Optimization of multistage cross current extraction by iterative dynamic programming

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Abstract: In this study the optimal extraction of lignin and solvent distribution in a multistage cross current extraction cascade system is treated for the solution of constrained non-linear maximization problem. The objective was to simultaneously minimize the solvent loss and maximize solvent convergence in a finite cascade of extractors. Different possible loops are calculated through iterative dynamic programming, and the loops are restricted to certain objective functions. An analytical solution is developed which enables the amount of the make-up solvent to be chosen such that the total profit is maximized. The tabular results are represented on a general graph, giving the optimal combination and number of stages for different solvent distribution conditions.

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1. Introduction

Multiple optimum solutions of a multistage problem are already employed by the chemical and biochemical industries [1-3]. In the chemical manufacturing processes many criteria exist for the selection of the best optimum conditions under which to design and operate a project. It may be desired to increase the yield of final product, or to maintain its quality above a certain minimum, or more generally it is desired to increase the sales value and to reduce the operating costs for any operation [1,4]. An established practice to optimize the multistage problems is to apply the dynamic programming technique [5]. In dynamic programming various possible decisions are made at each individual stage so as to reduce the expected cost. The serial computation is employed in this optimization approach in logic that the decisions of the next stage are dependent upon the results of the previous stages involved [4, 5]. A mathematical model needs to be developed for such a process involving an objective function, which deals with the variables and the constraints for each individual stage. The constraints are subjected to certain inter stage dependencies.

Lagrange multipliers are traditionally used for constrained external problems. However, the Lagrangian multipliers method involves the differential functions and the multiplier approach becomes computationally unmanageable. These difficulties can be overcome by the use of dynamic programming. In dynamic programming multivariable optimization problems are solved individually into a series of one variable optimization problems each of these involving a small number of total variables. The individual one variable problem may be solved using standard methods of differential calculus or using simple search procedures.

In literature quite, a few applications of dynamic programming to chemical engineering problems are being reported. Aris et al., (1961) has solved problems concerned with reactor design and a problem of cross-current extraction cascade [6]. Similarly, Nemhauser et al., (1961) utilized dynamic programming to study the optimization of separation processes in detail [7]. In a study, Bellman et al., (1961) has proposed the basic theory of dynamic programming [8] by studying multistage decision processes in detail. Later, Rudd & Blum, (1962) extended the dynamic programming approach of Aris et al., (1961) and determined the optimum operating conditions for cross-current extraction with product recycle. Their work involved the iterative table entries and back substitutions [9]. These concepts were implemented on a multistage recycle system with recycle back to the first stage in detail by using the same dynamic approach by Van Cauwenberghe, et al., [10]. Optimization of multistage separation processes was explored by Nemhauser (1963) by using dynamic programming approach [7, 11, 12], they reported the mass transfer effect in solid separation problems in particular. Hence the use of dynamic programming is appropriate for the design in the chemical industry where the objective function for a complicated system can often be obtained by dividing the overall system into a series of stages. Optimizing the resulting simple stages can lead to the optimal solution for the original complex problem.

The present work uses a similar approach for analyzing the solvent distribution problem in the case of a cascade of four equal-sized extraction tanks. Based on our previous research Rashid et al., [13], it is evident that due to high efficiency of pyridinium formate ([Py][For]) a protic ionic liquid i.e. (≥ 90 % lignin extraction), it is economical to use it for further extraction cycles pertaining to the make-up solvent, in order to achieve more economical utilization of the solvent. All possible considerations have been taken into account and the final design choice of optimal solvent distribution can be made from a purely economic point of view. For this case the solvent distribution in a multistage cross current extraction cascade system is treated for the solution of constrained non-linear maximization problem. Different possible routes are calculated through iterative dynamic programming, and the routes are restricted to certain objective functions. In this particular arrangement, an analytical solution is developed which enables the optimum route and the amount of the make-up solvent to be chosen such that the total profit is maximized.

2. Materials

The starting material used for the current study include, pyridinium formate (synthesized as previously established procedure Rashid et al., [14]), Fresh biomass feed empty fruit bunch (EFB) samples were supplied by FELCRA, Nasaruddin Oil Palm Mill, Bota, Perak, Malaysia and stored at ≈ 5 °C. The raw biomass was washed with 2 % detergent solution in order to remove oil and greases. The washed biomass samples were dried under natural sun light for 24 hrs before being grinded and crushed using power cutting mill (Pulverizette 25). Subsequently, the grinded biomass samples were sieved to attain the smallest particle sizes of (0.1-0.3mm). All the chemicals were used as received. Triple distilled water was used for all washing purposes.

2.1 Methodology

2.1.1 General Schematics and Concept of Cross Current Extraction Cascade

The solvent [Py][For] and biomass feed continuously pass through different extraction stages in order to achieve the desired specification which is the solvent should achieve the saturation level and the biomass is nearly free of lignin. In chemical industry a common interest is the optimization of an objective function by the most suitable choice of the independent design variables. An objective function can be defined in which the individual costs or profits from each sub-unit in the complex process may be summed and the total optimized with respect to the design or operating variables using the common optimizing techniques [3, 4]. These concepts can be employed for the efficient concept of bio refinery which comprises of the efficient separation of oil palm biomass constituents as well as the efficient utilization of the solvent. To evaluate this particular need some sets of experiments were performed to collect data. Experiments up to two iterations containing (stage 1, 2 and 3) were conducted to collect data for efficiency graph (figure 2) and solvent loss in each stage. All the experiments were carried out on optimized conditions obtained from the second order polynomial equation for lignin extraction from our previously published work Rashid et al., [13]. To begin with, it was first considered as the operation

of a single stage (figure 1(a)). The solute "lignin" is to be extracted using fresh solvent "S_o", in a series of "n" extraction tanks with "R_n" and "E_n" be the concentration of solute in the Raffinate and Extract streams leaving the nth stage. Based on this a general scheme of cross current extraction cascade is shown in figure 1(b). The biomass feed "F_o" has been introduced in first extraction stage. The concentration of the biomass feed entering in first stage has comparatively high initial lignin contents "C_o" as compared to second and third extraction stages. The fresh solvent "S_o" due to high extraction efficiency does not achieve the allowable lignin capacity (determined experimentally (0.710 g/ml) Rashid et al., [14]) through first extraction stage so the exit solvent "S_1" of this stage enters in the second stage which has comparatively low lignin contents "C_1".
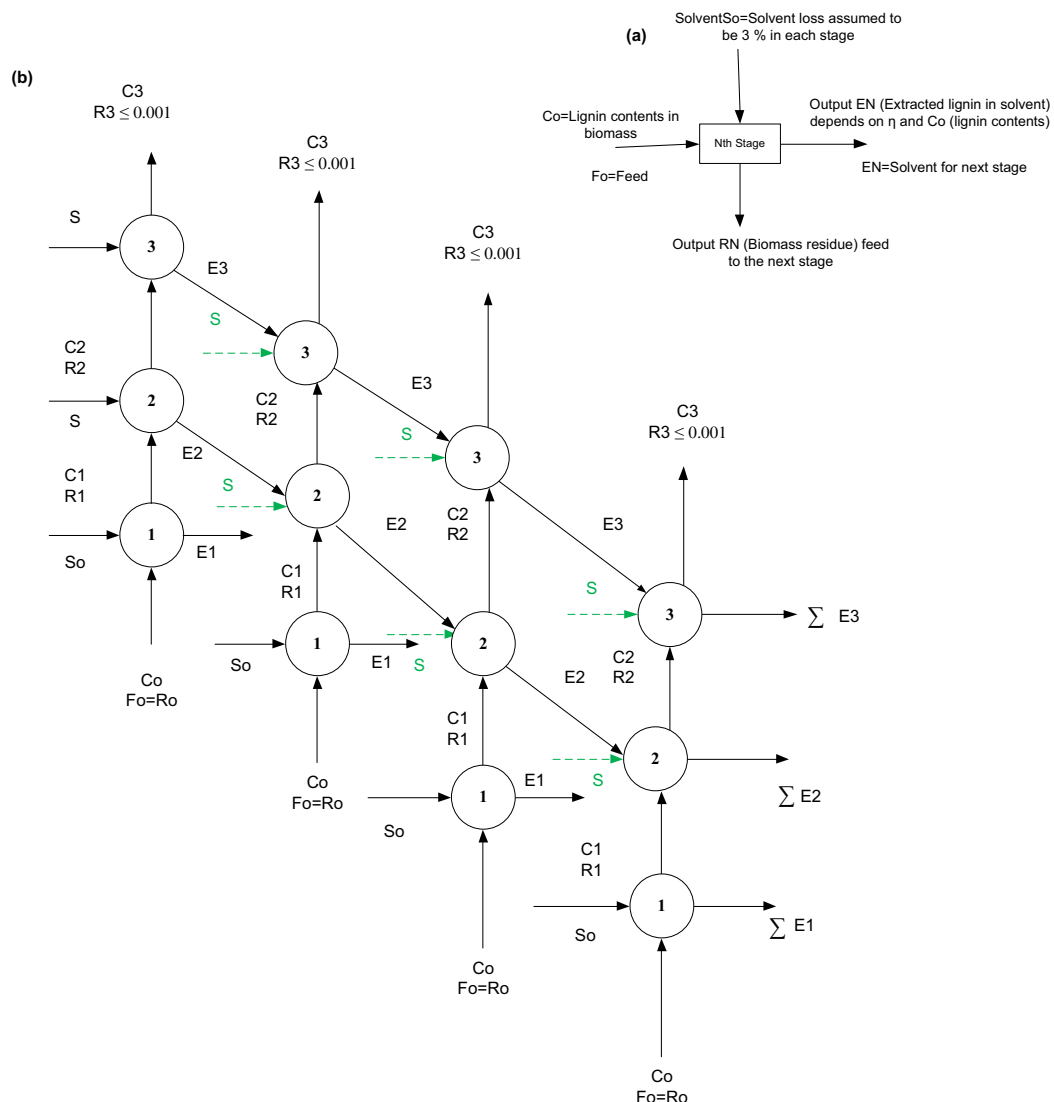

Figure 1. General schematic of cross current extraction cascade.

A certain amount of fresh solvent "S_o" was added to make the established wt % constant. The lower lignin contents "C_1" allows the solvent to extract the maximum remaining lignin contents to "C_2". This continues until the solvent achieves the saturation at 0.710g/ml. The scheme in Figure 1(b) is used for the selection of optimal solvent distribution for lignin extraction considering various routes. Various possible routes are systematically determined for optimal solvent utilization and is presented in results

and discussion section. The mathematical model is formulated as a set of different equations and the details are discussed in the following sections;

2.1.2 Objective Function for Optimal Solvent Distribution

An objective function that serves as a comparison between the various routes suggested above is required. The formulation of the objective function and the notations used are detailed in this section. The extract "E_n" and raffinate "R_n" streams are in equilibrium, then; $E_n = R_n$. The problem is the maximization of a specified function f_n to the final state by an optimal choice of the decision θ_n.

$$E_n = f_n (E_{n-1}, \theta_n) \qquad n = 1, 2, N \tag{1}$$

where "θ_n" is the decisions that may be made for the solvent distribution for the operation of nth stage. The profit "P" obtained in each stage will be taken as being given by the sales income from the lignin extracted less the cost of the extracting solvent [Py][For] used [4, 15]. Thus if "λ_S" is the unit cost of the added solvent (\$/ml) and "λ_L" is the unit income from the sale of the extracted lignin (\$/gm), the profit from the nth stage may be written as;

$$\psi = \lambda_L \sum_{n=1}^{N} E_{n+1} - \lambda_S (V_o) \tag{2}$$

In actuality, the feed as well as the solvent used is all directly fed into the first extractor. There is a drop in the amount of solvent used between the first and second stages and then it rises slowly from the second to the nth stage. This extra amount of solvent used, "V_L" which is independent of the number of extractors in the system, also gives rise to an additional cost, which must be subtracted from the profit, this solvent loss was determined experimentally which is almost 3% for each stage, hence the objective function becomes,

$$\psi = \lambda_L \sum_{n=1}^{N} E_{n+1} - \lambda_S (V_o - V_L) \tag{3}$$

Extraction from Biomass, the equation governing this process is

$$R_n = F_{n*} C_n \tag{4}$$

the initial concentration of lignin in biomass is C_o, then

$$C_{n+1} = [C_n - (\eta_{n+1} * C_n)] \tag{5}$$

The efficiency is a function of E_n (gm/ml), experimentally obtained Figure 2;

$$\eta_n = -0.0021 * E_n^2 - 228 * E_n + 92.455 \tag{6}$$

As the solvent has a saturation limit of 0.710 g/ml; before reaching that limit it has a high efficiency in extracting lignin. In a given stage there is a combination of streams coming from different previous stages. The volume of solvent at stage "N" can be calculated as:

$$V_{n+1} = V_n - (V_n * W) \tag{7}$$

Where "W" represents the % loss of solvent (i.e amount left (ml) of the initial solvent used in each stage) achieved between stages which was determined experimentally. With a material balance for lignin around stage n (where n = 1, 2, 3) we get;

$$E_{n+1} = R_n * \eta_{n+1} / V_n + E_n \tag{8}$$

The solution has been implemented by using solver add-in tool, Microsoft Excel (nonlinear solving method). for the calculation at every stage and also for determining the optimal route to get the maximum profit using dynamic programming analysis of Rudd et al., [9]. The routes are based on the best selection of make-up solvent. The optimum route was considered which gives the minimum solvent loss and maximum lignin removal.

3. Results and discussion

The biomass feed introduced in the first stage enters at comparatively high lignin contents as the second and third extraction stages. The lignin contents of the biomass feed EFB were determined experimentally by TAPPI T222 om-02 method which were found to be 17.8 %. The first stage operates at 17.8 % lignin contents, the second stage operates at 1.42 % lignin contents and stage three operates at 0.11 % lignin contents as ≈ 90% of lignin is being extracted in each consecutive stage. The solvent in the first stage due to its high efficiency does not achieve the allowable lignin saturation ($\sum E_n + 1 \approx 0.710$ g/ml) so the exit solvent is reused in the second and third stages. The lower initial lignin contents in stage two and three allows to remove the maximum remaining quantity of lignin contents but the solvent still has not achieved its saturation concentration. Hence it is economical to use it for further extraction cycles pertaining to the make-up solvent, in order to achieve more economical utilization of the solvent. The systematic determination of routes for optimal solvent utilization is shown in figure 1(b). Stage 1, (figure 1(b)) serves as a root stage of the cross current extraction cascade. The Feed "F_o"has to pass through stage 1 before passing through the stage 2 and 3. The possible routes for solvent saturation are shown in figure 1(b).

The plotted curve is labelled: $Y = -0.0021X^2 - 228X + 92.455$
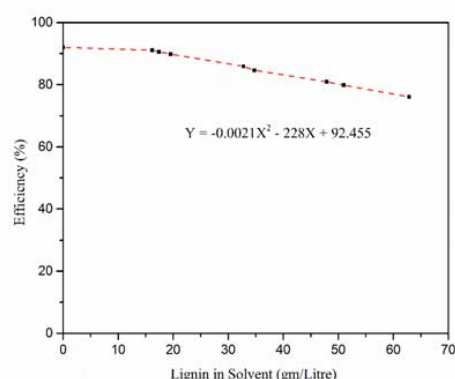
Figure 2. Efficiency vs amount of lignin in solvent.

The routes are based on the best selection of make-up solvent, for example route #1 follows fresh solvent at each stage 1 and no make-up solvent is involved in this scheme, and iterations are run until the constraints ($\sum E_1 \approx 0.710$ gm/ml) are achieved. In route #2 fresh solvent enters at stage 2 and passes downstream through stage 2 with makeup solvent, route # 3 involves the entry of fresh solvent at stage 3 and passes downstream with make-up solvent. The route # 4 involves the fresh solvent from stage 2 and passes through next loop's stage 2 and 3 respectively. The last route #5 involves the fresh solvent enters at stage 1 and 2 and the stage 3 involves make up solvent. All the routes iterations were run until the constraints ($\sum E_n + 1 \approx 0.710$ g/ml) are achieved.

3.1. Selection of Optimal Route

Arranging stages into cascades allows more separation or less energy input than is possible in a single stage. The solution of the mathematical model formulated has been implemented by using solver add-in tool, Microsoft Excel Microsoft Corp. for the calculation at every stage and also for determining the optimal route to get the maximum profit using dynamic programming analysis of Rudd et al., [9]. For the route # 1, fresh feed "Fo" with "Co" lignin content enters at stage1, as the fresh feed contacts with fresh solvent in all stages so there is no drop-in extraction efficiency "ηn". The computations for the route # 1 are given in table 1.

Table 1. Computation for "E_n" for route # 1 for single iteration

Stage	V_o(ml)	F_o (gm)	C_n (%)	E_n (gm/ml)	η_n	R_n (gm)
			Route # 1			
1	80.00	7.894	0.1780	0.01612	0.9201	0.0938

For the route # 2 feed "F_1" after first extraction and reduced lignin contents "C_1" enters in stage 2, fresh solvent enters into stage 2 and follows downstream through stage 2 with make-up solvent. As the solvent is reused for multiple extractions of stage 2 so there is a drop in % extraction efficiency "η_n". This drop in" η_n" can be calculated from equation (6). The computations for the route # 2 are given in table 2.

Table 2. Computation for "E_n" for route # 2 for single iteration

Stage	V_o(ml)	F_o (gm)	C_n (%)	E_n(gm/ml)	η_n	R_n (gm)
			Route # 2			
1	80.00	7.894	0.1780	0.0162	0.9201	0.0938
2	66.85	6.597	0.0142	0.0013	0.9201	0.0074

For the route # 3 the same process is considered but the stage 2 is replaced by stage 3. The calculations for the route # 3 are given in table 3.

Table 3. Computation for "E_n" for route # 3 for single iteration

Stage	V_o(ml)	F_o (gm)	C_n (%)	E_n (gm/ml)	η_n	R_n (gm)
			Route # 3			
1	80.00	7.894	0.1780	0.0162	0.9201	0.09389
3	66.30	6.543	0.0011	0.0001	0.9201	0.00059

For the route # 4 it is considered that the feed "F_1" after first extraction and reduced lignin contents "C_1" enters at stage 2, fresh solvent enters into stage 2 and this solvent enters further into next stage 2 and 3 of the next loop (Figure 1(b)) with make-up solvent. The computations for the route # 4 are listed in table 4.

Table 4. Computation for "E_n" for route # 4 for single iteration

Stage	V_o(ml)	F_o (gm)	C_n (%)	E_n (gm/ml)	η_n	R_n (gm)
			Route # 4			
1	80.00	7.894	0.178	0.0162	0.9201	0.09389
2	66.85	6.597	0.0142	0.0013	0.9201	0.00740
2	66.85	6.597	0.0142	0.0013	0.9215	0.00740
3	66.30	6.543	0.0011	0.0001	0.9186	0.00059

The route # 5 is a conventional one in which the feed passes through stage 1,2 and 3 respectively. The fresh solvent enters at stage 1 and 2 and the stage 3 is working with make-up solvent (table 5).

Table 5. Computation for "E_n" for route # 5 for single iterition

Stage	V_o(ml)	F_o (gm)	C_n (%)	E_n (gm/ml)	η_n	R_n (gm)
			Route # 5			
1	80.007	7.894	0.1780	0.0162	0.9201	0.09389
2	66.857	6.597	0.0142	0.0013	0.9201	0.00740
3	66.309	6.543	0.0011	0.0001	0.8847	0.00059

The solvent loss, E_n and R_n concentrations for all of the routes and stages are determined. In table 6 it can be observed that route # 5 which is the most conventional route gives the maximum convergence but on the other hand it can be seen that the solvent loss and number of stages are maximum per iteration. The optimum cost for the individual stage and process will be higher, hence route # 5 is rejected. From the table 6, the feasible solvent distribution and combination of stages seems to be route # 2, which produces an equal profit as route # 5, showing that route # 2 cascade is a fairly good approximation. This table also shows that the extraction is more complete, if more stages are used. Although this requires more solvent and more loss of solvent consequently.
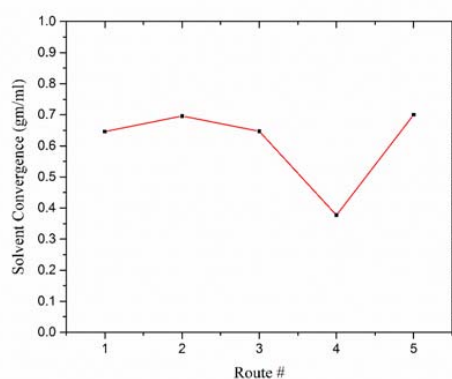

Figure 3. Solvent convergence plot for various routes.

It can be suggested that, in order to attain optimal operation fresh solvent should be utilized in the first stages and should be reused during further downstream. From table 6 it can also be concluded that route #1 requires less solvent and hence the solvent loss is minimum, but on the other hand the raffinate concentration in the exit stream is comparatively higher than other routes which is also another parameter to consider for bio-refinery concept. Hence the selection of the route is dependent on the concentration of R_n for lignin extraction as it should be minimum in the remaining biomass residue.

Table 6. Overview of various studied routes

Cascade Route	Convergence E_n (gm/ml)	Solvent Loss (ml)	Profit ($)	No.of Iterations	Stages Iteration
Route # 1	0.646	96	108	10	4
Route # 2	0.696	176	113	10	8
Route # 3	0.647	176	107	10	8
Route # 4	0.377	168	88	10	8
Route # 5	0.700	255	113	10	12

From figure 4 it is clear that the profit is a function of solvent loss as greater the solvent loss lesser is the profit achieved [10]. Conversely it is evident from figure 4 that solvent convergence is higher for the longer route as more lignin is available to be extracted and hence the profit is increased. Pertaining to this fact it can be observed from figure 4 that the route # 5 is giving maximum solvent loss but instead the profit is found to be increased.
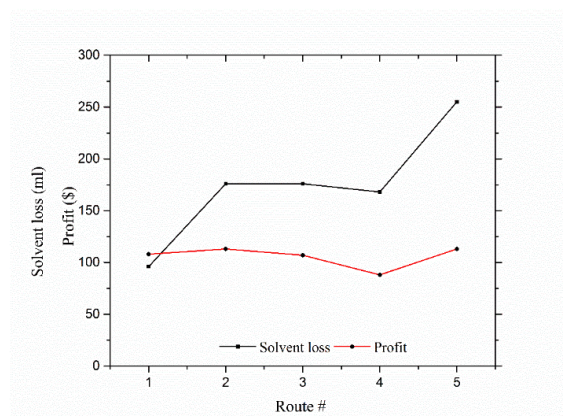
Figure 4. Profit and solvent loss relation for lignin crosscurrent extraction cascade.

This may be due to the reason that more lignin is available to be extracted in route # 5 and hence the profit is greater, but this increase in profit is very small that it cannot be considered on the cost of the solvent loss. Hence, from figure 3 and figure 4 it is evident that route # 2 and 3 are comparable but due to higher convergence achieved by route # 2, it is selected as optimum route.

4. Conclusıons

A multistage cross current extraction cascade system is treated for the solution of constrained non-linear maximization problem for optimum solvent distribution. Different possible routes are calculated through iterative dynamic programming and the route which gave maximum solvent saturation with minimum solvent losses was suggested for further large-scale applications. This is of a great significance from an industrial viewpoint, as a significant amount of solvent and material and energy could be reduced if such a process is implemented.

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