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Analysis of the Factors Affecting the Oil Recovery from Oilseeds in a Multistage Crosscurrent Leaching Process

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ABSTRACT

The analysis of the total oil recovery from oilseeds in a multi-stage crosscurrent leaching process is presented. The model created using mass balance equations for three stages was used to analyze the factors affecting total oil recovery, T_o*. The key performance factors (raw meal feed flow rate, solvent feed flow rate in each stage, solvent distribution factor for each stage, percent oil in the raw meal (*P_{oil}*), solids to solution ratio in the underflow (N), solvent to solids ratio (SSR), solvent to oil ratio (SOR) and wash ratio (WR) were varied while assessing* T_o*. While* SSR_i *and* SOR_i *ranged from 0.1 to 10 and from 0.1 to 100, respectively, N varied between 1 and 7. The variation of* T_o *with the key variables were fitted using polynomial functions, whose degree increased with N at fixed values of solvent feed flow rate (*V_S*) and raw meal flow rate (F).* T_o *was the highest when* α_1 *and* α_2 *ranged from 0.05 to 0.3 and 0.05 to 0.2, respectively. The highest percent contribution to* T_o *was from Stage 1 (50 - 80%) followed by Stage 3 (10 to 40%). Optimal settings for the newly introduced parameters (*SSR_i*,* SOR_i *and* α_1*) and solvent distribution factors are recommended for maximizing the total oil recovery.*

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INTRODUCTION

Leaching as a unit operation, multi-stage arrangements and solvent distribution

Unit operations are the basic processes involved in the raw material reparation, separation, and purification of products. These include distillation, leaching, absorption, adsorption, evaporation, drying, crystallization, among others, which are implemented according to the nature of the materials involved. Leaching is used for separation, which depends on solubility difference between the inert carrier solid and the solute held by the solids. The aim of leaching is to remove the target component (solute) by dissolving it in a convenient solvent. Thus, the desired component in a solid phase is separated by contacting the solute rich solid with the solvent. The desired component leaches from the solid into the solvent and the two phases are separated. The desired component is then recovered from the liquid phase by further processing. Leaching, also known as solid-liquid extraction is a mass transfer phenomenon. The separated solution is commonly named extract or overflow, and the inert solid with solution retained is named raffinate or overflow. Equipment is available in the market to conduct leaching under batch, semi continuous, or continuous operating conditions. However, a wider knowledge of the key factors affecting the efficiency of leaching process is lacking, especially for cross current operations.

This separation process can be done in a single or multiple stage connected in series using countercurrent or crosscurrent mode of operation. In this study, a multistage crosscurrent mode was simulated intensively, using mass balance equations. Fresh or spent solvent (containing a small amount of solute) can be added to each stage. This scheme is rarely used in a commercial process because of the large volume of solvent required and low concentration of solute in the extract. Also, lack of knowledge on solvent distribution between the stages hinders its industrial application. The solvent feed can be by equal amounts across the stages or specifically distributed across stages based on the pre-determined factors, as presented in this study.

Principles of leaching and its applications in the process industry

One of the major applications of leaching is in the extraction of oil from oil seeds using a solvent. Various methods for recovering oil from the seeds have been investigated. Solvent extraction (or leaching) is the most widely used technique, due to high efficiency in oil recovery (90 to 98%). The major disadvantage of solvent extraction technique is its high energy input during mixing and toxicity of solvent. The need for expensive equipment for separating the oil and the solvent is another added cost in large scale plants. This has led to the development of other methods such as enzyme-based techniques (Sharma *et. al.*, 2002).

Other uses of the leaching process include the metal industry for removing minerals from ores (acid solvents) and the sugar industry for removing sugar from beets (water as a solvent). The oilseeds industry uses leaching for removing oil from soybeans, ground nuts, sunflower, etc., (where n-hexane (Baskar *et. al.*, 2019; Manyele, 2008) petroleum ether (Manyele, 2008), hexane or similar organic solvents are used). Some common examples for leaching in food industry include extraction of soluble compounds from roasted and ground coffee in the soluble coffee production, and extraction of proteins from soybeans in the production of isolated soybean protein. A significant application of leaching processes is orientated to the environment, as is the removal of heavy metals (lead and cadmium) from contaminated soils using EDTA (Hao *et. al.*, 2017), the recovery of zinc from alkaline batteries waste using microwave or ultrasound assisted leaching (Sadeghi *et. al.*, 2017) and a process for recycling tin, bismuth and copper from E-wastes and wastewater using ammonia leaching and hydrochloric acid leaching (Jeon *et. al.*, 2017).

Leaching is usually operated at an elevated temperature to increase the solubility of the solute in the solvent which can add to energy costs. Furthermore, regardless of all other factors, a mixing temperature near 60-65°C shows the best yield values (Baskar *et. al.*, 2019; Al-Hamamre, 2014; Khasawneh, 2017; Sheet, 2018; Lubls, 2013). The diffusion coefficient of oil from the solid matrix into the solvent will be expected to increase with rise in temperature and this will also improve the rate of extraction. In some cases, the upper limit of temperature is determined by secondary considerations, such as the necessity to avoid enzyme action during the extraction of sugar.

The mass balance equations involve three component (solid, solvent and solute) systems. The feed is usually prepared by grinding or chopping and then mixed with a liquid solvent. The desired material dissolves (to some extent) and leaves the solid matrix when the liquid is drawn off as overflow. The solids containing some retained solution are then removed as underflow, leading to unrecovered oil and solvent losses. The mass balance equations must account for oil in all streams. In general, the amount of solution in the underflow depends on the properties of the solution, which are dependent on its composition. The amount of solute present causes "stickiness" of the solution. As a result, "draining data" is typically collected, to relate the solids-to-solution ratio to the composition of the solution. In the stages, comprising of the mixer and separator, the contact times can be made long enough for any desired approach to equilibrium. Since mixer-separator assemblies take much of floor space, they usually are employed in batteries of at most four or five units of a large variety of compact equipment, like tower arrangements. The relations between their dimensions, the operating conditions, and the equivalent number of stages forms the key design features.

Solvent-related factors affecting leaching operation performance

The factors influencing extraction rate and total oil recovery include: entering feed flow rate and composition, choice of liquid solvent(s) (Baskar *et. al*., 2019; Manyele, 2008) operating temperature and pressure, etc. The solvent feed flow rate, on the other hand, affects the total oil recovery by making solvent available for the solute to dissolve. The solvent must wet all the solids in Stage 1, Thus, the solvent flow rate is related to both solid matrix and the oil it contains. The solvent flow rate into each stage was further expressed in the form SSR and SOR. Thus, the optimal range of solvent distribution between the three stages is critical as it determines the range of SSR and SOR. The nature of solvent (n-hexane, ethanol or petroleum ether) (Manyele, 2008; (Lubls, 2013; Abu-Arabi, 2000) and particle size also affect the total oil recovery (Baskar et. al., 2019; Manyele, 2008a; Manyele, 2008b). The liquid chosen should be a good selective solvent and its viscosity should be sufficiently low for it to circulate freely (Khasawneh, 2017). Generally, a relatively pure solvent will be used initially, but as the extraction proceeds the concentration of solute will increase and the rate of extraction will progressively decrease, first because the concentration gradient will be reduced, and secondly because the solution will generally become more viscous. Other solvents used for oil extraction include benzene and carbon tetrachloride (Lubls, 2013) and isopropanol (Abu-Arabi *et. al*., 2000). The performance of a number of solvents (diethyl ether, petroleum ether, hexane, acetone and ethanol) has also been evaluated (Abu-Arabi *et. al*., 2000). Methyl ethyl ketone has also been used as a solvent, results of which were compared to other solvents (Abu-Arabi *et. al*., 2000). Thus, the purity of the solvent used in the leaching process poses another challenge.

The SSR and SOR have a strong effect on total oil recovered. The nature of solvent, temperature, and SSR affects extracts from grape pomace (Pinelo *et. al*., 2005). The SSR between 1 and 5 maximized the amount of phenolic extracts (Pinelo *et. al*., 2005). The analysis of the effect of SSR on total oil recovered was aimed at preventing excessive solvent use. Similar study was carried out by varying the feed to-solvent ratio (Laitinen, 1999). Since leaching operation uses solvent to extract oil trapped in the inert solid matrix, the SSR and SOR need to be optimized (Osman *et. al*., 2013).

Particle size influences the extraction rate in a number of ways. The smaller the size the greater is the interfacial area between the solid and liquid, and therefore the higher is the rate of transfer of oil to the solvent and the smaller is the distance the solute must diffuse within the solid. On the other hand, with a very fine material the circulation of the liquid is impeded, separation of the particles from the liquid and drainage of the solid residue can be more difficult. The small particle size is preferred as it allows for a large interfacial area between the solid and liquid. Agitation of the solvent increases the eddy diffusion and therefore increases the transfer of materials from the surface of the particles into the bulk of solution (Kaskar *et. al*., 2019).

If diffusion of the solute through the porous structure of the residual solids is the controlling factor, the material should be of small size so that the distance the solute has to travel is small. On the other hand, if diffusion of the solute from the surface of the particles to the bulk of the solution is the controlling factor, a high degree of agitation of the fluid is required. Agitation of the solvent-raw meal mixture is important because this increases the eddy diffusion and therefore the transfer of material from the surface of the particles to the bulk of the solution. Intensive agitation of suspensions of fine particles prevents sedimentation and more effective use is made of the interfacial surface.

The focus and findings of former studies on leaching operations

Table 1 summarizes the literature on leaching problems solved by other researchers, methodology used and the key results obtained. The summary further shows how the results were presented, and finally the contribution of those studies to the body of knowledge on design and operation leaching operations.

Table 1: Methodology, key results and analysis from the past research on oil exaction from oilseed and fish meal.

Problem addressed	Extraction Methodology used	Key results	Presentation of results	Citation
Extraction of Jojoba oil by leaching process	Multifunctional extraction	Effect of solvents (n-hexane, diethyl ether, and petroleum ether) in oil extraction	Compared solvents used in relation to oil yield, operating cost and energy consumption	(Khasawneh, 2017)
Leaching of oil from tuna fish liver by using methyl-ethyl ketone	Distillation method with different solvents (acetone, n-hexane, benzene, carbon tetra chloride)	Determined the maximum yield of fish liver oil at different temperatures	Maximum yield of fish liver oil obtained in relation to temperature and duration.	Lubls, 2013)
Effected of leaching temperature and time in terms of yielding of fish oil by the fraction of lipid	Leaching utilizing different solvents (hexane and isopropanol)	Effect of solvent type temperature and time; optimized the parameters based on oil yield	Multiple regression analysis and the second-order model fitted for each response. Time and temperature had a significant impact on oil yield.	(Abu-Arabi *et. al*., 2000)
Evaluation of oil production from safflower seeds using different kinds of solvents	Applied classical solvent extraction, Soxhlet and ultrasonic extraction method with different solvents (diethyl ether, petroleum ether, hexane, acetone and ethanol)	Identify the best methods in terms of time and solvent consumption	The types of solvent and extraction methods were compared in terms of process viability and the compositions of final product	(Takadas, 2017)

Simulation on the extraction of sunflower oil using supercritical carbon dioxide	Used conventional vacuum distillation and solvent extraction methods at a commercial scale	Yield of sunflower oil produced using SC-CO_2 in relation to methods used	Differentiating the conventional vacuum distillation and solvent extraction methods against SC-CO_2 on extracting thermolabile molecule from oil bearing seeds	(Filho, 2012)

In Table 1, the methods used for oil extraction include leaching (Abu-Arabi *et. al*., 2000; Khasawneh, 2017; Lubls, 2013) hydraulic pressing, Soxhlet extraction (Takadas, 2017) and supercritical carbon dioxide (Nimet, *et. al*., 2012). The key results from these studies include maximum yield of oil at different operating conditions and temperatures, effect of the type of solvents used (Abu-Arabi *et. al*., 2000; Khasawneh, 2017) effect of mixing time and solvent consumption (Takadas, 2017).

The main objective of this study was to develop the crosscurrent computer model and simulate the effect operating parameters on the total oil recovery. To achieve the results, the following tasks were accomplished: development of a computer model and simulation of the leaching operation for a multi-stage cross-current system; establishment of the range of different solvent related ratios: S/F (solvent to feed ratio), *N* (solids to solution ratio), solvent-to-oil ratio (SOR_i), solvent-to-solids ratio (SSR_i) in the respective stages and for a wide range of leaching plant sizes and operating conditions; relating different solvent-related ratios with the total of oil extracted or plant efficiency; and, studying the variation of oil concentration in the overflow with solvent feed flow rate.

METHODS AND MATERIALS

Assumptions made to simplify the model

To enable modelling and simulation of a leaching system several assumptions were made to make the system ideal.

Setting the feed flow rates for raw meal, *F*, and solvent, V_{s1}, is a challenge that need to be addressed. This is because, the solvent fed must "wet" the solid (fill pores, etc.) and so will typically pick up more liquid in the 1st Stage than in subsequent stages. Together with the variation of solvent flow rate with solute concentration in the first and subsequent stages, the contribution of oil recovered in the first stage to the total oil recovered was analyzed, as well as in other stages. The solvent fed was assumed to be sufficient to dissolve the solute out of the solids in the first stage, where fresh raw meal is fed, and that the successive stages has also a sufficient solvent flow rate to dilute the solution and deviate most of oil into the overflow streams.

Mixing of the solid and the solvent is critical. Typically, perfect mixing is assumed, as is the idea of an equilibrium stage, that is, the solid and liquid phases on each stage are assumed to be in equilibrium. These assumptions imply that all the liquid within the stage has the same composition and so the overflow and the liquid carried in the underflow are identical, that is $Y_i = X_i$. This is known as the *uniform solution* assumption which leads to a linear equilibrium curve.

It was further assumed that there is no adsorption of solute onto the solid particles, such that after coming in contact with solvent in the first stage, all of the solute is dissolved into the solvent, so that $Y_i = X_i$.

In an ideal equilibrium leaching stage, the overflow is assumed to be free of carrier solid, that is, none of the solid is dissolved. Hence, a constant flow rate of the solids through the stages, $B_i = B_o$, was assumed. To determine how much liquid leaves entrained with the solids in the underflow, the assumption was constant solution underflow, which implies that every stage has the same, fixed ratio of solution to solid in the underflow stream.

Therefore, the set values of the ratio of solids-to-solution, *N*, is constant and applies to all stages. Since the amount of inert solids is unchanged, this assumption leads to the fact that same amount of solution is held in the underflow from each stage.

Model Development and Mathematical Formulations

Process flow sheet for crosscurrent leaching operation

The flow of the feed solution and solvent from stage to stage was selected to be crosscurrent. In multi-stage crosscurrent extraction the raffinate is successively contacted with fresh solvent as shown in Figure 1. In multistage crosscurrent leaching, the underflow stream from Stage 1, W_1, carries the solids and becomes the feed stream for Stage 2 and W_2 stream from second stage become the feed stream for the third stage. In each stage, the mixture is contacted with a fresh stream of solvent. The overflow solution carrying the oil, exits from each stage, as the products of varying oil content. The solute concentration in the underflow is denoted as *X*, while that in overflow is denote as *Y*, both changing from one stage to another. The stages are numbered in the direction of solids flow.

For a single stage, using Soxhlet extractor connected with solvent and oil recovery, ratios like kg oil recovered per kg solvent used, percent of volatile material driven off from the raw meal and kg oil recovered per kg volatile matter driven off during extraction in a fixed contact time and particle size of soybean, groundnuts and sunflower seeds have been reported (Manyele, 2008a, 2008b).

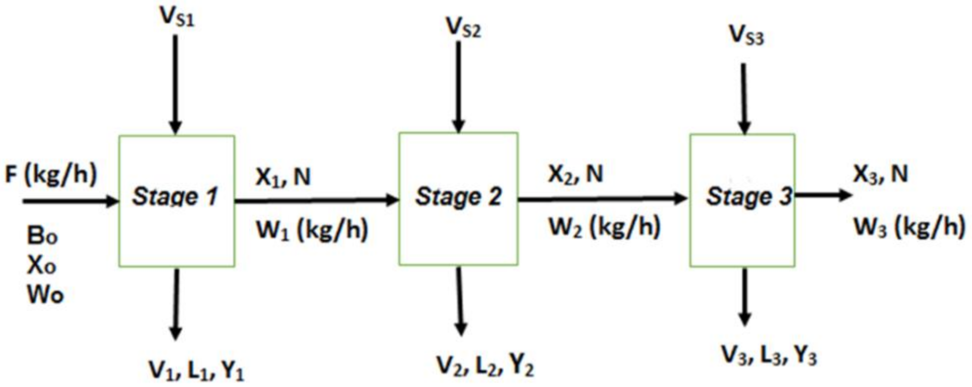


Figure 1: Simplified process flow diagram for crosscurrent leaching operation with three stages to enable mass balances.

Mathematical model formulation

The oil content in the raw meal, S_o, was given as per (1):

$$S_o = X_o W_o = P_{oil}\, F \qquad (1)$$

where X_o is the oil concentration in the solution within the raw meal fed to the plant, W_o is the flow rate of the solution, P_{oil} is the percent of oil in the raw meal and *F* is raw meal flow rate or plant capacity. In case the raw meal feed is free of the solvent, then $X_o = 1.0$ and $W_o = S_o$. Defining B_o as the inert solids flow rate in the raw meal, then the feed point F_p, is given by Cartesian coordinates (X_o, N_o), where

$$N_o = \frac{B_o}{W_o} \qquad (2)$$

Similarly, for the solution and solids exiting the i^{th} stage and assuming that solids are inert and do not dissolve in the solvent, $B_i = B_o$, then the oil concentration in overflow solution leaving the *i*th stage, is given as:

$$Y_i = \frac{L_i}{V_i} \qquad (3)$$

where L_i is the amount of oil extracted or quantity of oil in the overflow solution with flow rate given as V_i. In the underflow, the concentration of the solution leaving the *i*th stage, is defined as:

$$X_i = \frac{S_i}{W_i} \qquad (4)$$

where S_i is the amount of oil in underflow stream. Since the solution in the overflow has the same properties as that in the underflow, then $X_i = Y_i$, which leads to:

$$\frac{L_i}{V_i} = \frac{S_i}{W_i} \qquad (5)$$

With the total solvent flow rate, Q_s, the distribution of solvent can take different forms such as: equal flows across the stages, leading to (6):

$$V_{si} = \frac{Q_s}{n} \qquad (6)$$

where n = number of stages; or implementation of solvent distribution factors, α_i ($i = 1, 2, \ldots, n$) as shown in Figure 2. In this study, the flow rate distribution was optimized for maximum total oil recovery by varying V_{si}, or α_i, based on the relationships between the three factors:

$$\alpha_1 + \alpha_2 + \alpha_3 = 1 \qquad (7)$$

and the determination of solvent feed rate across the stages using the set values of α_I, as per (8):

$$V_{si} = \alpha_i Q_s \qquad (8)$$

Based on solution and solute balances across an *i*th stage,

$$W_{i-1} + V_{si} = V_i + W_i \qquad (9)$$

and

$$\begin{aligned} Y_iV_i = X_{i-1}\,W_{i-1} - X_iW_i \\ + V_{si} \\ = V_i + W_i \end{aligned} \qquad (10)$$

Using (10) and also by $N = B_o/W_1$, and the equilibrium relationship, $Y_i = X_i$, the solute concentration in the overflow from a given stage is related to the incoming underflow solution concentration according to (11):

$$Y_i = X_{i-1}\left(\frac{W_{i-1}}{W_{i-1} + V_{si}}\right) \qquad (11)$$

From (11), if N and B_o are constant throughout the stages, then:

$$Y_i = X_{i-1}\left[\frac{\frac{B_o}{N}}{\frac{B_o}{N} + \alpha_i Q_s}\right] \qquad (12)$$

and therefore,

$$Y_i = X_{i-1}\left[\frac{B_o}{B_o + \alpha_i N Q_s}\right] \qquad (13)$$

(13), also defined as operating line equation, relates the overflow solution concentration from the *i*th stage with the solute concentration in the underflow solution from the previous stage based on mass balances. In (13), a shift is applied on the stage numbers such that Y_{i+1} is related to X_i.

While the oil extracted from the *i*th stage is given by (14):

$$L_i = Y_i \times V_i \qquad (14)$$

the unrecovered oil remaining in the underflow leaving the nth stage is given as per (15):

$$R_n = X_n \, . W_n \qquad (15)$$

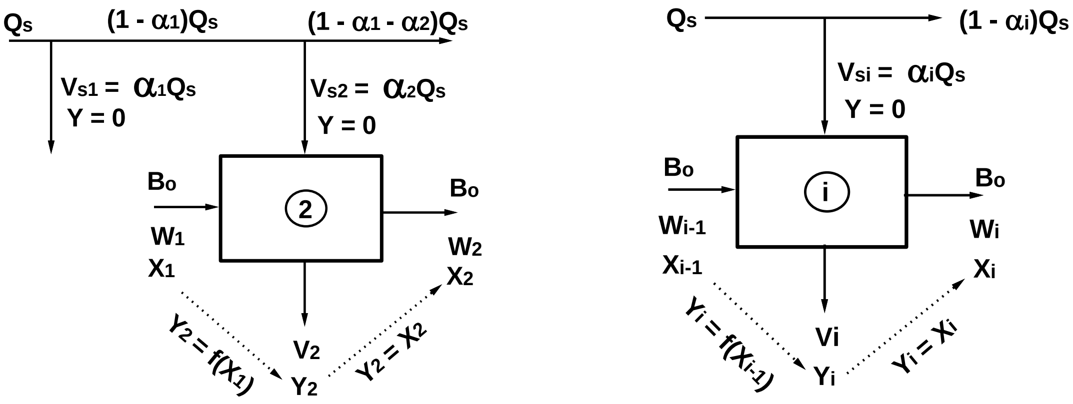
The unrecovered oil is normally left in the solution adhering to the treated solids, which can be traced in the underflow exiting the last stage (in this case 3rd stage) or $n = 3$ into (15), leading to (16):

$$R_3 = X_3W_3 \qquad (16)$$

The percent unrecovered oil or oil lost P_{OL} is given based on the total oil fed in the plant with raw meal as per (17):

$$P_{OL} = 1 - E_R = \left(\frac{X_3W_3}{\mathrm{P_{oil}}F}\right) \qquad (17)$$

where E_R is the percent oil extraction or recovery efficiency.



Specific case for stage 2 Generalized case for the i^{th} stage.

Figure 2: Relating solute concentration between the incoming underflow solution and the extracted solute in the overflow across the i^{th} stage for crosscurrent leaching.

If the overflow streams are combined, for $n = 3$, then the total oil recovered, from all stages, T_o, is given by (18):

$$T_o = \sum_{i=1}^{n} Y_i V_i = \sum_{i=1}^{n} L_i \qquad (18)$$

and the overall solution concentration is thus given as per (19):

$$Y_{ov} = \frac{\sum_{i=1}^{n} Y_i V_i}{\sum_{i=1}^{n} V_i} = \frac{\sum_{i=1}^{n} L_i}{\sum_{i=1}^{n} V_i} \qquad (19)$$

The oil extraction efficiency was estimated based on the percent oil content in the raw meal, that is:

$$\eta_{po} = \frac{T_o}{P_{oil}\, X\, F} X100\% \qquad (20)$$

or, based on total feed flow rate, as per (21):

$$\eta_{RM} = \frac{T_o}{F} X100\% \qquad (21)$$

The percent contribution of the i^{th} stage to the total oil recovered is given as per (22):

$$P_{cont} = 100\ X \left(\frac{L_i}{T_o}\right) \qquad (22)$$

Solvent to feed ratio is a key factor determining the performance of the multistage leaching process. For the 1st stage the solvent to feed ratio, S_F is given by:

$$S_F = \frac{V_{s1}}{F} \qquad (23)$$

For the i^{th} stage, the solvent-to-inert solids ratio, S_B, was defined as per (24):

$$S_B = \frac{V_{s1}}{B_o} \qquad (24)$$

In a given stage, the underflow containing solute (oil), solvent and inert solids from the previous stage are mixed with fresh solvent, V_{si}. Based on this mixture, the solvent fed-to-solute present ratio, S_{SL}, was defined as per (25):

$$S_{SL} = \frac{V_{s1}}{X_{i-1} W_{i-1}} \qquad (25)$$

Another key parameter denoting the separation between overflow and underflow in a given stage is the wash ratio, WR_i. The wash ratio values for the i^{th} stage were determined according to (26):

$$WR_i = \frac{V_i}{W_i} \qquad (26)$$

After mixing the solvent with the underflow, before separation takes place, the total solvent present-to-solids ratio, SSR_i, in the i^{th} stage was defined as:

$$SSR_i = \frac{(1 - X_{i-1}).W_{i-1}}{B_o} \qquad (27)$$

The total solvent present-to-oil ratio in the ith stage, SOR_i was defined in (28):

$$SOR_i = \frac{(1 - X_{i-1}).W_{i-1} + V_{si}}{X_{i-1}.W_{i-1}} \qquad (28)$$

Thus, the total solvent in the underflow exiting the i^{th} stage was determined as per (29):

$$S_u = (1-X_i).W_i \qquad (29)$$

The following equations were developed based on mass balances across an i^{th} stage. The solution balance across the i^{th} stage gives (30):

$$W_{i-1} + V_{si} = W_i + V_i \qquad (30)$$

while the solute balance across the i^{th} stage gives (31):

$$W_{i-1}X_{i-1} + V_{si}Y_{VS} = X_iW_i + Y_iV_i \qquad (31)$$

where Y_{vs} is the solute concentration in the incoming solvent stream. If the solvent is free of solute, then $Y_{vs} = 0$, and

$$W_{i-1}X_{i-1} = Y_iV_i + X_iW_i \qquad (32)$$

For the given stage, i, $X_i = Y_i$, and therefore, (32) leads to:

$$\mathrm{X_i} = \mathrm{Y_i} = \frac{\mathrm{W_{i-1}X_{i-1}}}{\mathrm{W_i{+}V_i}} \qquad (33)$$

Flow Chart for the Simulation Model

Figure 3 shows the flow chart of the multistage crosscurrent leaching model used to simulate the effect of various operating parameters on the total oil recovery. As stated before, three stages were modeled incorporating several input variables starting from the feed characteristics (F, P_{oil}, B_o, X_o and W_o) followed by solvent feed rate variables (Q_s, V_{si}, α_i), and the constant underflow (kg solids per kg solution, N). The mass balance equations were generated to determine the overflow characteristics (V_i, X_i) and the underflow characteristics (W_i, X_i).

A condition was imposed to assess if the total oil was the maximum. When the total oil was not the maximum new solvent distribution factors α_I were selected until the condition was satisfied. At different stages of the model, the new variables describing the operating conditions of the leaching process were determined and tabulated, such as WR_i, SSR_i and SOR_i, as defined earlier.

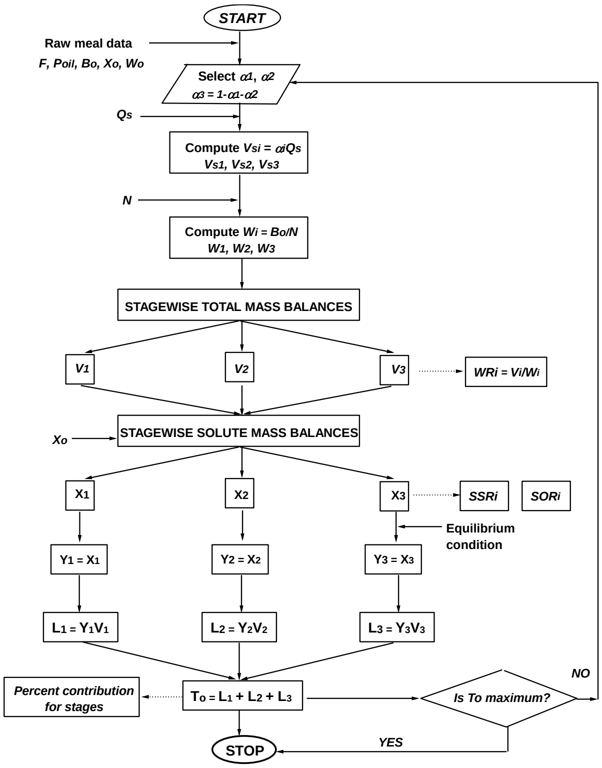


Figure 3: Flow chart for the crosscurrent leaching model implemented in this study.

RESULTS AND DISCUSSION

Variation of Oil Concentration in the Overflow with Solvent Flow Rate

The solute concentration in the overflow from a given stage is an important factor determining the efficinency of the leaching process. It leads to the total oil recovery when the three concentration values are used to determine the oil received in each stage stated in (3), (4) and (5). Figure 4 shows the effect of increasing solvent flow rate into respective stages on concentration of the overflow solution, Y_i. It was observed that Y_i decreases with V_s according to 4th, 5th and 6th order polynomial functions for 1st, 2nd and 3rd stages. The good fit at increasing order of polynomial function from Stage 1 to 3, indicates that Y-V_s relationship increases in complexity at Stages 2 and 3. This can be attributed to the fact that in Stage 1, fewer variables affect Y_1 compared to Y_2 and Y_3. For all solvent flow rates, Y_1 was the highest, while Y_2 and Y_3 were closer especially at higher solvent flow rates. The closer and constant values of Y_2 and Y_3 at higher V_s values indicate that increasing V_s has no effect on the solute concentration in the last two stages.

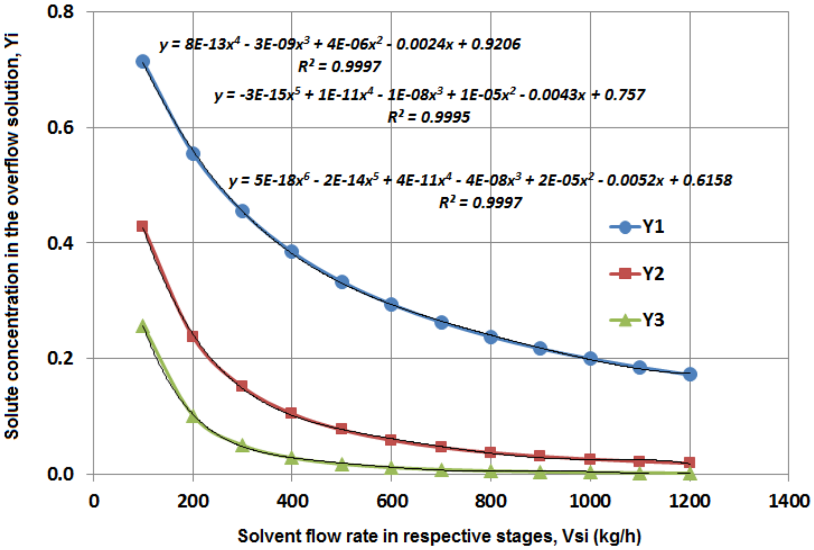


Figure 4: Variation of solute concentration in the overflow solution from respective stages, Y_i, with the equal solvent flow rates, V_{si} ($N = 5$, $F = 1000$ kg/h).

Variation of SSR and SOR with Raw Meal Feed Rate

The operating range for feed and solvent flow rates was assessed by observing SSRi in each stage. When the plant capacity increases, the quantity of inert solids also increases in the system for a given oil content in the oil seed, P_{oil}. Figure 5 presents the range of values of the solvent-to-solid ratio by *SSRi*, considering the variation of percentage in the raw meal oil interring the leaching plant with different raw meal feed flow rate kg/h. Figure 5 is presented on a logarithmic scale for both SSR_i and F, for P_{oil} ranging from 18 to 40%. The SSR_i decreases with increasing F, indicating that more solid enters the stages, lowering the *SSRi*, since the solvent flow rate was kept constant at 300 kg/h across each stage ($Q_s = 900$ kg/h) and $\alpha_i = 1/3$). Results show that P_{oil} has least effect on the SSR_i. Moreover, SSR_3 values were the highest compared to Stages 1 and 2. The higher values of SSR_3 then SSR_1 and SSR_2 can be attributed to the fact that in the last stage, most of the solute has been extracted into streams V_1 and V_2, leaving the solution W_3 with very low concentration of oil (that is very dilute), leaching to high solvent concentration in the stream W_2, and hence, additional solvent, V_{s3}, adds up towards larger amount of solvent present in the Stage 3 mixer and settler, and hence higher SSR_3. This is based on the fact B_o is constant and the ratio *SSR* depends solely on the solvent present. The lowest *SSR* values were observed at $P_{oil} = 18\%$ in Stage 1. Also at various values of P_{oil}, the lowest *SSR* values were observed in Stage 1. This can attribute to the fact that compared to other stages, stage1, handles most of the oil, before part of it is extracted via V_1. Therefore, the solvent fed to Stage 1, forms a thick solution (highest oil concentration) with least amount of solvent in

Stage 1, hence low values of *SSR_1*. At a given *F* and *V_s*, increasing *P_{oil}* should increase *SSR_i*, since *B_o* is taken up by oil, increasing *SSR_i*.

During extraction, a focus is on how much solvent is applied for the given amount of oil in the respective stages. This was expressed using the SOR values. Figure 6 presents the values of *SOR_i* in the three stages of a leaching plant as a function of raw meal flow rate, at constant values of *P_{oil}*. Results show that increasing raw meal flow rate lowers *SOR* values in all stages. Given the total solvent present in the stage, a large raw meal flow rate implies that more oil is present in the stages and hence lower *SOR_i*. Highest values of *SOR* were observed in Stage 3, for all set values of *P_{oil}*. This can be attributed to the fact that in Stage 3, there is lowest amount of oil left that is very dilute solution attributable to decreased amount of solute, after being extracted in Stages 1 and 2 and hence highest *SOR* values.

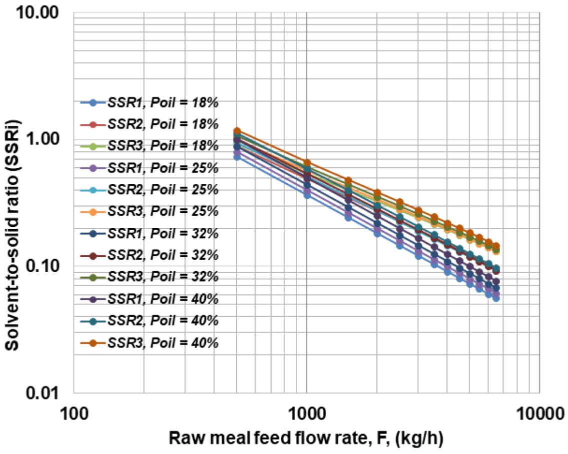


Figure 5: Variation of of solvent to solid ratio with raw meal feed flow rate for different oil content in oilseeds.

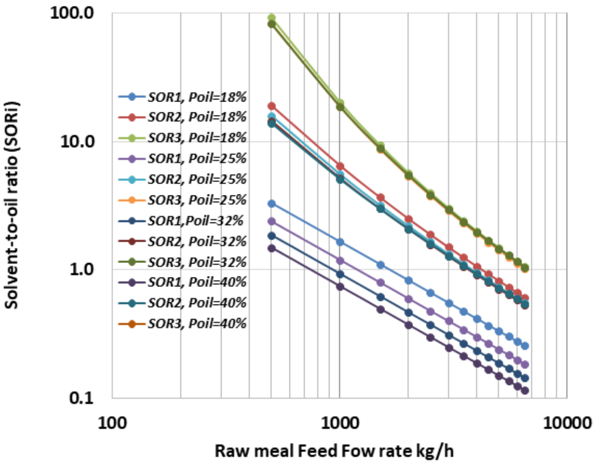


Figure 6: Variation of solvent-to-oil ratio in different stages (*SSR*) with raw meal feed flow rate for different oil-content in the raw meal at *V_{si}* = 300 kg/h.

In Stage 1 on the other hand, the mixer receives the largest amount of oil, and hence, with fixed solvent flow rate, *SOR_i* values were the lowest, corresponding to highest values of *Y_1* and *X_1*. In each stage, there is higher solvent to oil ratio at lower percent oil in the raw meal (*P_{oil}* = 18%) than at higher percent oil (*P_{oil}* = 40%), attributable to presence of smaller amount of the oil in the system at lower *P_{oil}*, leading to a higher *SOR*.

Effect of Solvent Feed Rate on Solvent-to-Solids Ratio (SSR) and Solvent-to-Oil Ratio SOR in each stage

To express the wetting of solids during mixing with solvent, *SSR* values were determined for each stage and for different plant capacities. Figure 7 presents the variation of *SSR_i* with solvent flow rate into respective stages. For all the three stages, increasing *V_{si}* increases the SSR*$_3$* at constant *P_{oil}* or *N*. It was observed that varying *P_{oil}* did not show appreciable effect on *SSR_i*, and that the latter varied strongly with stages. The variation of *SSR_i* with *V_{si}* changed from Stage 1 (where a linear relationship was observed) to Stage 2 where the curves were fitted with quadratic functions. This was attributed to the additional variables in the process, that is, W_1 and X_1. In Stage 3, on the other hand, the relationship between SSR_3 and V_{s3} were fitted using a 3rd order polynomial function, indicating the most complex relationship. This was again attributable to increased number of variables, that is, additional X_2, W_2, Y_3 and N.

The effect of increasing N was observed for SSR_2 and SSR_3. For SSR_1 (in Stage 1), N did not have any effect, basically because mixing of the solvent and inert solids does not involve the effect of underflow characteristics, until the 2nd and 3rd stages. It was further observed that N had a strong effect on Stage 2, where the curves were far apart compared to Stage 3. Varying N did not change the nature of variation between SSR_i with V_{si}, respectively, both of which were fitted using 2nd order polynomial functions.

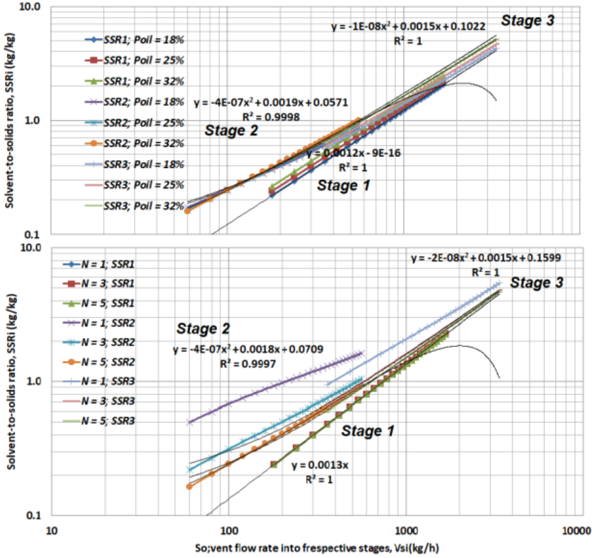


Figure 7: Effect of increasing solvent flow rate in respective stages on SSR_i at F = 1000 kg/h, and P_{oil} = 25%).

In both cases of varying P_{oil} and N, highest value of SSR were observed in Stage 3, indicating that more solvent was fed into Stage 3 compared with the previous stages, since B_o was constant. This was observed also based on results presented in Figure 4, where Y_3 were the lowest, due to both large quantity of solvent and also due to most of the oil having been extracted in Stages 1 and 2. Assuming that there is no trace of solvent in the raw meal and that B_o is constant throughout the leaching stages, then (34) shows the number of factors affecting the SSR values in each stage. The three equations show a strong correlation between the number of parameters affecting the SSR_i and the order of polynomial functions used to fit the respective data.

$$\begin{aligned} SSR_1 &= f(Vs_1) \\ SSR_2 &= f(Vs_2, W_1(1-X_1)) \\ SSR_3 &= f(Vs_3, W_2(1-X_2)) \end{aligned} \tag{34}$$

The SOR signifies whether we have enough solvent to dissolve the oil and carry it out of the system into the overflow. Optimal values of SOR exist for maximum oil recovery as shown later. Figure 8 shows the variation of SOR_i with solvent feed flow rate into respective leaching stages, V_{si}, at constant P_{oil} and also at constant N. Increasing V_{si} increases the solvent-to-oil ratio, values of which are lower in the 1st stage than in the 2nd stage. The values of SOR_i varied depending on thethe solvent flow rate across respective stages. In Stage 1, V_{s1} ranged from 200 to 2000 kg/h, and from 50 to 600 kg/h in Stage 2. The largest flow was into the 3rd stage, which ranged between 300 and 3500 kg/h. The difference in the solvent flow rate range across the three stages was set to maximize the total oil recovered using the distribution factors. The effect of P_{oil} was negligible and solvent-to-oil ratio strongly depended on stage number.

Therefore, SoR_1 increases with Vs_i according to polynomial functions of 1st, 2nd and 3rd order for Stages 1, 2, and 3, respectively, with least influence of P_{oil}. The linear relationship in Stage 1 indicates a simple mixing problem that determines SOR_1, while in the 2nd and 3rd stages, other factors come into play, making the relationship more complex. These additional factors include N, V_1 and Y_1 for Stage 2, and N, V_2, and Y_2 for the Stage 3. Detailed analysis of the variables affecting SOR_i is presented in (35):

$$\begin{aligned} SOR_1 &= f(Vs_1) \\ SOR_2 &= f(Vs_2, X_1) \end{aligned} \tag{35}$$

$$SOR_3 = f(Vs_3, X_2)$$

Equation (36) shows that the number of key variables is equal to the order of polynomial fits. Higher values of *N*, however, led to higher SOR_2 and SOR_3. The effect of *N* was more pronounced in the 3rd stage, where the curves of constant *N* were far apart compared to those in the 2nd stage. The nature of variation of SOR_i with V_{s2} and V_{s3} depends strongly on stage numbers. Increasing N does not affect SOR_1 and comes into play during separation after mixing of the two streams in accomplished. Thus, *N* will affect Stages 2 and 3, since the streams mixed in those two stages results from the separation according to selected values of *N* in Stages 1 and 2.

Effect of *N* on Total Oil Recovery

The settings for solids to solution ratio in the underflow are critical towards the total oil recovery. The effect of increasing N or effectiveness of decantation process on To was studied at a fixed plant size and solvent flow rates into stages. Figure 9 presents the variation of total oil recovered from a three-stage crosscurrent leaching plant with *N* for fixed equal solvent feed flow rates into respective stages. The total oil recovered increased with *N*, reaching a limit, for $N \geq 5$, after which no further increase in T_o was observed. The data was fitted well with polynomial functions of 4th order for V_s = 200 kg/h and 5th order for $V_s \geq$ 300 kg/h. Increasing *N*, decants most of the solution into the overflow streams, thus increasing T_o, and vice versa. Similarly, increasing V_s increases the total oil recovered similar to results presented in Figure 9.

Higher values of *N*, on the other hand, signify expensive decanting equipment, which may not justify the extra extraction efficiency. For example, there is little change in total oil recovered at V_s = 500 kg/h when $N \geq 3$.

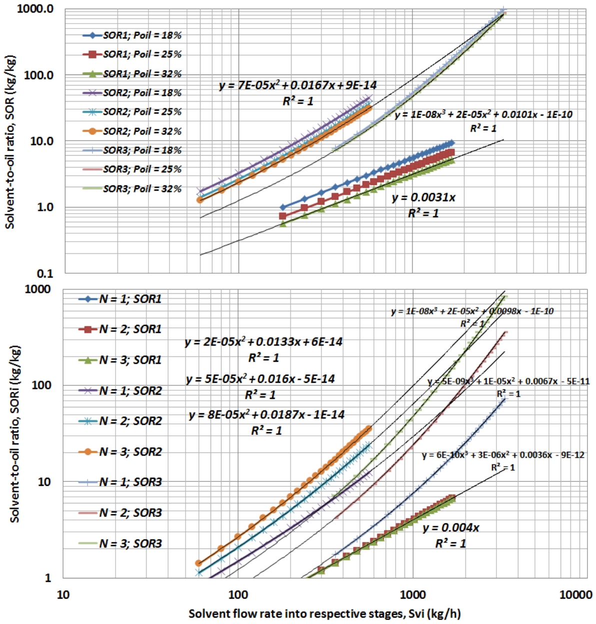


Figure 8: Variation of solvent-to-oil ratio (*SOR$_i$*) with equal-solvent flow rate into each stage (a) at various oil contents in the raw meal, *P$_{oil}$*, and (b) solids-to-solution ratio in the underflow, *N*.

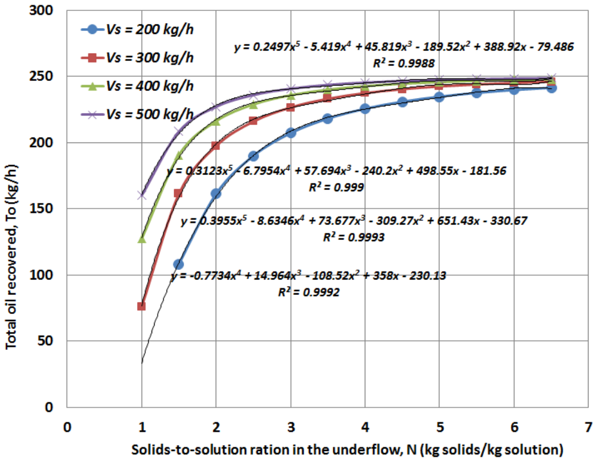


Figure 9: Variation of total oil recovered with solids-to-solution ratio, *N* for equal constant fresh solvent flow rate in each stage V$_s$ (*F* = 1000 kg/h and *P$_{oil}$* = 25%).

Figure 10 presents the variation of the total oil recovered with *N* for constant values of raw meal feed flow rates, *F*. Similar to Figure 9, the total oil recovered increased with *N*, reaching a limit, after which no further increase in T_o was observed. The limit T_o values were observed to be distinctly different depending on F. The lower feed flow rate (signifying smaller oil extraction plants), the effect of N is negligible since T_o levels off immediately, that is, for $N > 1$. Thus, for smaller plants, $N \leq 2$ is acceptable for oil extraction. The data was fitted well with polynomial functions of 4th order for all values of raw meal feed flow rates, with R^2 values increasing with F. For small leaching plants ($F \approx 500$ kg/h), N has no significant effect on T_o. Compared to large plants ($F \approx 2000$ kg/h) where N has a strong effect, a higher value of $N \geq 6$ is required for extracting the oil from raw meal, necessitating expensive decanting equipment. Considering Figures 9 and 10, asymptotic curves at higher N values was observed. Considering the cost of solvent and decantation or pressing equipment for higher N, avoiding unnecessary high values of V_{si} and N is recommended.

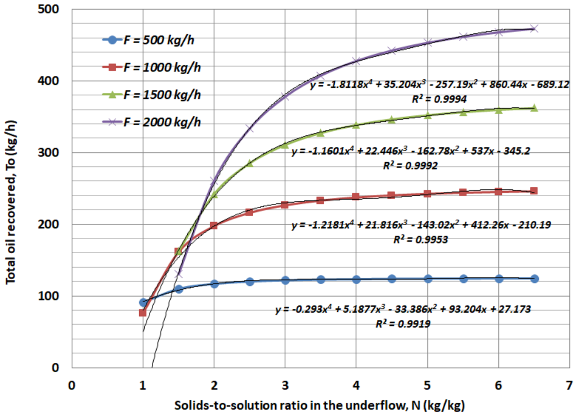


Figure 10: Variation of total oil recovered with *N* for constant values of raw meal feed flow rates or leaching plant size.

Effect of Solvent Flow Rate into Respective Stages on Total Oil Recovered at constant *N*

In multistage crosscurrent leaching, separate solvent flow rates are applied to each stage. Given the prevailing oil content, solids and solution in the underflow, the total oil recovered from each stage will vary differently with solvent flow rate. Figure 11 shows the effect of increasing solvent flow rate across respective stages under fixed values of N. Increasing V_s increases the total oil recovered as shown also in Figure 9. Initially T_o increases with V_s for Stage 1, reaching asymptotic values of the total oil recovered at higher values of V_{si}. For $N \geq 3$, the clear effect of V_s is negligible. In general, T_o increases with solvent flow rate across each stage in a manner depending on the set value of N. This shows that at lower values of N the percent oil recovery is low. In Stage 1, T_o varied with V_s according to 3rd order polynomial functions. In Stage 2, the polynomial functions relating T_o with V_s were more complex, changing from 2nd to 5th order. In Stage 3, T_o increased linearly with V_{S3} for all set values of N. The solvent flow rate across a given stage is a key factor towards improved plant performance in terms of total oil recovered. Increasing solvent flow rate across a given stage should lead to high oil recovered but also high process cost due to solvent requirements. The results in Figure 11 indicate distinct behaviour for the three stages, as V_{si} was increased. The results show that the stages are independent or uncoupled systems behaving differently with N and V_{si}. In all cases, $N = 1$ leads to lowest total oil recovery compared to $N > 1$. For $N > 3$, no appreciable effect of N on the relationship between T_o and V_s was observed, whereby, the curves of the same shape coincide.

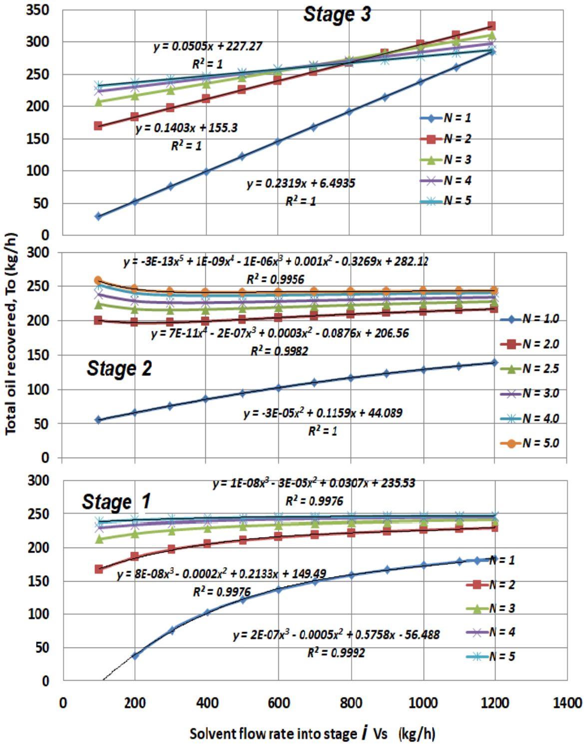


Figure 11: Variation of total oil recovered with solvent flow rate in respective stages for fixed values of *N*.

In Stage 3, linear relationships were observed for T_o vs V_{si} for $N > 1$, and also, the slope of the curves decreased with N. The results show that for $N > 1$ a common value of T_o exist when $V_s = 800$ kg/h. When the feed into Stage 3 was changed to $V_s = 1200$, T_o was the same for $N = 1$ and $N = 5$, showing that further increase in V_s, at $N = 1$, can exceed the corresponding values of the total oil recovered at $N \geq 2$. The value of V_s at the point of intersection for $N = 1$ and 2 was determined to be $V_{s3} = 1624.5$ kg/h, at which, $T_o = 383.22$ kg/h total oil recovered. Depending on the availability of solvent to feed into Stage 3, a value of N exists which leads to maximum total oil recovered. For $V_{s3} \leq 800$ kg/h, $N = 5$ gives the maximum total oil recovered. For $800 \leq V_{s3} \leq 1624.5$ kg/h, $N = 2$ gives the maximum total oil recovered, and finally, for $V_{s3} > 1624.5$ kg/h, $N = 1$ gives the maximum total oil recovered, as summarized in Table 2.

Based on Table 2, Figure 12 shows the optimal values of N corresponding to the range of V_{s3} for maximum total oil recovery. The quantities are referred to as optimal based on the fact that maximum amount of oil is obtained which can maximize sells and profit. The plot shows the linear equations between T_o and V_{s3} for each of the three ranges of V_{s3} corresponding to $N = 5$, 2 and 1, respectively. Thus, given a solvent flow rate into Stage 3, the expected total oil to be recovered can be determined from Figure 12.

Table 2: Solvent flow rate range in Stage 3 for maximum total oil recovery

N	V_{s3} Range (kg/h)	Equation for T_o	Remarks
5	$V_{s3} \leq 800$	$T_o = 0.0505V_{s3} + 227.27$	Maximize overflow by pressing hard the cake, leaving very little solution in the underflow leaving 0.2 kg solution per kg solids.
2	$800 \leq V_{s3} \leq 1624.5$	$T_o = 0.1403V_{s3} + 155.30$	Allow 2 kg solids per kg solution, which is lower pressing requirement for the cake and slightly high solution is left in the underflow.
1	$V_{s3} > 1624.5$	$T_o = 0.2319V_{s3} + 6.4935$	Least pressing requirements; leaving 1 kg solids per kg solution (equal amount of solution and solids. This may need extra care of spent cake to recover more solvent and minimize losses.

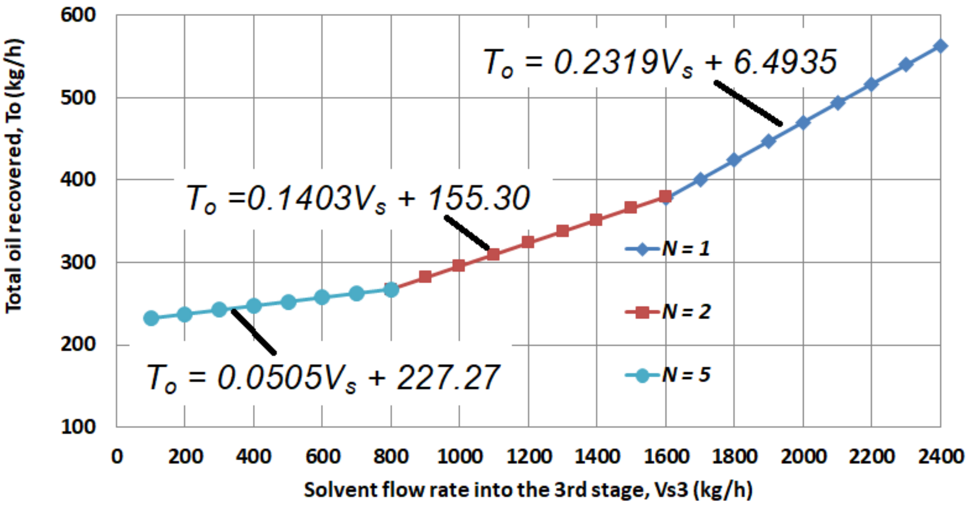


Figure 12: Determination of optimum values of *N* for different ranges of solvent flow rate into Stage 3 for maximum total oil recovery.

Identification of the optimal range of solvent distribution factors

The utilization of (8) and Figure 3 involved testing a range of values of α_i leading to maximum total oil recovery. To determine the effect of quantity of solvent fed into a given stage on the total oil recovered, the distribution factors were applied on Q_s as shown in Figure 3. Three factors α_1, α_2 and α_3 as per Equations 7 and 8 were applied to Q_s leading to V_{s1}, V_{s2} and V_{s3} data, respectively. In Figure 13, four different sets of α_i values were tested while searching for maximum values of T_o.

Based on (8), changing α_i implies feeding a specific solvent flow rate to the given stage. Highest values of T_o resulted from setting $\alpha_1 = 0.3$, $\alpha_2 = 0.1$ and $\alpha_3 = 0.6$. The range of values around these set values was tested. Thus, the range of α values for maximum total oil recovered was estimated to be α_1 = 0.05 to 0.3, α_2 = 0.05 to 0.2, and α_3 = 0.5 to 0.95. Figure 13 shows further that the maximum values of T_o, corresponding to optimal values of α_1 has a local maximum, indicating that further turning of α values can lead to highly optimal values of T_o.

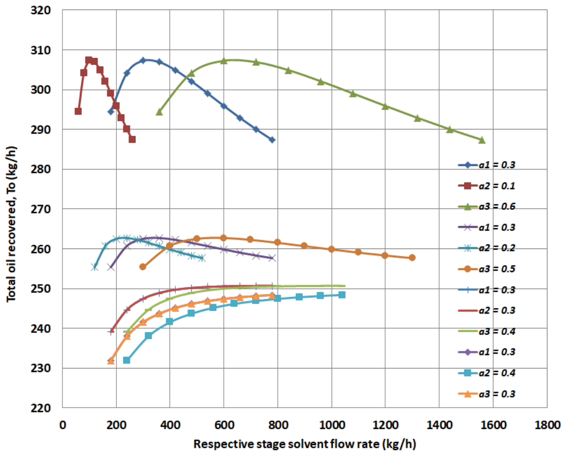


Figure 13: Variation of total oil recovered with respective stage solvent flow rate, V_{si} (kg/h) based on flow distribution factor, α_i.

For example, the maximum values of T_o were reached at V_{s1} = 300 kg/h, V_{s2} = 100 kg/h and V_{S3} = 600 kg/h, that is, when Q_s = 1000 kg/h was distributed using $\alpha_1 = 0.3$, $\alpha_2 = 0.1$ and $\alpha_3 = 0.6$. The ranges of V_{s1} and V_{s2} were narrow (10 to 1,000 kg/h only) compared to V_{s3} (500 to 3,500 kg/h) for the same range of total oil recovered.

Percent Contribution of each Stage to the Total Oil Recovery based on solvent feed flow rate

During multi-stage crosscurrent leaching, the overflow streams from the three stages are normally combined to form the final product from which oil recovered is further separated. The values of total oil recovered and the overall concentration of the oil in the combined overflow were determined based on (15) and (16). Based on the low solute concentration in the overflow from Stage 3 and the higher solvent flow rate into Stage 3 aimed at attaining higher total oil recovery, it is worth assessing the contribution of the stages to the total oil recovered. The low concentration of oil in the overflow from the 3rd stage (as shown in Figure 4) is attributable to large solvent flow rate or the higher value of α_3 compared to α_1 and α_2.

Figure 14 compares the percent contribution of the stage to the total oil at fixed values of N. For each value of N (3, 4 and 5), three curves showing the values of percent contribution are plotted against the solvent flow rate into each stage, V_{si}. The highest contribution was from the first stage, attributable to solvent coming in contact with raw meal containing the highest quantity of oil, which is separated via V_{si} for a selected value of N. The highest percent contribution to the total oil recovered was observed to originate from Stage 1, where the solvent comes in contact with the raw meal. The contribution to total oil recovery by Stage 1 increased with solvent flow rate into the stage, V_{s1}, according to quadratic functions, as shown in Figure 14. The contribution by Stage 1 was highest for range of α_1 and V_{s1} studied (that is, 250 to 1200 kg solvent/h) which ranged between 48 and 80%, could have different results for the solvent flow rate into Stage 1 below 200 kg/h, because the contribution from Stage 3 could also play a large role in that solvent flow rate range.

The next high contribution was observed to come from Stage 3, being highest at $N = 5$. Together with the low concentration of oil in Stage 3, higher contribution than Stage 2 was observed due to higher quantity of oil (= Y_3V_3) despite the lower value of Y_3. The contribution from Stage 3 ranged from 10 to 40% when V_{s3} was varied between 500 and 2,000 kg solvent/h. The percent contribution in Stage 3 decreased with solvent flow rate for all values of N, also, according to 2nd order polynomial functions. Therefore, it can be concluded that the 3rd stage is critical and is needed for maximum total oil recovery.

In the 2nd stage, the least contribution to total oil recovered was observed for all values of N. The percent contribution by Stage 2 ranged from 10 to 15% only, with a clear decreasing trend for $V_{s2} > 400$ kg solvent/h. The percent contribution to the total oil recovered increased slightly reaching a maximum value before decreasing again. Higher contribution in Stage 2, was observed for $N = 5$, similar to the observation made in Stages 1 and 3. The variation of percent contribution to the total oil recovered in the 2nd stage was also observed to follow 2nd order polynomial function.

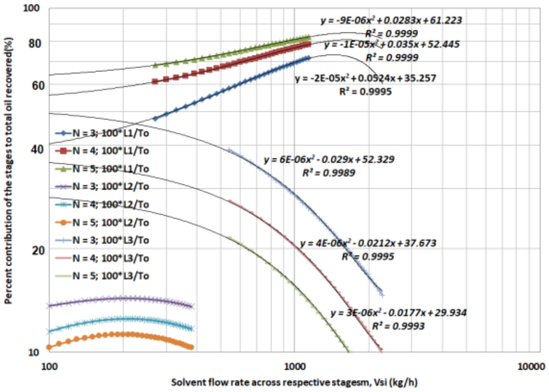


Figure 14: Variation of the respective stages contribution to the total oil recovered with the solvent flow rate into the respective stages at fixed values of N.

Effect of SSR and SOR on Total Oil Recovery

Among the factors affecting oil recovery during crosscurrent leaching is the solvent-to-solids ratio across each stage, SSR_i. When solids are flowing into each stage and coming in contact with solvent, there is a need to establish the optimum ratio of solvent-to-solids which leads to maximum oil recovery. To simulate the effect of increasing SSR on the total oil recovery, the feed flow rate was fixed at 1,000 kg/h with P_{oil} = 18%. Figure 15 shows the variation of total oil recovered with the SSR for Stages 1, 2 and 3, at F = 1000 kg/h and N = 5. The total oil recovered increased with SSR in each stage, reaching a maximum before decreasing again. For each stage, the oil recovered had a peak value. It was noted also that, the maximum total oil recovered occurs at lower SSR in Stage 2, followed by Stage1 and at highest SSR in Stage 3. With constant inert solids flow across the system, this implies also that the peak total oil recovered occurs at higher solvent flow rate into Stage 3, almost twice the flow rate of solids.

For Stages 1 and 2, the oil recovery was highest at lower solvent flow rates, about 1/5 to ¼ of the inert solids flow rate. Therefore, results presented in Figure 15 helps to determine the optimum solvent flow rate across each stage leading to maximum total oil recovery. Former researchers did not observe the peak percent copper recovery when they varied SSR (Osman et. al., 2013), possibly due to narrow SSR range from 1.5 to 3.0 only, while this study assessed a wider range from 0.1 to 8. Optimization of jojoba oil extraction yield has been studied by varying the seed-to-solvent ratio (F/V_s), from 5 to 15 and observed that the optimum ration was at 12.9 kg seed/kg solvent, equivalent to F/V_s = 0.08 kg solvent per kg raw meal fed [18]. It should be noted that F/V_s values are slightly lower than SSR values reported in this paper due to exclusion of the quantity of oil in the denominator. The recovery is on the other hand, aided by higher temperature (65°C), particle size (0.48 mm) and high mixing speed of 1000 rpm (Osman et. al., 2013).

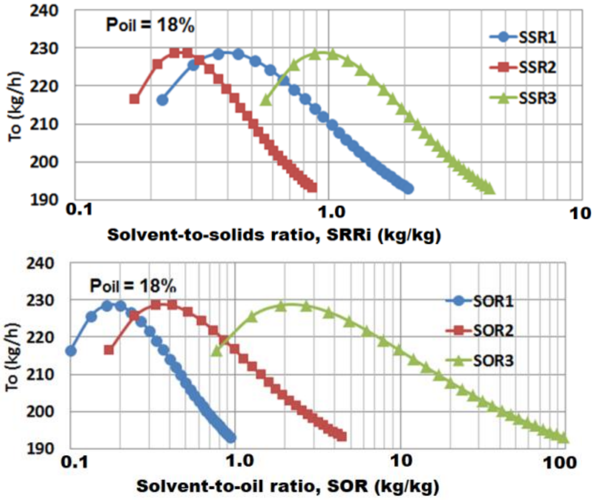


Figure 15: Variation of total oil recovered with SSR and SOR_i in respective stages (N = 5 and F = 1,000 kg/h).

Knowing the SSR_i values, will allows the operators to set the necessary solvent flow rates to achieve the maximum total oil recovery. The shift in the peak value of T_o along the horizontal axis signifies that the leaching stages should be operated differently in terms of solvent flow rates, to achieve the given maximum total oil recovery. Thus, for a fixed raw meal flow rate (F) and percent oil in the raw meal (signifying different oilseeds), the solvent flow rate giving the ratios, SSR_i corresponding to maximum total oil recovery should be selected. At P_{oil} of 18%, the maximum T_o was reached when SSR_2 = 0.26, SSR_1 = 0.3 and SSR_3 = 1.0 being highest in Stage 3. Between Stage 1 and 2, results show that $SSR_1 > SSR_2$, attributable to the fact that in the first stage large amount of solvent is required to wet all the fresh raw meal and allow the solute to dissolve completely into the solvent. In the Stage 2, on the other hand, the solvent adhering to the solids after Stage 1 based on the set value of N, is available for extraction, and little amount of solvent is required since the solute in the solids is already dissolved.

Similarly, the data for SOR was varied by selectively varying the solvent flow rate across each stage, while monitoring the total oil recovered ((15) and (23)). The results presented in Figure 16 show the variation of total oil recovered (kg/h) with SOR. The SOR_i data in Figure 15 ranged from 0.1 to 1,000 kg solvent/kg oil. The same amounts of total oil recovered were obtained at lowest SOR_i for Stage 1, followed by Stage 2 and lastly, at higher SOR values in Stage 3. The observed T_o values at low SOR in Stage 1 can be attributed to largest quantity of oil available in Stage 1. The peak T_o was observed at SOR = 0.2 and 0.4 kg solvent/kg oil in Stages 1 and 2, respectively. In Stage 3, on the other hand, the SOR value for which the maximum total oil recovered was reached was 2.0 kg solvent/kg oil. Therefore, a large quantity of solvent is required in the last stage in order to recover maximum total amount of oil in the raw meal. At SOR_3 = 2.0 kg solvent/kg oil, the loss of solvent is higher through the underflow, controlled by setting higher $N = 5$.

Therefore, because Stage 3 operates at a very low oil concentration as observed in the solution leaving as overflow or underflow (Figures 4 and 15), its necessity require justification. This was concluded based on data presented in Figure 15, whereby, the contribution of Stage 3 to the total oil recovered was higher than that of Stage 2. Higher SOR_3 values, therefore, does not signify least amount of oil being extracted, but it is due to largest amount of solvent fed into Stage 3.

Effect of Wash Ratio on Total Oil Recovered

The wash ratio relates the amounts of solution in the overflow with that in the underflow streams. The WR is also critical as it parts the solution into two portions (overflow and underflow). At fixed values of N, the amount of oil carried out by the overflow depends on WR. The variation of total oil recovered with WR_1, is presented in Figure 16 for Stages 1, 2 and 3. In Figure 16 there are marked differences between the variations of total oil recovered with WR_i across the three stages. In Stage 1, the total oil recovered increased faster with WR_1 at $N = 1$, and slowly with WR_1 at higher values of $N = 7$.

For higher values of N, the total oil recovered changed slightly with WR_2. It was observed that T_o is slightly higher at low WR_2 and drops slightly with increasing WR_2.It was remaining constants at higher values of $WR_2 \geq 2$.

Linear relationships were observed in Stage 3 between T_o and WR_3 for all values of N. Moreover, a constant value of T_o = 240 kg/h was observed for all values of N when WR_3 = 1.8. The total oil recovery increases faster with WR_3 at $N = 1$, slopes of which dropped as N was increased further. While for Stages 1 and 2, the set values of N have distinct ranges of the total oil recovered, which increases with N from 1.0 to 7.0, in Stage 3, there is no distinct range of T_o. For instance, at $N = 1.0$, increasing WR_3 changed T_o from 120 to 275 kg/h, in a very short range of WR_3 (0 to 2 only), while at $N = 5$, T_o was changed from 240 to 275 kg/h when WR_3 changed widely from 0.2 to 8.0. The results presented in Figure 19, resemble those presented in Figure 11, where T_o varied with V_{si} at constant values of N. The linearity of curves in Figures 11, 12 and 17 is solely a characteristic of Stage 3, not observed in other stages.

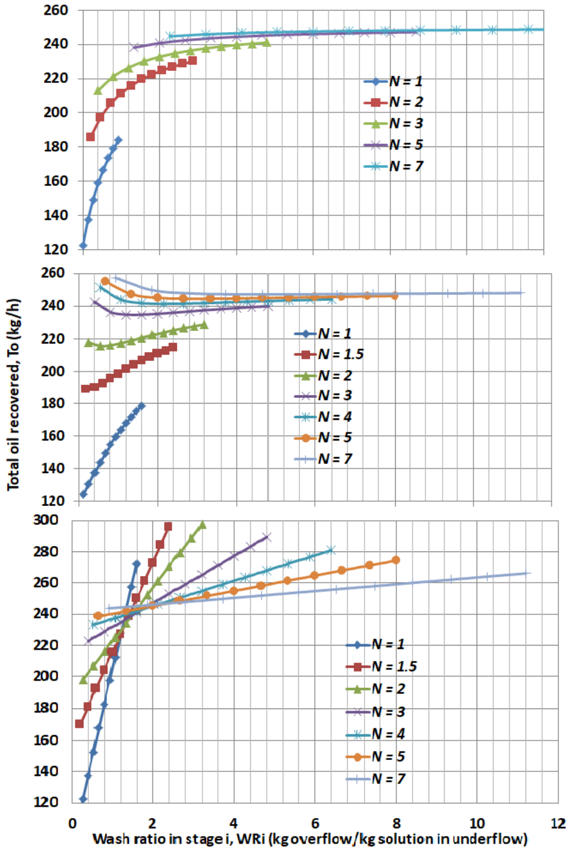


Figure 16: Variation of total oil recovered with wash ratio, WR_i, in respective stages.

CONCLUSION AND RECOMMENDATION

Based on the key assumptions, the crosscurrent leaching operation was modelled using a large number of equations as summarized in the model flowchart. This signifies that the crosscurrent leaching operation with three stages is a complex system.

The oil concentration in the overflow solution was highest for Stage 1 and lowest for Stage 3. While the oil concentration was higher in the overflow from Stage 2 than that from Stage 3, large solvent feed flow rate into Stage 3, led to higher oil recovery and contribution to total oil recovery in Stage 3 (10 – 40%) than Stage 2 (10 – 15%). The percent contribution was highest in Stage 1 (50 – 80%).

The SSR_i decreased logarithmically with raw meal feed flow rate, F being highest at highest P_{oil} of 40% (indicating low solids content). The SOR_i on the other hand decreased also logarithmically with F, with highest values observed at lowest values of P_{oil}, due to smaller oil content fed to the plant. At lower levels of P_{oil}, the relationship between SOR_i and F was no longer linear. The SSR_i increased with V_{si} at constant values of N and P_{oil}, while SSR_i increased linearly with V_{si} in Stage 1, the variations were fitted well with second order polynomial functions for Stages 2 and 3, values of which were highest in the last stage. The 2nd order was attributed to the two key parameters affecting the SSR_2 and SSR_3, that is, V_{s2} and $W_1(1-X_1)$ for SSR_2 while SSR_3 was observed to depend on V_{s3} and $W_2(1-X_2)$. Similarly, while SOR_1, varied linearly with V_{si} (attributable to only one parameter, V_{s1}), SOR_2 and SOR_3 varied with V_{si} according to 2nd order polynomial functions, attributable to two key parameters (that is, Vs_2 and X_1 for SOR_2) while SOR_3 was affected by V_{s3} and X_2.

The total oil recovery increased with N at contract F, according to fourth order polynomial functions, which shows saturation at higher values of $N > 3$. Also, at constant values of total solvent feed flow rate, V_s, the total oil recovered increased faster with N towards a maximum according to polynomial functions of fifth order for $N > 3$. The total oil recovery varied differently with V_{si} depending on stage number and N. While all of the curves were fitted with 3rd degree polynomial functions in Stage 1, the order increased from 2nd to 5th as N was increased from 1 to 5 in Stage 2. In Stage 3, on the other hand, the total oil recovered varied linearly with V_s slopes of which decreased with N. Depending on the range of solvent flow rate, it was possible to select a value of N leading to a maximum total oil recovery. The effect of solvent flow rate into a given stage of a multistage crosscurrent plant on total oil recovered is complex showing that the operation of the plant requires extensive knowledge of the operating parameters.

There exists optimal range of solvent flow rate into a given stage, which leads to the maximum total oil recovery. This necessitates proper distribution of solvent into the three stages, using distribution factors. It can be concluded that the optimal range was $\alpha_1 = 0.05$ to 0.3, $\alpha_2 = 0.05$ to 0.2 and $\alpha_3 = 0.5$ to 0.95. Given the optimal range of distribution factors, the contribution of the stages to the total oil recovered also ranged from 50 to 80% for Stage 1, 10 to 15% only for Stage 2, and 10 to 40% for Stage 3.

Both SSR_i and SOR_i show the optimal values at which the total oil recovery reached the maximum values depending on the oil content in the raw meal, P_{oil}. The total oil recovery varied widely with WR_i depending on N, and stage number. It was concluded that detailed analysis of the key operating parameters need to be carried out before oil extraction process is initiated, for maximum oil recovery from the feed raw meal. The study has contributed strongly to the pool of knowledge on oil extraction by crosscurrent mode of operation.

REFERENCES

A. Sharma, S.K. Khare and M.N. Gupta. (2002). "Enzyme Assisted Aqueous Extraction of Peanut Oil". *Journal of the American Oil Chemists' Society*, **79**:215-218. https://doi.org/10.1007/s11746-002-0463-0

G. Baskar, G. Kalavathy, R. Aiswarya and A.I. Selvakumari. (2019). "Advances in bio-oil extraction from nonedible oil seeds and algal biomass", in A. Kalam, (Ed.), Woodhead Publishing Series in Energy, Advances in Eco-Fuels for a Sustainable Environment, Woodhead Publishing, pp. 187-210.

S.V. Manyele and I. Kahemela. (2008). "Effect of Particle Size on the Performance of Solvent Extraction of Edible Vegetable Oil from Oilseeds". *The Tanzania Journal of Engineering and Technology*, **2**(2):1-13.

S.V. Manyele. (2008). "Comparison of Solvent Extraction Performance Factors between Sunflower and Soybean Oilseeds using Petroleum Ether". *The Tanzania Journal of Engineering and Technology*, **2**(3):34-48.

Q. Hao, J. Tian, X. Li andL. Chen. (2017). "Using a hybrid of green chemistry and industrial ecology to make chemical production greener". *Resources, Conservation and Recycling*, **122**: 106-113. https://doi.org/10.1016/j.resconrec.2017.02.001

S.M. Sadeghi, G. Vanpeteghem, I.F.F. Neto and H.M.V.M. Soares. (2017). "Selective leaching of Zn from spent alkaline batteries using environmentally friendly approaches", *Waste Management*, **60**: 696-705. https://doi.org/10.1016/j.wasman.2016.12.002

S.-H. Jeon, K. Yoo and R.D. Alorro. (2017). "Separation of Sn, Bi, Cu from Pb-free solder paste by ammonia leaching followed by hydrochloric acid leaching." *Hydrometallurgy*, **169**: 26-30.

Al-Hamamre and J. Yamin. (2014). "Parametric study of the alkali catalyzed transesterification of waste frying oil for biodiesel production". *Energy Conversion Management*, **79**:246–254.

R.F. Khasawneh. (2017). "Leaching of Jojoba oil". *Journal of Natural Science Research*, **7**(10): 49–62.

E.A.E. Sheet. (2018). "Effect of preheating waste cooking oil on biodiesel production and properties". *Energy Source*, Part A, **40**(2): 207–213. DOI: 10.1080/15567036.2017.1410594.

M.R. Lubls. (2013). "Leaching of Oil from Tuna fish liver by using solvent of Methyl Ethyl Ketone". *Jurnal Rekayasa Kimia dan Lingkungan*, **9**:188 -196.

M.K. Abu-Arabi, M.A. Allawzi, H.S Al-Zoubi, A. Tamimi. (2000). "Extraction of Jojoba oil by pressing and leaching". *Chemical Engineering Journal*, **76**(1): 61-65.

F. Takadas and O. Doker. (2017). "Extraction method and solvent effect on safflower seed oil production". *Chemical and Process Engineering Research*, **51**:9-17.

M. Pinelo, M. Rubilar, M. Jerez, J. Sineiro and M.J. Núñez. (2005). "Effect of Solvent, Temperature, and Solvent-to-Solid Ratio on the Total Phenolic Content and Antiradical Activity of Extracts from Different Components of Grape Pomace". *Journal of Agricultural and Food Chemistry*, **53**(6):2111-2117. DOI: 10.1021/jf0488110.

A. Laitinen and J. Kaunisto. (1999). "Supercritical fluid extraction of 1-butanol from aqueous solutions". *Journal of Supercritical Fluids*, **15**:245-252. DOI: 10.1016/S0896-8446(99)00011-X.

A. Osman, D.Nezungai , F.K. Crundwell and J. Lloyd. (2013). "Analysis of the effects of changes in operating conditions on the agitated leaching of copper". *The Southern African Institute of Mining and Metallurgy, Base Metals Conference*, pp. 235-254. https://www.saimm.co.za/Conferences/BM2013/235-Osman.pdf.

G. Nimet, E. Antonio da Silvaa, F. Palúa, C. Darivac, L. dos Santos Freitas, A.M. Neto, L.C. Filho. (2012). "Extraction of sunflower (Heliantus annuus L.) oil with supercritical CO_2 and subcritical propane: Experimental and modeling". *Chemical Engineering Journal*, **168**: 262-268. https://doi.org/10.1016/j.cej.2010.12.088

I. Al-Sheikh, J.A.A. Yamin. (2019). Modeling and optimization of jojoba oil extraction yield using Response Surface Methodology. *Journal of Pharmacy & Pharmacognosy Research*, **7**(5):367-380.