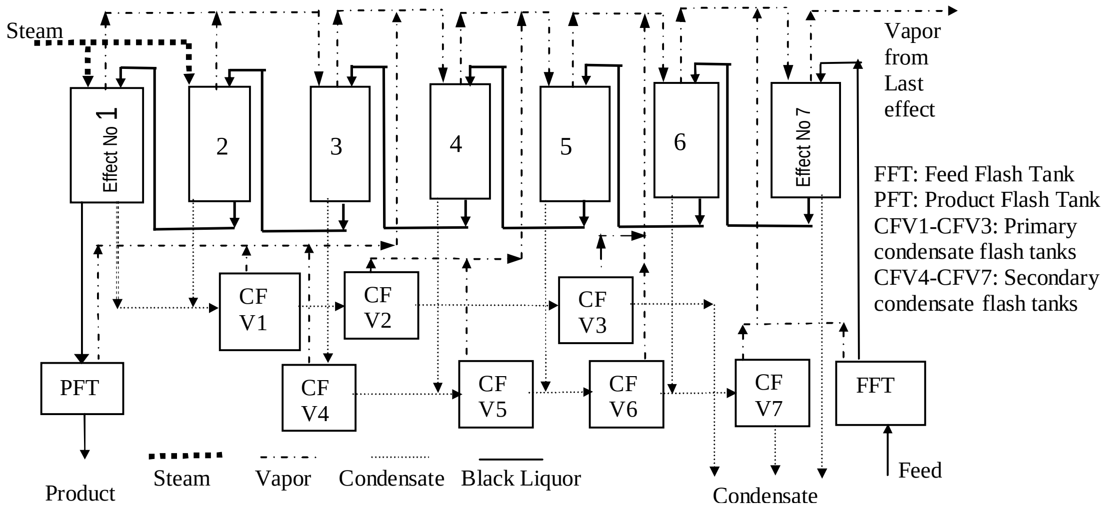


Fig. 1 Schematic diagram of SEFFFE system

The schematic diagram of a SEFFFE system with backward feed flow sequence is shown in Fig. 1. The first two effects of it require live steam. This system employs feed, product and condensate flashing to generate auxiliary vapor, which are then used in vapor bodies of appropriate effects to improve overall SE of the system. The last effect is attached to a vacuum unit. The base case operating and geometrical parameters for this system are given in Table 2 which shows that steam going into first effect is 7 °C colder than that into second effect. This is an actual scenario and thus it has been taken as it is during simulation. The plausible explanation is unequal distribution of steam from the header to these effects leading to two different pressures in the steam side of these effects.

Table 2

Base case operating and geometrical parameters for the SEFFFE system

S. No	Parameter(s)		Value(s)
1	n		7
2	n_s		2
3	T_S	Effect 1	140 °C
		Effect 2	147 °C
4	x_F		0.118
5	T_F		64.7^{o}C
6	F		56200 kg/h
7	T_{Le}		52 °C
8	Feed flow sequence		Backward
9	Heat Transfer Area	Effect 1 and 2	540 m^2 each
		Effect 3 to 6	660 m^2 each
		Effect 7	690 m^2

3. Model development

3.1. *Boiling Point Rise (τ)*

For development of a correlation for τ of black liquor, the functional relationship is taken from well established TAPPI correlation (Ray et al., 1992). For i^{th} effect where concentration of black liquor is x_i, τ is given as:

$$\tau=C_3(C_2+x_i)^2 \tag{1}$$

To develop Eq.1 different samples were collected from the SEFFFE system and experiments were conducted under controlled conditions in the R & D section of the industry to determine τ as a function of temperature as well as concentration of black liquor. It should be noted that plant data (such as liquor temperature and concentration) used in the present study were measured after calibrating the sensors. Additional measurements of temperature and concentrations were also performed in those places where routine measurements were not performed. Based on value of τ, obtained from experiment, a correlation similar to Eq. 1 is developed as given below:

$$\tau=20(0.1+x_i)^2 \tag{2}$$

Eq. 2 predicts the plant data, given in Table 3, with an average error of 2.4%. In fact, the τ of black liquor depends on its chemistry and so it is affected by changing the black liquor. Also no data for τ of black liquor on FFFE is available in the literature. Thus, it is not possible to validate the correlation of τ developed through Eq. 2 against data from other paper industry as well as from available literature.

Table 3

Data for determination of τ

x_i	0.0767	0.091	0.106	0.13	0.169	0.244	0.369	0.462	0.47
τ_i	0.60	0.70	0.80	1.10	1.40	2.30	4.30	6.20	6.40

3.2. *Development of model of an effect*

By taking mass and energy balances over i^{th} effect of a SEFFFE system, shown in Fig. 2, following equations can be developed.

Overall mass balance around evaporation section

$$L_{i+1}=L_i+V_i \tag{3}$$

Overall mass balance around steam chest

$$V_{i-1}=CO_{i-1} \tag{4}$$

Partial mass balance for solids provides

$$L_{i+1}x_{i+1}=L_ix_i=L_Fx_F \tag{5}$$

Overall energy balance gives

$$L_{i+1}h_{Li+1}=L_ih_{Li}+V_iH_{Vi}+\Delta H_i \tag{6}$$

where ; $\Delta H_i=U_iA_i(T_{i-1}-T_{Li})$ (7)

$$T_{Li}=T_i+\tau_i \tag{8}$$

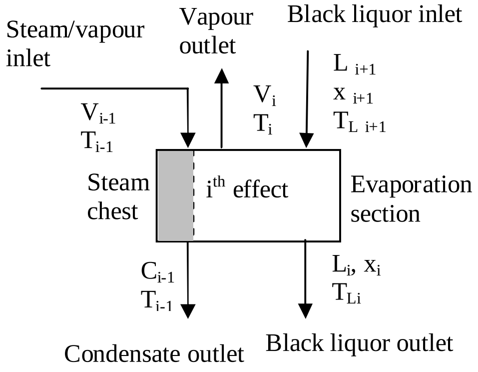


Fig. 2 Block Diagram of an evaporator

$$h_L = C_{PL}\,(T_L - C_5) \tag{9}$$

$$C_{PP} = C_1 * (1-C_4x) \tag{10}$$

The values of coefficients C_1, C_4 and C_5 are 4187, 0.54 and 273, respectively.

Energy balance on steam/vapor side gives rise to:

$$V_{i-1}=\Delta H_i/(H_{Vi-1}-h_{i-1}) \tag{11}$$

$$V_{bi}=(\Delta H_i + Q_{loss})/(H_{Vi-1}-h_{i-1}) \tag{11a}$$

Combining Eqs. 2 to 10 and eliminating V_i, x_i, h_{Li}, ΔH_i and T_{Li} one gets following cubic polynomial equation in terms of L_i:

$$a_1L_i^3+a_2L_i^2+a_3L_i+a_4=0 \tag{12}$$

where, coefficients a_1, a_2, a_3 and a_4 of the cubic polynomial are functions of input liquor parameters and other known parameters like A_i and U_i of the i^{th} effect. The expressions for coefficients a_1, a_2, a_3 and a_4 are:

$$a_1=H_{vi}-C_1T_i-C_1C_2^2C_3+C_1C_5 \tag{12a}$$

$$a_2=L_{i+1}h_{Li+1}+U_iA_i(T_{i-1}-T_i-C_3C_2^2)+L_{i+1}x_{i+1}(C_1C_4T_i-2C_1C_2C_3+C_1C_3C_2^2C_4-C_1C_4C_5)-L_{i+1}H_{vi} \tag{12b}$$

$a_3=(L_{i+1}x_{i+1})^2(2C_1C_2C_3C_4-C_1C_3)-2C_2C_3U_iA_iL_{i+1}\,x_{i+1}$ (12c)

$a_4=(C_1C_3C_4L_{i+1}x_{i+1}-C_3U_iA_i)(L_{i+1}x_{i+1})^2$ (12d)

In the present work all coservative equations as well as physical properties of liquor are used to develop a single cubic polynomial model for an effect. This is an advancement over the existing models as in these physical properties are computed first and then conservation equations are solved to get the results of the model of an effect. However, in this model all these computation can be carried out in a single step. Moreover, in general, the number of equations used to describe an effect by earlier investigators, is 3 or 4 in contrast to only one used in the present work. This helps in reducing the overall size of the problem and also the burden of computation to a large extent.

3.3. Development of model for liquor flash tank

For liquor (feed/product) flash tank, in which liquor (L_{in}) of concentration (x_{in}) is entering at T_{Lin} and being flashed at T_{out}, a similar cubic model, as presented in Eq. 12, is proposed. The modified expressions for constants a_1 to a_4 are described below. As a consequence of flashing vapor, V_{fout}, is generated.

$a_1=H_{vout}-C_1T_{out}-C_1C_2^2C_3+C_1C_5$ (12e)

$a_2=L_{in}h_{Lin}+L_{in}x_{in}(C_1C_4T_{out}-2C_1C_2C_3+C_1C_2^2C_3C_4-C_1C_4C_5)-L_{in}H_{vout}$ (12f)

$a_3=(L_{in}x_{in})^2(2C_1C_2C_3C_4-C_1C_3)$ (12g)

$a_4=(L_{in}x_{in})^3C_1C_3C_4$ (12h)

The cubic equation, Eq. 12, is solved to get its real root(s). Out of real roots only one root, which has a value equal or less than black liquor feed rate, is selected for further processing. Once, this root is known, other parameters like exit liquor concentration, temperature and vapor produced (V_i) are computed using Eqs. 5, 8 and 3. Use of Eqs. 7 and 11 provides the quantity of vapor required (V_{i-1}) to provide the necessary heat for the evaporation.

3.4. Development of model for condensate flash tank

Material and energy balances over condensate flash tank yields following relation to determine exit condensate flow rate (CO_j), for a known condensate flow rate, CO_i, entering at a temperature, T_i, with specific enthalpy, h_i, and being flashed at temperature, T_j . The overall mass and energy balance give:

$CO_j=CO_i(H_{Vj}-h_i)/(H_{Vj}-h_j)$ (13)

and $V_{fout,j}=CO_i-CO_j$ (14)

3.5. *Development of model for a re-heater*

Re-heater is modeled to achieve a targeted rise in black liquor temperature (T_T) using bled vapor from the SEFFFE system.

$V_{ph}=LC_{pL}(T_T–T_{Lin})/(H_v-h)$ (15)

Where, $T_T=T_{L,i-1}+0.5(T_i–T_{L,i-1})$

3.6. *Development of empirical correlations for Q_{loss} and U*

It was considered that Q_{loss} from a given effect is entirely due to Natural Convection and thus can be expressed as $q = f(\Delta t)$ Coulson and Richardson, (1996). This equation was regressed using plant data and the Eq.16 is developed which basically a plant specific equation. However, the functional relationship between q and Δt may hold good for other evaporators as well.

This is a fact that correlations for the predication of U for flat falling film evaporators are hardly available in open literature. Thus for the simulation of SEFFFE system it was thought necessary to develop a correlation for U based on plant data. It is also a well known fact that plant data are not recorded properly and are in most of the cases deficient in terms of providing a complete picture. The propose SEFFFE system had both of these weaknesses. These problems were tackled by collecting large sets of data from the plant and then screening out those sets for correlation development which satisfy material and energy balances. Additional data from intermediate points of the evaporator systems were also collected to help in conducting mass and energy balances around each effect. It was found that out of the collected data sets, about 70% are of no use. The screened sets are only used for development of correlations for prediction of U.

3.6.1. *Correlation of Q_{loss}*

Analogous to $q = f(\Delta t)$, a simplified empirical correlation for heat losses to environment from different effects of a SEFFFE system is developed as given below:

$Q_{loss} \alpha\ (\Delta t)^{1.25}$

Where, Δt is difference of temperature between vapor body and ambient. Regression, using values of (Δt) and corresponding values of computed heat losses, yields following empirical correlation:

$$Q_{loss}=1.9669*10^{3}(\Delta t)^{1.25} \qquad (16)$$

Predictions from Eq. 16 show an error limit of –33 to +29%. In the present SEFFFE system the average Q_{loss} was of the tune of 4% of total energy input to the system. It appears that the present Q_{loss} is at a higher side in the plant may be due to degraded insulation.

3.6.2. Correlation of U

Many investigators such as, Gudmundson (1972) and Beccari et al. (1975), have proposed mathematical models to predict U but these were for LTV evaporators. Recently, Xu et al. (2004a, 2004b) and Prost et al. (2006) have developed correlations for the prediction of U but for horizontal as well as vertical tube falling film evaporators and not for FFFEs. The only work, which appears to be available, is that of Pacheco et al. (1999). They proposed a correlation for U of a FFFE for concentrating sugar cane juice as a function of ΔT and x. Moreover, a statistical analysis of plant data for SEFFFE system, shown in Table 4, illustrates that besides ΔT and x, U also depends on flow rate of liquor. It appears that no correlation is available in the literature, which can be directly used in the present investigation for the prediction of U. Thus, it becomes necessary to develop a correlation of U for FFFE system.

An analysis of U values of all seven effects for four data sets, shown in Fig. 3, clearly indicates that the effect No.1 & 2 follow a different trend than all other effects i.e. 3 to 7. The values of U are substantially low for effect No.1 & 2. This lower value of U is primarily due to higher concentration of black liquor (43% to 53%) handled by these effects which accelerates crystallization fouling. In fact, in the vicinity of 48% solid concentration the scale formation starts (Süren, 1995). This occurs due to crystallization of inorganic species sodium carbonate (Na_2CO_3) and sodium sulfide (Na_2SO_4), present in the black liquor, on the metal surface. These form the double salt Burkeite ($2Na_2SO_4.Na_2CO_3$) when they co-crystallize (Hedrick and Kent, 1992, Schmidl and Frederick, 1999 and Chen and Gao, 2004). The above salts are also present in the black liquor considered for the present investigation as shown in Table 1. This phenomenon causes U to fall drastically in first two effects.

Therefore, two different empirical correlations are developed, one for effect Nos. 1 to 2 and other for effect Nos. 3 to 7. The normalized Power law equation, shown in Eq. 17, is used for both the correlations using divisors 2000 $W/m^2/K$, 40 °C, 0.6 and 25 kg/s as these are higher than the respective highest values encountered in the plant data.

$$(U/2000)=a(\Delta T/40)^b(x_{avg}/0.6)^c(F_{avg}/25)^d \quad (17)$$

The estimated values of U from plant data, for all seven effects, are used to estimate unknown coefficients a, b, c and d of Eq. 17 as shown in Table 5 using constrained minimization technique of Sigma Plot software. To show the extent of fitting plant and computed data for U from Eq. 17a & b are plotted in Fig. 4, which clearly shows that correlations, Eq. 17a & b predict the U values within an error limit of ±10%. In the absence of any data for U of FFFEs employed for concentration of black liquor or any developed correlation in this regard, it was not possible to compare these equations with the work of others. Thus, the above correlations are industry as well as liquor specific. However, the functional relationship between U and other parameters as given in these equations can be effectively utilized to develop correlations of U for other situations also.

Table 4

Cross correlation coefficients between parameters U, ΔT, X_{avg} and F_{avg}.

	U	*delta T*	*Xavg*	*Favg*
U	1			
delta T	-0.96999	1		
Xavg	0.928086	-0.92326	1	
Favg	-0.81645	0.875356	-0.90815	1

Table 5

Value of Coefficients of Eq. 17

Effect No.	a	b	c	d	% Error	Eq. No.
1 and 2	0.0604	-0.3717	-1.227	0.0748	-11.32 to 7.25	17(a)
3 to 7	0.1396	-0.7949	0.0	0.1673	-11.75 to 8.20	17(b)

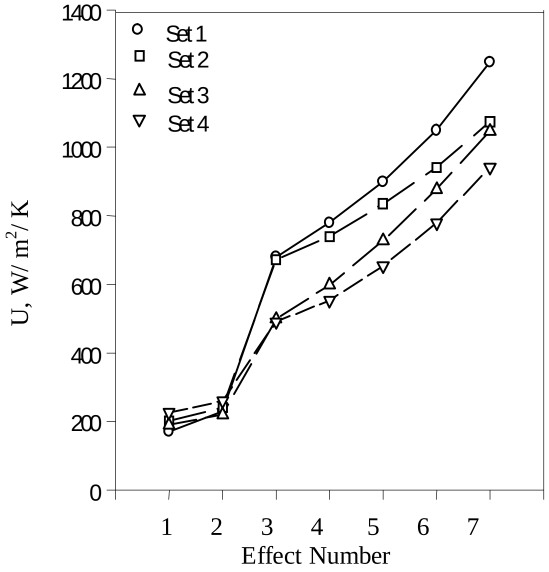


Fig. 3 Profiles of U from Plant Data

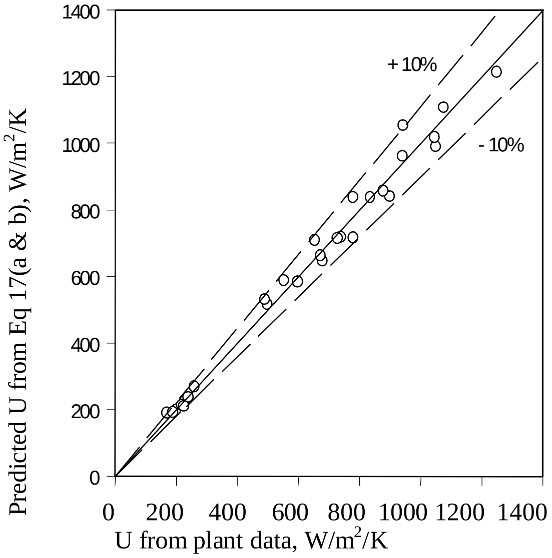


Fig. 4 Comparison of U from plant data and those predicted from Eq. 17(a & b)

3.7. *Development of generalized model for a MEE system*

The modified block diagram of ith effect is shown in Fig. 5, which accommodates any flow sequencing and liquor splitting. The black liquor feed rate to ith effect can be expressed as:

$$L_{i+1} = y_{oi}\, L_F + \sum_{\substack{j=1\\ j\neq i}}^{n} y_{ji} L_j \qquad (18)$$

Where, y_{oi} is the fraction of the feed (after feed flash), which enters into ith effect and y_{ji} is the fraction of black liquor which is coming out from jth effect and enters into the ith effect.

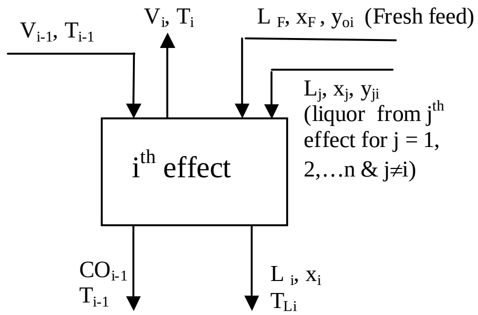


Fig. 5 Block Diagram of an evaporator for cascade simulation

Total mass balance around i^{th} effect gives;

$$y_{o,i}\, L_F + \sum_{j=1}^{n} y_{j,i} L_j = L_i + V_i$$

or $$\sum_{j=1}^{n} y_{j,i} L_j - L_i = V_i - y_{o,i} L_F \qquad (19)$$

The expression, developed for i^{th} effect and shown in Eq. 17, can be represented for all n effects by a Matrix Equation as given below:

$$\mathbf{Y_0}\, L_F + \mathbf{Y}\,\mathbf{L} \qquad (18)$$

Where, $\mathbf{Y_0} = \begin{bmatrix} y_{01} & y_{02} & y_{03} & \ldots\ldots & y_{on} \end{bmatrix}^T$

$$\mathbf{Y_f} = \begin{bmatrix} y_{11} & y_{21} & y_{31} & \cdots & y_{n1} \\ y_{12} & y_{22} & y_{32} & \cdots & y_{n2} \\ y_{13} & y_{23} & y_{33} & \cdots & y_{n3} \\ : & : & : & \cdots & : \\ y_{1n} & y_{2n} & y_{3n} & \cdots & y_{nn} \end{bmatrix}$$

And $\mathbf{L} = \begin{bmatrix} L_1 & L_2 & L_3 & \ldots\ldots\ldots & L_n \end{bmatrix}^T$

Where, **Y** is the flow fraction matrix. Its diagonal elements, y_{jj} are equal to zero.

For development of a general model of an evaporator system, mathematical model for i^{th} effect as given by Eq. 12b to 12d is generalized by replacing the inlet liquor flow term, L_{i+1}, by expression given in Eq. 18.

Further, vapor required in i^{th} effect steam chest i.e. V_{bi}, calculated after solving the model of an effect and V'_{i-1} (vapor available for supply to i^{th} effect steam chest) can be modified to incorporate flash vapor produced by feed-, product- and condensate- flashing along with vapor produced in the $(i-1)^{th}$ effect. Further, vapor bled to re-heater is deducted from it and thus, vapor available for i^{th} effect can be obtained to provide the required heat. This has been clearly shown in Fig. 6. The values of vapor denoted by V'_{i-1} and V_{bi} should be equal for an exact solution. An index called "Performance Index (PI)" is defined as a measure of the difference in V'_{i-1} and V_{bi}.

$$PI=\Sigma((V'_{i-1}-V_{bi})/V_{bi})^2 \qquad (26)$$

$$\text{Where, } V'_{i-1} = V_{i-1} + V_{fout} - V_{ph} \qquad (27)$$

The summation term shown in Eq. 26 is for 'n_s+1' to 'n' effects, where first n_s effects are fed with live steam. The summation of V_{bi} for first n_s effects gives total steam consumption, and summation of V_i from first n_s effects is the vapor fed to $(n_s+1)^{th}$ effect vapor chest, as shown in Fig. 6.

$$SC = \sum_{i=1}^{n_s} V_{bi} \qquad (28)$$

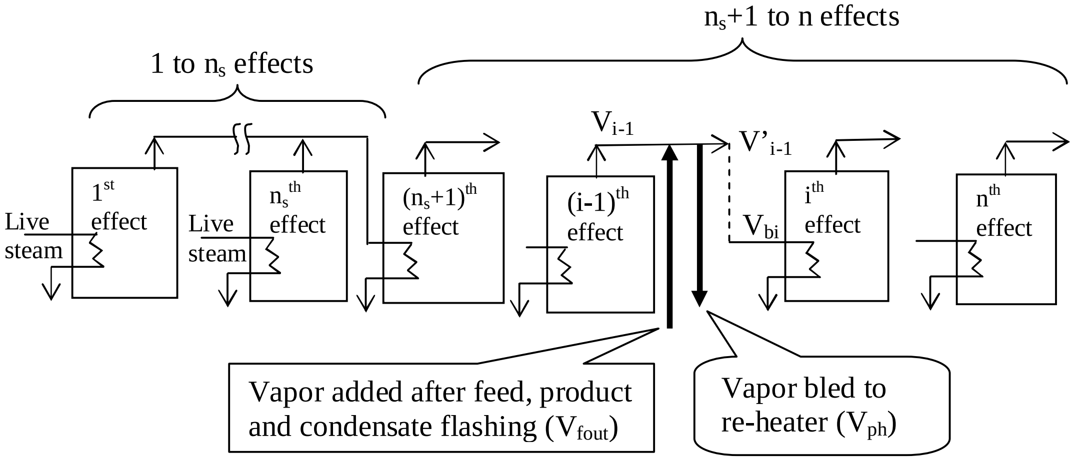


Fig. 6 The schematic diagram of vapor flow in a MEE system consisting of 'n' effects

4. Boolean and flow fraction matrices

To express feed flow sequence in the present investigation, Boolean matrix is used. The order of the matrix is (n+1)×(n+1), where first column denotes the feed stream and subsequent columns are source effects 1 to n and first n rows are sink effects and last row is product stream. A unit value of element b_{ij} indicates that liquor exiting from $(j-1)^{th}$ effect enters i^{th} effect. Boolean matrix, shown below, is for backward flow sequence of the SEFFFE system. In this matrix the element $b_{13} = 1$ shows that liquor exits 2nd effect and enters the first effect.

$$\begin{array}{r|cccccccc|l}
 & \text{(Feed) F} & 1 & 2 & 3 & 4 & 5 & 6 & 7 & \xleftarrow{\text{Source effect}} \\
 & & & & & & & & & \downarrow \text{Sink effect} \\
 & 0 & 0 & \boxed{1} & 0 & 0 & 0 & 0 & 0 & 1 \\
 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & 2 \\
 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 3 \\
\boldsymbol{B} = & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 & 4 \\
 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 0 & 5 \\
 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 1 & 6 \\
 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 7 \\
 & 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 & \text{P (Product)}
\end{array}$$

To incorporate splitting of black liquor feed and/or intermediate liquor streams a flow fraction matrix $\mathbf{Y_f}$ of size (n+1)×(n+1) is defined. It is an augmented form of matrix $\mathbf{Y}$ with an extra column for feed (1st in the matrix) and an extra row for product (8th row in the matrix). For a flow sequence when feed is splitted equally to enter 6th and 7th effects and then combined liquor output of these effects enter 5th effect, the flow fraction matrix $\mathbf{Y_f}$ is shown below:

$$\boldsymbol{Y_f} = \left| \begin{array}{cccccccc} 0 & 0 & 1 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \boxed{1} & \boxed{1} \\ \boxed{0.5} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ \boxed{0.5} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 & 0 & 0 & 0 & 0 \end{array} \right|$$

Similarly, placements of condensate, feed and product flash tanks as well as placement of re-heaters are also decided by respective Boolean matrices.

This method of representation helps to alter the connectivity of the system through data file and helps in accommodating different operating strategies with ease.

5. Solution of the model

A complete analysis for the solution of model is given in Table 6, which indicates the input and output variables and equations to be solved. The solution of the mathematical model starts with assumed values of operating pressures for effect number 1 to (n-1) based on equal ΔP in all effects. It gives the values of vapor required (V_{bi}) along with the vapor available (V'_{i-1}) for each effect and then Performance Index (PI) is calculated using Eq. 26. If it is greater than desired accuracy (say, $5*10^{-6}$), next iteration is to be performed. This will require new and improved estimates of P_i for i = 1 to (n-1). The solution technique is described in the work of Bhargava et al. (2007).

Table 6

Input and Output Parameters of the model

S. No.	Input Parameter	Equations to be solved	Output Parameter	Remarks
1.	n, n_s, P_1 to P_n, T_1 to T_n, F, x_F, T_S, H_1 to H_n, h_{L1} to	Eq. 12, 12e to 12h, mass and component balance	L_{out}, x_{out}, T_{Lout}, V_{fout}	

	h_{Ln}, C1 to C5, **B** for feed flashing	around FFT		
2.	**B** for a flow sequence, effect number (i^{th} effect) ΔT, x_{avg}, F_{avg}	Eq. 17, 17a and 17b	U	These steps are solved for each effect depending on **B**.
3.	**B** for a re-heater, T_{Li-1}, T_i, H_V, h, L	Eq. 15	V_{ph}	
4.	U, $\mathbf{Y_f}$, H_V, h_L, x_{i+1}, A, C1 to C5	Eq. 12, 12b to 12d, Eq. 3, 5, 7, 8, 11	L_i, x_i, V_i, T_i, V_{i-1}	
5.	Δt	Eq. 16	Q_{loss}	
6.	Q_{loss}	Eq. 11a	V_{bi}	
7.	**B** for condensate flashing, H_V, h	Eq. 13, 14	CO, V_{fout}	
8.	**B** for product flashing, T_L, H_V, h_L, C1 to C5, x_L	Eq. 12, 12e to 12h, mass and component balance around PFT	V_{fout}, x_P (x_{out}), L_{out} (product flow rate)	
9	V_{ph}, V_{i-1}, V_{fout} (generated from feed, product & condensate flashing)	Eq. 27	V'_{i-1}	
10	V'_{i-1}, V_{bi}	Eq. 26	PI	
11	V_{bi}, n_s	Eq. 28	SC	

5.1. Algorithm for solution of model

To simulate the mathematical model developed in the present work computer program is developed in FORTRAN. A complete solution is provided in Appendix A. The stepwise algorithm is given below:

1. Read values of input parameters, given in Table 6.
2. Convert flow fraction matrix, $\mathbf{Y_f}$, to $\mathbf{Y}$, by removing first column and last row. Compute [**Y-I**] and invert it to obtain matrix **A** as defined in Eq. 21.
3. Determine sequence of computation using Boolean matrix ***B*** for feed flow sequence.
4. Assume initial set of operating pressures for effect numbers 1 to (n-1).
5. Calculate steam/vapor and condensate properties using all the pressures including live steam and last effect pressures.
6. Decide the operating conditions for feed flash tank, as dictated by its Boolean matrix. Feed flash calculations are carried out only if feed temperature is more than its operating temperature by solving cubic polynomial as given by Eqs. 12, 12e to 12h. This provides L_{out}, x_{out}, T_{Lout} and V_{out}.
7. Start computation for the selected first effect as per the sequence of computation as decided in Step no. 3.
8. Compute total liquor flow rate to the effect considered, as given by Eq. 18, and also calculate its temperature and concentration.
9. Check re-heater Boolean matrix for placement of re-heater if any before this effect. If yes, carry out calculations to determine liquor outlet temperature and quantity of vapor required to preheat the liquor using Eq. 15.
10. Initially, for the calculation of U of an effect, using Eq. 17 (a) or (b), consider x_{avg} and F_{avg} equal to inlet liquor -concentration and - feed rate as computed in Step 8. Also calculate ΔT considering τ based on inlet liquor concentration.

11. Compute outlet liquor flow rate, L_i, by solving cubic polynomial as described by set of Eqs. 12 and 12 (a) through (d). Using value of L_i compute V_i, T_{Li} and x_i.
12. Compute V_{bi} employing Q_{loss} and ΔH_i. Compute Q_{loss} and ΔH_i using Eq. 16 and Eq. 11, respectively.
13. Compute x_{avg} and F_{avg} for the effect. If absolute value of difference between computed values and assumed values of these parameters is more than the prescribed error limit (10^{-5}) then repeat the computation starting from Step No. 10. Otherwise proceed to the next Step 14.
14. The procedure from step 8 to 13 is repeated for all the effects based on the sequence of computation determined in step 3.
15. Compute condensate flash, as decided by condensate flash Boolean matrix, using Eq. 13&14 to determine exit condensate flow rate and flash vapor generated respectively.
16. Product flash Boolean matrix decides the product flash calculation. Methodology as given for feed flash in step 6 is adopted for the computation. It also gives exit liquor flow rate, concentration, temperature and product flash vapor generated.
17. Total vapor available for an effect (V_{i-1}) is computed by adding vapor produced in preceding effect with feed-, product-, and condensate-flash vapor and then subtracting vapor required in re-heater. This procedure is carried out for (n_s+1) to n^{th} effect, as in first n_s effects live steam is used.
18. Performance index (PI) is computed as per Eq. 26. If the value of PI is less than desired accuracy ($5x10^{-6}$) then stop otherwise proceed to Step 19.
19. Solve the complete model as described in Bhargava et al. (2007) for modified values of pressures for effect numbers 1 to (n-1).

6. Validation of the model

To validate the model simulation runs are carried out using base case operating parameters, given in Table 2.

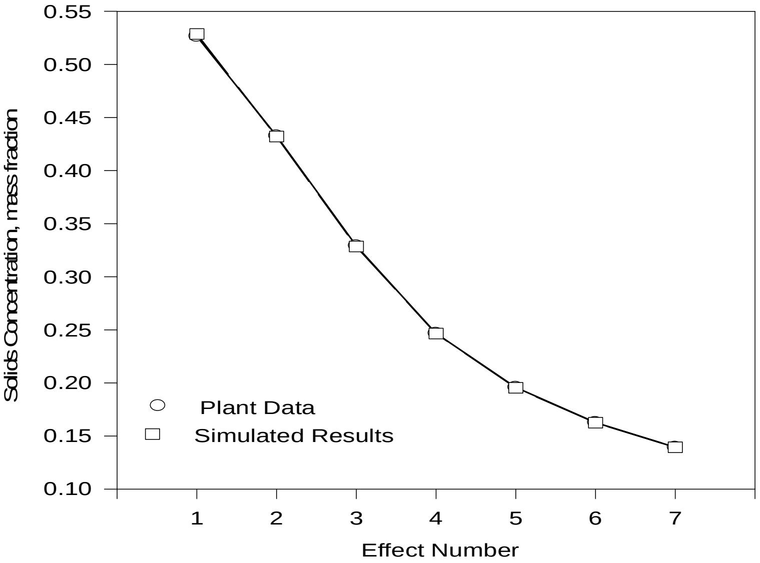


Fig. 7. Comparison between Solid concentration in liquor from plant data and that predicted by model

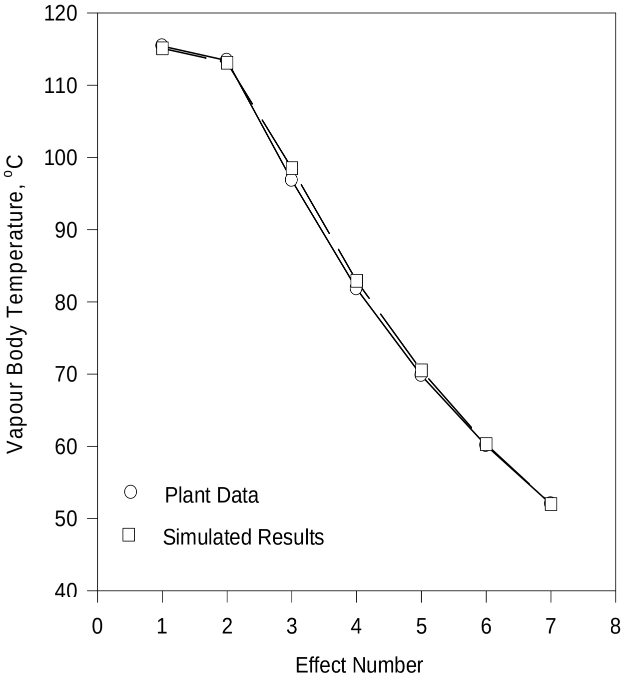


Fig. 7 and 8 have been plotted to show the comparison between experimental data obtained from the mill for concentration of black liquor and vapor body temperature of different effects with that obtained from model respectively. Predicted results show that the liquor concentration match within an error band of -0.2 to +0.4%, and the vapor temperature of different effects match within an error limit of –0.26 to +1.76%. The present model computes the temperature difference (ΔT) for each effect with a maximum relative error of 23% between plant data and simulation result. However, for the similar MEE system the published model (Bremford and Muller-Steinhagen, 1994) reported a maximum error of 43.43% for the prediction of temperature difference in each effect. Thus, it appears that the present model predicts the plant data fairly well in comparison to the published model.

7. Results and discussions

After establishing the reliability of the present model, it was thought logical to study the variation of output parameters such as SC, SE and x_P with change in input parameters, T_S, T_{Le}, T_F, x_F and F, so that better operating conditions can be identified which will give maximum SE for the SEFFFE system. In the present investigation, the input parameters are varied within a range, as given in Table 7, around the base case values to study its effect on output parameters. The ranges of input parameters, shown in Table 7, are considered after analyzing the prevailing practices in Indian paper mills.

Table 7

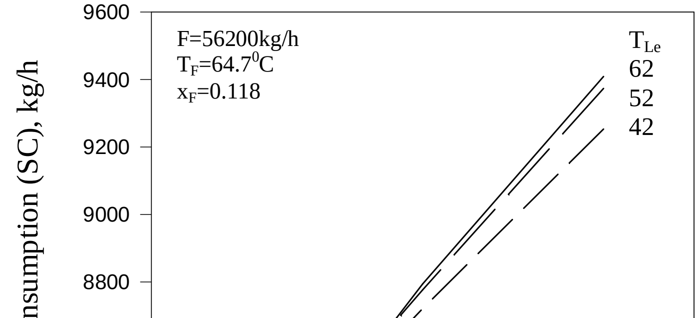
Ranges of operating parameters of a SEFFFE system

Parameters	Variation in value
T_S	120^{o}C-160^{o}C
x_F	8%-16%
T_{Le}	42^{o}C-62^{o}C
T_F	44.7^{o}C-84.7^{o}C
F	56200-78680 kg/h

It appears that the SE is the single most prominent parameter to evaluate the efficiency of the SEFFFE system as it varies with variation in operating parameters and geometrical parameters as well. Moreover, the contributions of SC and x_P are also included in it as the value of SE is the ratio of total water evaporated to total SC. In addition to it, the amount of evaporated water is also related to the value of x_P directly. Though by monitoring SE one can keep a watch on the economics of evaporation, the study of variations in parameters such as SC and x_P with input parameter offers better understanding of the process.

7.1. Effects of T_S and T_{Le} on SC, SE and x_P

Figs. 9 to 11 have been prepared to show the effect of T_s and T_{Le} on SC, SE and x_p for specified values of x_F, F and T_F as shown in these figures. Fig. 9 shows that with the increase in T_s, there is a considerable increase in the value of SC, for all values of T_{Le}. Whereas, for a constant value of Ts, when T_{Le} is varied, the value of SC does not change considerably. At the highest value of T_s the SC differs only by 1.68% when T_{Le} is varied from 42 to 62°C. As this difference is very small as compared to errors involved in the prediction of some variables through empirical correlations it can be concluded that the effect of T_{Le} on SC is insignificant.



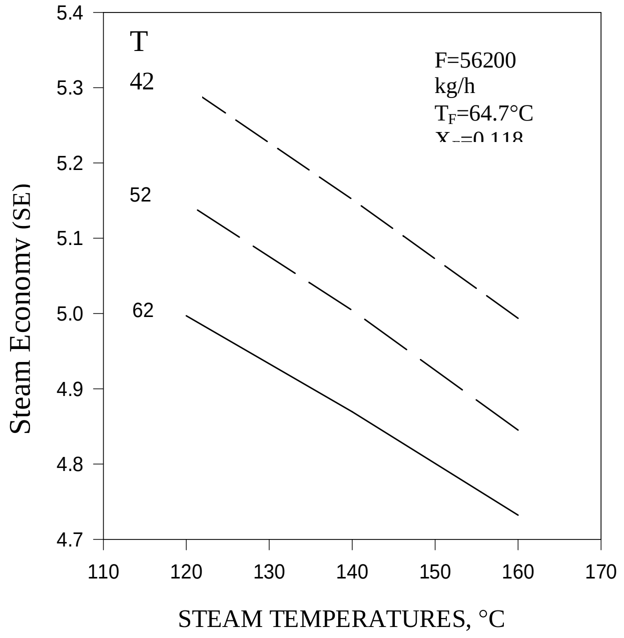


Fig. 10. Effect of T_S on SE with T_{Le} as a parameter

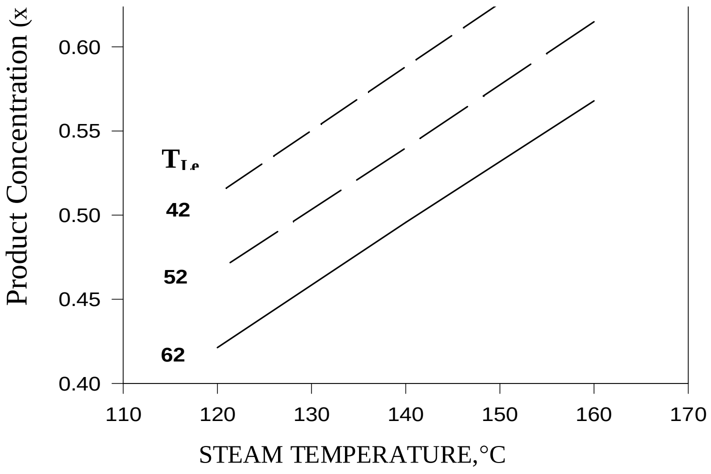


Fig. 11. Effect of T on x with T as a parameter

The increase in the value of SC with increase in T_s can be attributed to decrease in latent heat of condensation of the steam with increase in the value of T_S. Further, an increase in T_S, increases the temperature difference (ΔT) between steam and liquor, thus provides conducive environment to pump more heat in to the effect causing more evaporation. This in turn increases the liquor concentration in each effect. As a result of it, lowering of U with increase in T_S is observed in first two effects also where live steam is fed. The cumulative effect of above factors is well represented by cumulative values of $U\Delta T$ for first two effects (as the areas of these effects are same). It is seen from Table 8 that with the increase in T_S the value of $U\Delta T$ for first two effects increases as a result more heat is pumped to these effects. Thus, the value of SC increases with increase in T_s.

Table 8

Values of sum of $U\Delta T$ for first two effects

Cumulative Values of $U\Delta T$, W/K			
Value of T_{Le}, °C	Value of T_s, °C		
	160°C	140°C	120°C
42	9485.907	9253.592	8936.813

52	9597.748	9292.552	8920.395
62	9619.321	9301.958	8837.805

It is a fact that SE depends on total water evaporated and SC. For the SEFFFE system total evaporation depends on vapor produced from effects nos. 1 to 7 as well as those generated from feed and product flashing. Amount of total evaporation has a direct relationship with x_P also. Table 9 shows variations in SC and different components of total evaporation with variation in T_S when other input parameters such as T_{Le}, T_F, F and x_F are kept constant at 52 °C, 64.7 °C, 56200 kg/h and 0.118, respectively.

From Fig. 10 it can be seen that the value of SE decreases with the increase in T_S for all value of T_{Le} investigated. This phenomenon can be easily explained from the variation pattern of SC and total evaporation with T_S. With increase in T_S, SC increases rapidly. However, total water evaporated does not increase in the same ratio. For example, when T_{Le} is kept at 52°C and T_S is increased from 120 to 160°C it increases SC by 14.9% whereas, total evaporation increases by 8.2% only as evident from Table 9. The net result is that SE decreases with the increase in T_S. As has been seen in the case of variation of SC with T_{Le} for a given value of Ts, SE also does not vary appreciably with the variation in T_{Le}.

Table 9

Effect of variation of T_S on SC and total evaporation

T_s °C	SP*	SC, kg/h	Total evaporation, kg/h								
			Feed Flash	Prod. Flash	Amount of vapor generated from different effect number (s)						Total evap.
					1	2	3	4	5	6	7	
120	F=56200	8159	466	267	2622	4614	6482	6745	6566	6427	7805	41993

140	T_F=64.7 T_{Le}=52 x_F=0.118	8776	320	274	2784	4842	6680	7052	6874	6775	8316	43917
160		9373	185	266	2932	5042	6854	7283	7071	7044	8738	45415

* Specified parameter

Fig. 11 shows that the product concentration increases with increase in T_S and decreases with increase in T_{Le}. With the increase in the value of T_s more heat is pumped to effects and thus causes more water to evaporate. For example, for a given value of T_{Le} equal to 52°C when the value of T_S is varied from 120 to 140°C and 120 to 160°C, the total evaporation increases by 4.6% and 8.2% respectively as evident from Table 9. This leads to a higher x_P. However, for a given value of T_s equal to 140°C when the value of T_{Le} changes from 42 to 52°C and 42 to 62°C total evaporation decreases by 2.2% and 4.7%, respectively. This results in lowering of the values of x_P.

7.2. *Effects of T_S, T_F, x_F and F on SC, SE and x_P*

Table 10 shows the effect of variations of input parameters such as T_S, T_F, x_F and F on SC, SE and x_p. The trends of behaviors of SC, SE and x_P with in input parameters are also shown in Table 11.

Table 10

Effects of T_s, T_F, x_F and F on SC, SE and x_P

Parameter		T_S=120°C			T_S=140°C			T_S=160°C			Specified parameters
		SC	SE	x_P	SC	SE	x_P	SC	SE	x_P	
x_F	0.08	8653.2	5.08	0.368	9345.9	4.91	0.436	9946.2	4.76	0.509	F=56200 kg/h

	0.118	8158.5	5.15	0.467	8776	5.00	0.54	9373.2	4.85	0.615	T_F=64.7°C
	0.16	7626.9	5.2	0.542	8233.4	5.07	0.622	8785.2	4.93	0.696	T_{Le}=52°C
T_F	44.7	8456.80	4.84	0.43	9123.50	4.70	0.485	9739.2	4.56	0.565	F=56200 kg/h
	64.7	8158.50	5.15	0.47	8776.00	5.00	0.535	9373.2	4.85	0.615	x_F=0.118
	84.7	7699.50	5.57	0.50	8316.90	5.39	0.57	8841.5	5.23	0.665	T_{Le}=52°C
F	56200	8158.5	5.15	0.467	8776	5.00	0.54	9373.2	4.85	0.615	x_F=0.118
	67440	9220.3	4.90	0.357	10101.2	4.68	0.395	10905.5	4.50	0.432	T_F=64.7°C
	78680	10115.3	4.67	0.296	11184	4.42	0.318	12228.7	4.19	0.339	T_{Le}=52°C

Table 11

Trends of SC, SE and x_P with change in T_s, T_F, x_F and F

Parameter	SC	SE	x_P	Specified parameter
T_s ↑	↑	↓	↑	x_F, F, T_F and T_{Le}
x_F ↑	↓	↑	↑	T_s, F, T_F and T_{Le}
T_F ↑	↓	↑	↑	T_s, x_F, F and T_{Le}
F ↑	↑	↓	↓	T_s, x_F, T_F and T_{Le}

The SC for the SEFFFE system depends largely on the cumulative value of $U\Delta T$ for first two effects. While comparing of above value, it is observed that it decreases by 10% when T_F changes from 44.7 to 84.7°C at T_s equal to 140°C. This clearly indicates that SC decreases with increase in T_F. Contrary to this, under above conditions, total evaporation increases by 4.4%. Due to increase in total evaporation and decrease in value of SC, SE increases with increase in T_F as is evident from Table 11. With rise in T_F more feed flash vapor is created and thus liquor with comparatively higher concentration enters into the 7th effect and after evaporation in subsequent effects produces a product with higher value of x_P. In other words it behaves as if the value of x_F has been virtually increased.

In fact, states of effect nos. 1 & 2 decide the SC. With the change in value of x_F from 0.08 to 0.16, the cumulative value of UΔT for first two effects is reduced by 11.6% due to increased concentration of liquor in these effects. As a result, the SC decreases when x_F is increased. For the same variation in x_F, however, total evaporation decreases by 9% and SC decreases by 11.9%. As the decrease in SC is more than that of evaporation, value of SE increases slightly (2.3%).

With the rise in value of F from 56200 to 78680 kg/h at T_s equal to 140°C, the cumulative value of UΔT of first two effects increases by 28%. This is due to increase in the value of F, which increases U considerably. This leads to higher SC in first two effects. However, the total water evaporated does not increase in the same proportion (it only increases by 12.6%). Thus the value of SE decreases with increase in F. The above computed results are from Table 10.

From above investigation, it is seen that for values of parameters T_S, T_L, T_F, x_F and F equal to 120°C, 84.7°C, 52°C, 0.118 and 56200 kg/h respectively, the SEFFFE system exhibits maximum SE of 5.57 with x_P and SC equal to 0.49 and 7700 kg/h, respectively. This value of SE is 11.6% more than the SE at which SEFFFE system is being operated currently. Thus, based on above analysis it can be suggested that only by changing the operating conditions, SE of the system can be improved without any prior modification in layout of the paper mill.

8. Conclusions

The salient conclusions of the present investigation are as follows:

1. The model developed in this investigation predicts liquor concentrations and temperatures of different effects within an error band of -0.2 to +0.4% and –0.26 to +1.8%, respectively. Also it simulates the plant data with considerably smaller amount of error in comparison to published model.

2. The correlations developed for τ and U predict the plant data with average absolute errors of 2.4% and 10%, respectively.
3. SE of the SEFFFE system can be improved by proper selection of values of operating parameters without any prior modification in the plant layout.

Nomenclature

A	Heat transfer area, m^2
a_{ij}	Element of matrix **A**
CO	Condensate flow rate, kg/s
C_P	Specific heat capacity, J/kg/K
h	Specific enthalpy of liquid phase, J/kg
H	Specific enthalpy of vapor phase, J/kg
I	Identity matrix
k	Iteration number
L	Liquor flow rate, kg/s
MEE	Multiple effect evaporator
n	Number of total effects
n_s	Number of effects supplied with live steam
P	Vapor body pressure, N/m^2
Q_{loss}	Heat loss, W
SC	Steam consumption, kg/h
SE	Steam economy
SEFFFE	Septuple effect flat falling film evaporator
T	Vapor body temperature, K
U	Overall Heat Transfer Coefficient, $W/m^2/K$
V	Vapor flow rate, kg/s

x	mass fraction
Y	Flow fraction matrix

Subscripts

avg	Average of inlet and outlet conditions
out	Exit condition
F	Feed
i	Effect number
L	Black Liquor
Le	Last effect
S	Steam
T	Target
V	Vapor
ph	Re-heater

Greek letters

τ	Boiling Point Rise, K

References

1. Agarwal, V. K., Alam, M. S., & Gupta, S. C. (2004). Mathematical model for existing multiple effect evaporator systems. *Chem. Eng. World, 39*, 76-78.
2. Ayangbile, W.O., Okeke, E.O., & Beveridge, G.S.G. (1984). Generalised Steady State Cascade Simulation Algorithm in Multiple Effect Evaporation. *Comp. Chem. Eng., 8*, 235-242.
3. Beccari, M., Biasi, L., Di Pinto, A., Prosperetti, A., Santori, M., & Tozzi A. (1975). Mathematical Model of an LTV Evaporator. *Int. J. Multiphase Flow, 2*, 357-361.

4. Bhargava, R. (2004). *Simulation of flat falling film evaporator network*. Ph.D. Dissertation, Department of Chemical Engineering, Indian Institute of Technology Roorkee, India.

5. Bhargava, R., Khanam, S., Mohanty, B., & Ray, A. K. (2007). Selection of optimal feed flow sequence for a multiple effect evaporator system. *Comp. Chem. Eng.*, (to be published, Reference CACE3561).

6. Bremford, D.J., & Muller-Steinhagen, H. (1994). Multiple effect evaporator performance for black liquor-I Simulation of steady state operation for different evaporator arrangements, *Appita J.*, *47*, 320-326.

7. Chen, F. C., & Zhiming, G. (2004). An analysis of black liquor falling film evaporation. *Int. J. Heat Mass Trans.*, *47*, 1657–1671.

8. Costa, A. O. S., & Enrique, E. L. (2002). Modeling of an industrial multiple effect evaporator system. *Proc. Congresso e Exposicao Anual de Celulose e Papel, 35th, Sao Paulo, Brazil, Oct. 14-17.*

9. Coulson, J.M. and Richardson, J.F. (1996). Chemical Engineering, Vol. 1, 5th Edn, Butterworth Heinemann Ltd.

10. El-Dessouky, H.T., Alatiqi, I., Bingulac, S., & Ettouney, H. (1998). Steady state analysis of the multiple effect evaporation desalination process, *Chem. Eng. Tech.*, *21*, 15-29.

11. El-Dessouky, H.T., Ettouney, H.M., & Al-Juwayhel, F. (2000). Multiple effect evaporation-vapor compression desalination processes, *Trans IChemE*, *78*, Part A, 662-676.

12. Gudmundson, C. (1972). Heat transfer in industrial black liquor evaporator plants Part-II. *Svensk Papperstidning Arg*, *75*, 901-908.

13. Hedrick, R. H., & Kent, J. S. (1992). Crystallizing sodium salts from black liquor. *TAPPI J.*, *75*, 107–111.

14. Holland, C.D. (1975). Fundamentals and Modelling of Separation Processes. Prentice Hall Inc., Englewood cliffs, New Jersey.

15. Itahara, S., & Stiel, L.I. (1966). Optimal Design of Multiple Effect Evaporators by Dynamic Programming, *Ind. Eng. Chem. Proc. Des. Dev.*, *5*, 309.

16. Kern, D.Q. (1950). Process Heat Transfer, McGraw Hill.

17. Lambert, R.N., Joye, D.D., & Koko, F.W. (1987). Design calculations for multiple effect evaporators-I linear methods, *Ind. Eng. Chem. Res.*, *26*, 100-104.

18. Mathur, T.N.S. (1992). *Energy Conservation Studies for the Multiple Effect Evaporator House of Pulp and Paper Mills*. Ph.D. Dissertation, Department of Chemical Engineering, University of Roorkee, India.

19. Miranda, V., & Simpson, R. (2005). Modelling and simulation of an industrial multiple effect evaporator: tomato concentrate. *J. Food Eng.*, *66*, 203–210.

20. Nishitani, H., & Kunugita, E. (1979). The optimal flow pattern of multiple effect evaporator systems. *Comp. Chem. Eng.*, *3*, 261-268.

21. Pacheco, C. R. F., Cézar, C. A., & Song, T. W. (1999). Effect of the solute concentration on the performance of evaporators. *Chem. Eng. Proc.*, *38*, 109–119.

22. Prost, J. S., Gonzålez, M. T., & Urbicain, M.J. (2006). Determination and correlation of heat transfer coefficients in a falling film evaporator. *J. Food Eng.*, *73*, 320–326.

23. Radovic, L.R., Tasic, A.Z., Grozanic, D.K., Djordjevic, B.D., & Valent, V.J. (1979). Computer design and analysis of operation of a multiple effect evaporator system in the sugar industry, *Ind. Eng. Chem. Proc. Des. Dev*, *18*, 318-323.

24. Rao, N. J., & Kumar, R. (1985). Energy Conservation Approaches in a Paper Mill with Special Reference to the Evaporator Plant. *Proc. IPPTA Int. Seminar on Energy Conservation in Pulp and Paper Industry*, New Delhi, India, 58-70.

25. Ray, A.K., Rao, N.J., Bansal, M.C., & Mohanty, B. (1992). Design Data and Correlations of Waste Liquor/Black Liquor from Pulp Mills", *IPPTA J.*, *4*, 1-21.

26. Schmidl, G. W., & Frederick, W. J. (1999). Controlling soluble scale deposition in black liquor evaporators and high solids concentrators. *Internal Report, IPST.*

27. Stewart, G., & Beveridge, G. S. G. (1977). Steady State Cascade Simulation in Multiple Effect Evaporation. *Comp. Chem. Eng.*, *1*, 3-9.

28. Süren, A. (1995). *Scaling of black liquor in a falling film evaporator,* Master Thesis, Georgia Institute of Technology, Atlanta, GA.

29. Xu, L., Wang, S., Wang, S, & Wang, Y. (2004). Studies on heat-transfer film coefficients inside a horizontal tube in falling film evaporators. *Desalination*, *166*, 215-222.

30. Xu, L., Ge, M., Wang, S, & Wang, Y. (2004). Heat-transfer film coefficients of falling film horizontal tube evaporators. *Desalination*, *166*, 223-230.