Countercurrent Liquid-Liquid Extraction in a Packed Tower

Solvent Extraction of Oil by Nitrobenzene

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A description and discussion are presented of the method by which results of countercurrent extraction in a tower are evaluated by comparison with laboratory batch countercurrent extractions. An equilibrium curve has been obtained for the oil-nitrobenzene system used. Data from these laboratory treats are presented in a convenient form in a chart, by which comparisons of extractions can be made rapidly.

THE purpose of this investigation was to find and study the variables that enter into liquid-liquid extraction in a packed tower. The liquid system studied was a lubricating oil consisting of naphthenic and paraffinic types, and the solvent used was nitrobenzene. Only scant information exists in the literature pertaining to liquid-liquid extraction in either a packed- or spray-type tower. Recently, however, articles have appeared by Elgin and Browning (3) and by Varteressian and Fenske (13) dealing with this subject. Mathematical analyses of continuous liquid-liquid extractions by countercurrent processes have been discussed by Saal and Van Dyck (12), Hunter and Nash (8, 9, 10), Evans (4), Cannon and Fenske (2), and others. Analogies between continuous liquid-liquid extraction and distillation have been thoroughly discussed by Saal and Van Dyck (12), in connection with their work on solvent extraction of hydrocarbons, and by Varteressian and Fenske (14). There is little information in the literature pertaining to advantages of the types of packing suitable for use in packed tower extractions, and only a few patents indicate preferences in kind, size, and type. In this connection the patents of Fenske (5) are to be noted. But information is available concerning packing and its effect on gas-liquid extraction. A recent paper by Baker, Chilton, and Vernon (1) indicates the type of work and general conclusions in this field of extraction. In the present work, attention was given to the packing material and its amount, size, and shape with regard to general characteristics in the liquid-liquid extraction process.

Laboratory Control Extractions

To measure the results of a countercurrent extraction, it was found most convenient to compare them with the results of laboratory equilibrium batch countercurrent treats (B. C. C. treats). Data for an equilibrium curve were obtained for the solvent-oil system by making a series of mixes Using the data and discussion as a basis, data are presented which were obtained from a number of experiments, together with a discussion and evaluation of the variables affecting extraction in a packed tower. It is believed that this information is presented in a form which can be used as a basis for design of equipment to effect separations by extraction of systems similar to the solvent-oil system used in these experiments.

of solvent and stock oil in different ratios and separating the two layers formed; such extractions are called "single-stage batch treats." Other extractions were made using two- and three-stage batch countercurrent treats. The results of such treats were not in agreement with predictions made according to the suggestions of Saal and Van Dyck and of Hunter and Nash. These methods are based upon a system of three pure components and are not rigidly applicable to the complex solvent-oil system used in this work. For this reason a comprehensive series of treats was made in the laboratory under equilibrium conditions for use as standards of comparison. Figure 1 shows the scheme whereby countercurrent extractions were made by a batch process in the laboratory. Each batch was allowed to reach equilibrium before separation. and when successive values for R and E were constant in weight, the process was considered to have reached equilibrium. Equilibrium in each batch or stage was ensured by mixing thoroughly while heating to miscibility and then cooling to the desired temperature of extraction, whereupon the two immiscible phases separated. In nearly all such treats equilibrium was reached after four extraction cycles had been completed.

Analyses of Solutions

Solutions of solvent and oil were separated by distilling off the solvent under high vacuum. Weight determinations were made and used to make material balances as a check on the accuracy of the separations. The oil used and the oils obtained from the extract and raffinate solutions were evaluated by the viscosity-gravity constant (V. G. C.). The V. G. C. was proposed by Hill and Coats (6) as a measure of naphthenicity and paraffinicity. V. G. C. was selected for this work because it is easy to determine, is easily reproducible, is an additive property of an oil, and is a recognized method for measuring the degree of separation of an oil into naphthenic and paraffinic constituents (7). High values of V. G. C. indicate relatively high naphthenicity and low values indicate relatively high paraffinicity. The extraction system was oil with nitrobenzene as the solvent, but a three-component system was assumed to be present; the components were naphthenic oil, paraffinic oil, and nitrobenzene.



Equilibrium Curve

Equilibrium conditions between the two immiscible phases for this oil-solvent system were determined by laboratory B. C. C. treats, using one, two, and three stages. Table I gives data from these treats which were used to construct the equilibrium curve (Figure 2) and are presented as volume data. Volume data are permissible for use in this system since the volumes are considered to be additive. The data are plotted on rectangular coördinates in Figure 2, as previously proposed by Kurtz (11). The significance of the coordinates and the method of plotting in this way rather than on triangular coördinates follow. The abscissa is divided into any number of parts desired, to represent various ratios of two components (in Figure 2, V. G. C. represents mixtures of naphthenic, N, and paraffinic, P, oils). The ordinate represents percentage of solvent in any solution or mix. A solvent apex (in this work, ΦNO_2 apex) is chosen at any convenient point on the 100 per cent solvent line. To represent a solution consisting of $\overline{83}$ per cent N and P whose V. G. C. is 0.834, and 17 per cent of ΦNO_2 , the following procedure is used.

TAE	BLE I.	Exper Ba	IMENI ATCH C	CAL ANI) Smoo RCURRI	thed Ent Ex	DATA FROM TRACTIONS	MEQ1	JILIBRIT	M
Stages	Solvent Ratio	-Raffin V.G.C.	— Labo nate Vol. % ΦΝΟ2	v. G. C.	Data act Vol. % ΦNO2	E/R	V. G. C.	moothe nate Vol. % yield	ed Data- Extract V.G.C.	E/R
1	$0.5 \\ 1.0 \\ 1.5 \\ 2.0 \\ 2.5 \\ 3.0$	$0.844 \\ 0.834 \\ 0.827 \\ 0.821 \\ 0.817$	$19.8 \\ 17.1 \\ 15.5 \\ 13.2 \\ 12.8$	$0.897 \\ 0.893 \\ 0.889 \\ 0.883 \\ 0.883 \\ 0.875$	71.475.879.081.183.5	$\begin{array}{c} 0.36 \\ 1.38 \\ 2.40 \\ 3.79 \\ 7.04 \end{array}$	$\begin{array}{c} 0.844 \\ 0.834 \\ 0.827 \\ 0.822 \\ 0.818 \\ 0.815 \end{array}$	88.7 72.8 62.9 54.8 48.4 41.7	$\begin{array}{c} 0.897 \\ 0.893 \\ 0.889 \\ 0.884 \\ 0.880 \\ 0.875 \end{array}$	$\begin{array}{c} 0.35 \\ 1.25 \\ 2.36 \\ 3.61 \\ 5.41 \\ 7.42 \end{array}$
2	$ \begin{array}{c} 1 & 0 \\ 1 & 5 \\ 2 & 0 \\ 2 & 5 \\ 3 & 0 \end{array} $	0.822 0.815 0.812	13.7 13.7 12.7	0.889 0.886 0.881	70.9 79.3 81.2	1.57 4.11 8.43	0.826 0.818 0.814 0.810 0.808	$\begin{array}{r} 66.2 \\ 56.7 \\ 50.7 \\ 44.5 \\ 40.0 \end{array}$	0.897 0.892 0.887 0.882 0.878	1.592.754.226.008.45
3	$1.0 \\ 1.5 \\ 2.0 \\ 2.5 \\ 3.0$	0.821 0.810 0.800	13.6 11.5 10.5	0.899 0.890 0.880	70.6 77.7 83.0	1.83 4.33 8.83	$\begin{array}{c} 0.821 \\ 0.814 \\ 0.809 \\ 0.804 \\ 0.800 \end{array}$	$62.8 \\ 54.4 \\ 48.7 \\ 42.5 \\ 37.5 \\ \end{array}$	0.899 0.893 0.889 0.884 0.884 0.880	1.823.034.456.458.90

Place a straight edge on the base line at T and on the ΦNO_2 apex, draw a dotted line as shown, and mark the point R on this line where it crosses the ordinate of 17 per cent; R then represents the solution. The solvent ratio is the ratio of solvent to oil and is shown on the ordinate along with percentage of solvent. The relation between solvent ratio and percentage solvent is direct; for example, a solvent ratio of 2.0 means that a mix of 2 parts of solvent and 1 part of oil consists of 66.67 per cent solvent and 33.33 per cent oil.

The solid parts of the equilibrium curve in Figure 2 were constructed from experimental data and are shown with the tie lines for single-stage extractions. The quantitative relations involved in the equilibrium diagram here used may be illustrated as follows: Let a single-stage treat be made of an oil stock of 0.850 V. G. C. with nitrobenzene, in a solvent ratio of 1.0. S is marked on the base line corresponding to a V. G. C. of 0.850. Then the ratio of naphthenic oil of 0.900 V. G. C. to paraffinic oil of 0.800 V. G. C. in S is equal to the ratio of the distances PS and SN, or

$$\left(\frac{N}{P}\right)_{S} = \frac{PS}{SN} \tag{1}$$

On a line connecting S and the ΦNO_2 apex, the point M is marked, where the amount of solvent is 50 per cent of the mix and the solvent ratio is 1.0. The tie line passing through M



Figure 2. Equilibrium Curve for $0.85~V.\,G.\,C.$ Oil and Nitrobenzene at 50° F., with One-Stage Tie Lines

intersects the extract section of the equilibrium curve at E and the raffinate section of the curve at R. These two phases are the extract and raffinate solutions resulting from the treat, and are saturated with oil and nitrobenzene, respectively; also they are at equilibrium and are immiscible with one another. The dotted line drawn through the ΦNO_2 apex and

point R intersects the base line at T, giving the V. G. C. of the oil in the raffinate solution. The amount of P and N in this oil is

$$\left(\frac{P}{N}\right)_T = \frac{TN}{PT} \tag{2}$$

The ordinate of point R represents the amount of ΦNO_2 in the raffinate solution.

In the same way, a dotted line passing through point E intersects the base line at Ugiving the V. G. C. of the oil present in the extract, and the ordinate of point E gives the amount of solvent present in the extract. For U,

$$\left(\frac{N}{P}\right)_{U} = \frac{PU}{NU} \tag{3}$$

(4)

Further, the ratio of lines RM to ME gives the ratio of the amount of extract solution E to the amount of raffinate solution R; the total amount of R and E is represented by line RE and, if the amount RE is known, the amount of R and of E can be computed from

and

$$RE = RM + ME$$
$$\frac{E}{R} = \frac{RM}{ME}$$

Also, the ratio of raffinate oil to extract oil is indicated by the length of lines SU and TS, and

 $\frac{E}{R}$

$$\frac{T}{U} = \frac{SU}{TS} \tag{5}$$

The total oil in the two phases R and E is equivalent to the length of line TU. Thus the yield of raffinate oil may be measured by the ratio SU/TU. Then

Vol. % yield of raffinate
$$T = \frac{SU \times 100}{TU}$$
 (6)

or vol. % yield of raffinate oil =

$$\frac{(V.G.C.extract - V.G.C.stock) \times 100}{(V.G.C.extract - V.G.C.raffinate)}$$
(7)

All these relations are exactly the same as the corresponding quantitative relations in an equilibrium diagram on triangular coördinates. The use of rectangular coördinates in the manner just outlined is convenient and in many ways preferable to the use of triangular coördinates. It is especially useful when an extension of the base line is desired. For example, if it is desired in Figure 2 to plot an extract solution containing an oil of 0.910 V. G. C., it is necessary only to step the base line up to the 50 per cent ordinate and reduce an abscissa division along this ordinate to half its size on the base line.

From the preceding we see that an analysis of the extract and raffinate phases in any extraction can be represented on a triangular diagram in terms of composition, amounts of solutions, and yields of solutions or oils. Extractions have also



FIGURE 3. EQUILIBRIUM CURVE FOR 0.85 V. G. C. OIL AND NITROBENZENE AT 50° F., SHOWING ONE-STAGE TIE LINES AND TWO- AND THREE-STAGE CONNECTING LINES

been made by the B. C. C. process using two and three stages. Several of these are indicated in Figure 3; here it is evident that the connecting lines between the final extract and raffinate phases have different slopes from those of the tie lines for the single-stage treats, being flatter as the number of stages increases. Also, the final extract and raffinate solutions are not in equilibrium with each other (each stage, however, is in equilibrium), and for this reason a line connecting such points is called a "connecting" line rather than a "tie" line. It is not necessarily true that the extracts and raffinates for the one-, two-, and three-stage treats should all fall on the same equilibrium line for a system where there is decided mutual solubility. Laboratory results indicate that there is a difference in the positions of the equilibrium line for the same oil when treated in single-, two-, and three-stage B. C. C. treats. However, the results of such different stages of treating were sufficiently close to the one-stage equilibrium line so that an average or smoothed equilibrium line drawn through one-, two-, and



4. Comparison of Results of a Countercurrent Extraction with a B. C. C. Extraction FIGURE 4.

three-stage points was assumed to be valid and could be used without appreciable error, for the purpose of indicating saturated solution compositions which might be obtained in any form of countercurrent treating. This assumption has been made previously in the literature (8).

Use of the Equilibrium Diagram

Equilibrium curves made up in the way described were used as the basis for comparing the results of tower extractions with batch extractions. To illustrate, an extraction was made with oil stock S, and extract and raffinate solutions-Eand R resulted as shown in Figure 4. Since a saturated solution is a solution which, when plotted, lies on the equilibrium curve, and the two points E and R do not lie on the equilibrium curve, then these are not saturated solutions. It has been found in the laboratory, as mentioned above, that even though the final extract and raffinate solutions from a multistage B. C. C. treat are not in equilibrium with each other, they do nevertheless fall on the equilibrium line or very close to it, and are considered to be saturated solutions and may be joined by a connecting line. The two points E and R, then, represent solutions made up as follows: E is a solution of an oil of V. G. C., equal to U, and solvent: more of the solvent is present than is necessary to produce a saturated extract solution, E'. This excessive amount of solvent is indicated by line EE'. R is a solution of an oil of V. G. C. equal to T, with solvent insufficient in amount to produce a saturated raffinate solution, R'. The insufficient quantity of solvent or the excessive amount of oil is indicated by line RR'. If the two points E'and R', representing saturated solutions containing the same quality of oils as is present in E and R, are joined, this connecting line is analogous to the connecting lines for one-, two-, and three-stage treats. The slope of the dotted line E'R' is approximately the same as the two-stage B. C. C. connecting line passing through the same mix (or solvent ratio) point. Thus the extraction which gave the two oils of quality T and U was produced by an operation equivalent to a two-stage B. C. C. treat. Defining "equivalent theoretical stages" (E. T. S.) as the number of stages of equilibrium batch countercurrent treating necessary to produce extract and raffinate oils of given qualities, then for this extraction the E. T. S. is 2.0.

The intersection of line E'R' with the ΦNO_2 -S line is the mix point, M', which shows that, if oil stock were subjected to two-stage B. C. C. treatment, using the solvent ratio indicated by M', there would result saturated solutions corresponding to points E' and R'. Mix point M shows the solvent ratio which was actually used to obtain the two solutions Eand R. Thus the difference between M and M' represents the difference between the solvent ratio required under the actual operating conditions and the solvent ratio necessary to produce the same V. G. C. of oils, or the same degree of separation of the oil stock by B. C. C. treating. And this gives a measure of the efficiency of use of solvent, remembering that solution E contained more than enough solvent required for a saturated solution, whereas solution R contained less solvent than was required for a saturated solution. In other words, the deviation of both E and R from saturated solutions is measured and accounted for by a single point, M', on the solvent line. Following this, the "over-all per cent efficiency" (% Eff.) of use of solvent is defined as the ratio of the amount of solvent necessary to produce extract and raffinate oils of given qualities by means of equilibrium batch countercurrent treating, to the amount of solvent actually used, or

% Eff. =

(solvent ratio required by B. C. C. treating) \times 100 (solvent ratio necessary under actual operating conditions) (8)

or
$$\%$$
 Eff. = $\frac{M' \times 100}{M}$ (8A)

The foregoing analysis of the two points E and R led to the definition and significance of E. T. S. and % Eff. Both E. T. S. and % Eff. were found to be necessary bases for the comparison of extractions. Comparisons which follow are made in terms of both these measurements.

Extraction Chart

Figure 5 is based upon B. C. C. data and the equilibrium curve. By means of such a chart it is possible to determine rapidly a large number of interrelated facts concerning an extraction without calculation from the equilibrium curve. The chart was constructed for an oil of 0.850 V. G. C. treated with nitrobenzene at 50° F. by B. C. C. process. It was built up from experiments and an equilibrium curve as follows:

1. Stages of B. C. C. treating were placed on the ordinate and solvent ratios on the abscissa.

2. For one, two, three, or more stages of B. C. C. treats, the quality of extract and raffinate oils resulting from treats of different solvent ratios was marked on the plot. Lines were then drawn connecting points of the same quality. Extract lines in



FIGURE 5. EXTRACTION CHART FOR 0.850 V. G. C. OIL AT 50° F.

Figure 5 curve downward from right to left; raffinate lines curve downward from left to right.

3. Yields of raffinate oil corresponding to the stages and the qualities of oils obtained were plotted at the corresponding intersections of lines on the chart. For intermediate points where data were not available, the yields were calculated from Equation 7. For constant-yield lines it is convenient to select a yield and a V. G. C. of extract, and to calculate the V. G. C. of raffi-nate. Thus the yield lines were plotted easily and consistently.

4. For various stages and solvent ratios the corresponding ratios of extract and rafinate solutions were plotted. Actual E/R values from experiments were used, although values could have been calculated by use of Equation 4.

The chart is really a composite of all the information that can be computed from the three-component equilibrium diagram. It is set up most conveniently from experimental data and calculations from the equilibrium curve, and presents them in a direct manner. A similar chart made by plotting temperature in place of stages, and for a given number of stages, is quite useful for determining temperature and other operating conditions required to produce desired separations and yields by a given set-up of countercurrent extraction equipment.

If it were possible to predict the results of multistage B. C. C. treats from single-stage data, as Hunter and Nash attempt to do for such an oil-solvent system, it would be possible to construct charts for any range desired and for any oil stock, simply from single-stage extractions of a stock. As indicated previously, this cannot be done at present for an oilnitrobenzene system.

Use of Extraction Chart

The chart presents a clear picture of the relations between the variables in B. C. C. treating. Following a raffinate oil V. G. C. line from left to right, the chart shows: (a) The extract oil becomes less naphthenic (the extraction less selective). (b) Lower yields of the raffinate oil are indicated. (c) Extract-raffinate solution ratios increase. (d) Fewer stages are required. (e) Larger solvent ratios are required. Further, if a given quality of raffinate is to be produced by a given number of stages, the solvent ratio required and the E/R ratio (showing quantities of solutions to be handled) can be read directly. Also, it is evident from the chart that for highest yields of raffinates, low solvent ratios and high number of stages are required.

To evaluate the results of an extraction, the chart may be used in this way. Suppose an extraction is made using a solvent ratio of 1.50 and results in a raffinate oil of 0.820 V. G. C.,

an extract oil of 0.892 V. G. C., and an E/Rratio of 2.5. It is then desired to check the material balance and analysis and to compare the separation with a separation made by B. C. C. treating. A point is marked on the chart (Figure 5) where the 0.820 V. G. C. line crosses the 0.892 V. G. C. line and corresponds to the results of the extraction made. Then by reading the chart:

The E/R ratio corresponds to 2.5 which checks experimental results and is in effect a material balance showing that the analyses of the extract and raffinate solutions are correct.

2. The yield of raffinate oil is 58 per cent and can be compared with the actual yield obtained by analyses and used as a check. 3. The equivalent theoretical stages (E. T. S.)

are 2.02. (The extraction could have been made using 2.02 stages of B. C. C. treating.) 4. The solvent ratio is 1.43, which gives the amount of solvent by 2.02 stages of B. C. C. treat-ing required to produce the extraction. Since the ing required to produce the extraction. Since the solvent ratio actually used was 1.50, then the over-all solvent efficiency is $(1.43 \times 100)/1.50 =$ 95.3 per cent (Equation 8).

Another important use of the chart concerns actual operation during tower extraction. The E/R lines on the chart are nearly straight and very nearly vertical. Thus for any solvent ratio that may be used, there is relatively little change in the E/R value from one to three stages compared to the

change in value as the solvent ratio is changed. Therefore, when a treat is made in a tower, equilibrium conditions will be reached when the E/Rratio becomes constant and when it reaches a value that can be predicted approximately by the known solvent ratio and an estimated E. T. S.

Extraction Tower

Figure 6 represents the extraction tower and auxiliary equipment.

The tower was made up of a glass cylinder $2^{15}/_{19}$ inches i. d., 6 feet high, and jacketed with a glass cylinder 6 inches in diameter. The feeds to the bottom and top of the tower were maintained at 50° F. $\pm 2^{\circ}$ in all runs. The jacket was filled with circulating water and maintained at temperatures varying between 46° and 50° F., as temperatures of feeds demanded.

The oil feed to the bottom of the column was held at constant rate by means of a proportioning pump, which also allowed adjustment to give a wide range of oil feed rates. The oil was introduced through a 1/2-inch brass pipe, the end of which was covered with a brass plate having three 1/8-inch holes. The oil inlet was 8 inches above the bottom of the glass cylinder and midway between the center line of the column and the inside wall of the glass. A stool to support the packing and to distribute the oil consisted of a brass disk 1/4 inch thick, $2^3/4$ inches in diameter, perforated with a large number of 3/8-inch holes, and supported $8^{1}/2$ inches above the bottom by three 1/8-inch brass rods resting on the metal bottom of the column. For the graded glass packing runs the tower was packed as follows: On the bottom distributor plates were placed 4 inches of No. 1 rings (Table IV gives the size, etc.), followed by 5 inches of No. 2 and No. 3 mixed, 8 inches of No. 4, 4 inches of No. 3, 8 inches of No. 5, and finally 9 inches of No. 7, making a total of 55 inches of packing. After many combinations this arrangement was found to give the best results for this type of packing. When the rate of oil feed was too great to be handled by the packing, it was found that, in this grading of packing, flooding of the column occurred at three positions in the tower simultaneously. These positions were 7, 37, and 48 inches above the bottom of the packing.

The product from the bottom of the column (extract) was taken out through a 1/2-inch brass pipe threaded into the base. Thus there was an 8-inch space between oil inlet and bottom outlet, and an 81/2-inch space between bottom outlet and the bottom of the packing. This space is called the settling section at the bottom. A corresponding settling section was provided at the top of the column. In several runs 13-inch settling sections were used. From the bottom outlet the extract flowed upwards through a loop vented at the top and then down into a product tank. The loop was arranged so that its top could be raised or lowered to any desired position in order to regulate the level of the interface between the extract and raffinate layers. A low position of the loop gave high interface levels or, in other words, allowed raffinate to occupy most of the column. A high position of the loop gave high interface levels and thus allowed extract to occupy most of the column. An interface was always observed to be present between the two immiscible phases in the tower. In the tower operation drops of oil (or raffinate) rose through the extract layer until the interface was reached where-



FIGURE 6. FLOW SHEET FOR COUNTERCUR-RENT LIQUID-LIQUID EXTRACTION Packed glass tower, 2¹⁵/₁₆ inches i. d., operated at 50° F. *T* indicates thermometer.

upon they became a part of the upper raffinate layer, and drops of solvent (or extract) fell through the raffinate layer until they reached the interface and became part of the extract layer. Thus the solvent (and extract) formed the discontinuous phase above the interface level, while the oil (and raffinate) formed the discontinuous phase below the interface level.

The rate of nitrobenzene fed to the top of the tower was controlled by the constant-head box and adjustable siphon as indicated in Figure 6. After cooling, the solvent entered the column at the top of the packing through a pipe identical with that used for oil at the bottom, except that the pipe was bent in such a way that the outlet was directly in the center line of the column. Furthermore, the top of the packing was always arranged in a concave shape so that the outlet was approximately 1/4 inch below the uppermost packing at the walls of the column.

The nitrobenzene inlet for runs when 55 inches of packing were used was $8^{1/2}$ inches below the top product outlet. For lower heights of packing the nitrobenzene inlet tube was lowered accordingly, and the space above the packing (and nitrobenzene inlet) was partially filled with long glass rods for the purpose of reducing the quantity of raffinate in the tower above the packing. Product from the top of the column (raffinate) was removed through a pipe threaded into the top plate of the tower.

A connection is shown at the bottom outlet of the tower whereby part of the extract could be drawn through a pump and mixed with incoming oil. This arrangement was used in a number of experiments which are indicated in the data as runs with "recirculation."

Operation of the Tower

The following procedure was found to be the most satisfactory for starting and operating the tower. Cold water is cir-

culated through the chillers for solvent, oil, and recirculating extract, and also through the jacket. After cooling water temperatures become constant, solvent flow is started but oil flow is not started until enough solvent is in the column to cover the oil inlet. The extract loop is set to give approximately the desired interface level in the column. Flows and temperatures are then adjusted as desired, and the column is allowed to fill. Gradually in the top of the column a raffinate layer separates and displaces extract until the level so formed between the raffinate and extract phase remains constant. Flows of raffinate

and extract then become steady, and the extract loop may be raised or lowered as required to adjust the interface level to the exact position desired. As a run progresses, slight adjustments of the level are necessary because of the change of



Figure 7. Typical Curve Showing Time Required to Reach Steady E/R Conditions (Run 53)

the specific gravity of each phase as the process approaches equilibrium. Samples of extract and raffinate are taken every 20 or 30 minutes. These are weighed, and temperatures and gravities are determined. From these data volumes of extract and raffinate solutions are calculated at 60° F., and the E/R ratio is found. The calculations for E/R can be made quickly, and when succeeding determinations are constant, it is assumed that the column has reached equilibrium operating conditions. If E/R values are plotted against

time, curves similar to that in Figure 7 are obtained. Most of the runs required from 4 to 5 hours to reach equilibrium. This method for determining when the tower is operating at equilibrium was found to be very reliable and rapid. Specific gravity determinations alone are not dependable, nor is the assumption that equilibrium is reached when the interface level becomes steady. After the tower operation is at equilibrium, samples of extract and raffinate are taken for analysis, and data obtained for over-all material balances. The samples are analyzed and evaluated as described.

Experiments and Variables

Table II gives the results of a number of extractions made in the tower. All the data presented are from observation and analysis. By plotting the results either on the equilibrium curve (as in Figure 4) or on the chart (Figure 5), material balances and analytical data were checked. Material balances in all runs were within 5 per cent of the materials fed to the tower. Many of the runs shown were made with graded glass packing, and a number of variables were evaluated using such packing.

The following variables were studied in order to determine their effect on one another, their relative importance, and their quantitative influence on extraction: packing materials and shapes, sizes and arrangement of packing, recirculation of extract, solvent ratio, rate of feed (or throughput), interface level (or effect due to different phases being dispersed), height of packing, wetting of packing.

The results of the experiments were compared on the basis of the following: V. G. C. of extract and of raffinate oils produced (a measure of the degree of separation); yield of raffinate; nearness of each phase to saturation (by plotting on the equilibrium diagram); equivalent theoretical stages (E. T. S.) resulting from a given height of packing; over-all per cent efficiency of use of solvent (% Eff.), which is a direct measure of the deviation from saturation conditions; and finally, observation of the action in the tower through the glass jacket. For simplicity, comparisons can be made using only the E. T. S. and the % Eff. (for any given packing and height of packing), because, when these two measurements are given, the separation, yield, and saturation conditions are easily determined by reference to Figure 5. Thus a high value of E. T. S. and of % Eff. indicate that the tower has effected a better separation for the same amount of packing and solvent than is the case for low values of E. T. S. and % Eff.

Table III gives the results of the data for graded glass packing runs in terms of E. T. S. and % Eff. These data were taken from experiments made under widely varying operating conditions. As will be shown later, the quantitative effect of the principal operating variables was determined, and by means of such information the results of all experiments with graded glass were calculated to the same basis for comparison. Table II shows that the average deviation of the results is approximately 6 per cent. This figure seems to represent the accuracy of the experiments and attests to their reproducibility.

TABLE II DATA FROM COUNTERCURRENT EXTRACTION IN A GLASS TOWER AT 50° F.

Expt. No.	Pack- ing ^a	Height of Pack- ing	Inter- face Level % above	Oil Feed Rate	Sol- vent Ratio	Equiva- lent Theo- retical Sol- vent Ratio	% Eff.	E. T. S.	V. G. C. Raffi- nate Oil	V. G. C. Ex- tract Oil	Actual Instantaneou E/R at End of Run	¹⁸ Yield of Raffi- nate Oil
$1 \\ 2 \\ 3 \\ 99 \\ 40 \\ 423 \\ 44 \\ 445 \\ 446 \\ 47 \\ 449 \\ 501 \\ 525 \\ 52$	АААВНН СССССС СССССС	1nches 56 56 56 45 55 55 55 55 55 55 55 55 55	50000 51 14 80 83 74 25 33 40 29 40 78 78 78 78 78 73 78 82 80 70	Gal./hr. 2.60 3.31 3.31 0.810 0.940 0.954 0.372 0.256 0.347 0.291 0.691 0.491 0.598 0.398 0.319 0.593 0.328 0.460	$\begin{array}{c} 1.25\\ 1.99\\ 2.10\\ 1.63\\ 1.55\\ 1.59\\ 2.52\\ 0.95\\ 3.18\\ 2.40\\ 1.53\\ 3.06\\ 1.30\\ 1.52\\ 1.30\\ 1.52\\ 1.48\end{array}$	$\begin{array}{c} 1.00\\ 0.50\\ 1.02\\ 1.02\\ 1.14\\ 2.23\\ 0.67\\ 2.28\\ 1.95\\ 1.28\\ 2.75\\ 1.07\\ 1.29\\ 3.20\\ 1.27\\ 2.84\end{array}$		$ \begin{array}{c} <1 \\ <1 \\ <1 \\ <1 \\ .77 \\ 1.88 \\ 2.96 \\ 3.10 \\ 2.40 \\ 2.45 \\ 2.05 \\ 2.45 \\ 2.65 \\ 2.22 \\ 2.05 \\ 2.215 \\ 2.05 \\ 2.95 \\ 1.95 \end{array} $	$\begin{array}{c} 0.840\\ 0.841\\ 0.837\\ 0.830\\ 0.827\\ 0.825\\ 0.814\\ 0.813\\ 0.813\\ 0.813\\ 0.810\\ 0.813\\ 0.818\\ 0.810\\ 0.820\\ 0.820\\ 0.820\\ 0.806\\ 0.822\\ 0.810\end{array}$	0.882 0.850 0.852 0.894 0.895 0.895 0.895 0.885 0.885 0.885 0.886 0.889 0.896 0.881 0.898 0.895 0.877 0.894 0.884	$\begin{array}{c} & \cdot & \cdot \\ & 2 & \cdot 14 \\ & 2 & \cdot 02 \\ & 2 & 97 \\ & 5 & 25 \\ & 1 & \cdot 47 \\ & 6 & \cdot 57 \\ & 2 & \cdot 50 \\ & 6 & \cdot 57 \\ & 1 & 83 \\ & 2 & \cdot 45 \\ & 1 & 2 & 50 \\ & 2 & \cdot 55 \\ \end{array}$	61. 70 69.5 63.6 59.5 62.6 64.3 562.6 64.3 56.6 64.3 56.0 48.4 71.4 51.4 51.4 51.4 51.5 61.5 638.0 61.2 88.0 61.2 8
53 54 55 56 57 58 50 61 62 63 64 65 666 67 689 712 72 73	GGGGGGGG IJBCCCC KKKLLL	55 55 55 55 55 55 42 42 30 18 55 55 55 55 55 55 55 55 55 55 55 55 55	79 80 877 80 878 80 82 80 82 80 82 80 78 82 80 78 78 78 78 78 78	$\begin{array}{c} 0.300\\ 0.530\\ 0.95\\ 0.485\\ 0.452\\ 0.452\\ 0.562\\ 4.663\\ 2.804\\ 1.836\\ 1.015\\ 0.940\\ 0.806\\ 0.864\\ 0.707\\ 0.715\\ 0.715\\ 0.628\\ \end{array}$	$\begin{array}{c} 3.26\\ 3.14\\ 3.66\\ 3.14\\ 3.15\\ 2.66\\ 2.72\\ 3.05\\ 3.05\\ 3.08\\ 3.09\\ 3.04\\ 1.50\\ 3.08\\$	$\begin{array}{c} 2.63\\ 2.70\\ 3.03\\ 2.28\\ 2.53\\ 1.44\\ 0.70\\ 0.96\\ 2.76\\ 2.92\\ 1.43\\ 1.90\\ 1.87\\ 1.69\\ 0.92\\ 1.19\\ 1.97\\ 1.79\\ 1.82\end{array}$	81.5 86.0 82.7 80.23 54.2 90.57 90.57 90.57 90.57 60.5 55.65 60.5 60.5 55.65 64.63 59.5	$\begin{array}{c} 1.30\\ 1.95\\ 2.45\\ 1.92\\ 1.60\\ 0.85\\ 1.60\\ 0.85\\ 1.30\\ 1.53\\ 1.30\\ 1.90\\ 2.02\\ 1.58\\ 1.52\\ 1.58\\ 1.80\\ 1.60\\ 2.20\\ 1.65\\ \end{array}$	0.810 0.811 0.805 0.814 0.812 0.819 0.828 0.829 0.816 0.829 0.816 0.820 0.820 0.820 0.820 0.820 0.820 0.821 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.828 0.817 0.829 0.817 0.829 0.817 0.829 0.817 0.829 0.817 0.829 0.828 0.834 0.829 0.834 0.829 0.836 0.837 0.838 0.838 0.838 0.837 0.838 0.838 0.838 0.838 0.838 0.838 0.836 0.	0.882 0.881 0.882 0.884 0.884 0.884 0.882 0.886 0.887 0.892 0.895 0.879 0.892 0.878 0.879 0.892 0.886 0.886 0.886 0.888 0.888 0.885 0.885	$\begin{array}{c} 7.26\\ 7.05\\ 8.00\\ 5.33\\ 6.70\\ 4.30\\ 5.50\\ 3.70\\ 4.60\\ 9.85\\ 2.50\\ 5.50\\ 5.50\\ 5.50\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.88\\ 5.50\\ 5.80\\$	$\begin{array}{c} 14.4 \\ 44.3 \\ 44.3 \\ 40.0 \\ 52.7 \\ 47.7 \\ 62.7 \\ 62.7 \\ 62.7 \\ 68.2 \\ 45.2 \\ 45.2 \\ 45.2 \\ 55.0 \\ 56.8 \\ 63.7 \\ 51.3 \\ 54.0 \\ 54.0 \end{array}$

o packing.

A B

G. H.

No packing. 1/2 inch, Glass rings, $1/4 \times 3/8$ inch. Graded packing arrangement. Unglazed stoneware rings (3/8 inch) with first 5 inches from bot-tom of 1/2-inch glass rings.

^b No recirculation.

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Unglazed stoneware saddles, 1 inch. Unglazed stoneware saddles, 1/2 inch. Carbon rings, 1/2 inch. 30 inches of 1/2-inch carbon rings, under 25 inches of 1/4-inch carbon rings.

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Run No.	Height of Pack- ing In.	E. T. S. Calcd.	Deviation from Mean	% Devia- tion from Mean	% Eff. Caled.	Deviation from Mean	% Devia- tion from Mean
$\begin{array}{r} 423\\ 445\\ 445\\ 49\\ 53\\ 55\\ 55\\ 56\end{array}$	85 85 85 85 85 85 85 85 85 85 85 85 85 8	$\begin{array}{c} 2.49\\ 1.88\\ 2.35\\ 2.30\\ 2.76\\ 2.58\\ 2.51\\ 2.43\\ 2.51\\ 2.34\\ 2.30\\ 2.34\\ 2.53\\ 2.53\\ \end{array}$	$\begin{array}{c} 0.10 \\ -0.51 \\ -0.09 \\ 0.37 \\ -0.012 \\ 0.04 \\ 0.32 \\ -0.05 \\ -0.05 \\ 0.14 \end{array}$	$\begin{array}{c} 4.2\\ 21.4\\ 1.7\\ 3.8\\ 15.5\\ 0.2\\ 1.7\\ 13.4\\ 2.1\\ 3.8\\ 5.9\end{array}$	$\begin{array}{c} 82.7\\ 86.6\\ 71.2\\ 81.5\\ 93.0\\ 80.2\\ 91.4\\ 79.0\\ 94.5\\ 85.7\\ 86.0\\ 88.1\\ 92.0\\ \end{array}$	$\begin{array}{c} 2.8 \\ 1.1 \\ -14.3 \\ -4.0 \\ 7.5 \\ -5.3 \\ 5.9 \\ -6.5 \\ 9.0 \\ 0.2 \\ 0.5 \\ 2.6 \\ 6.5 \end{array}$	$\begin{array}{c} 3.3 \\ 1.3 \\ 16.8 \\ 4.6 \\ 8.8 \\ 6.2 \\ 6.9 \\ 7.6 \\ 10.5 \\ 0.2 \\ 0.6 \\ 3.0 \\ 7.6 \end{array}$
Av.	• •	2.39		6.2	85.5		5.9
$\begin{array}{c} 50 \\ 52 \end{array}$	55 55	$\substack{\textbf{2.01}\\\textbf{1.81}}$	No recirc No recirc	ulation sulation	$\begin{array}{c} 83.9\\ 85.5\end{array}$	No recir No recir	culation culation
Av.	• •	1.91	• • • •	• •	84.7		
57 58 59 60 ª Sol	42 42 30 18 lvent ra	2.23 2.22 1.91 1.16 tio = 2.0	; oil rate	 = 0.40 gal.,	77.1 83.4 62.9 54.4 /hr.; leve	= 80%	of packing
neight;	recircu	lation.					

TABLE III. SUMMARY OF RUNS WITH GRADED GLASS PACKING CALCULATED TO THE SAME SOLVENT RATIO, OIL RATE, AND INTERFACE LEVEL⁴

Packing Material

Glass, unglazed stoneware, and carbon were the materials used. The shapes were rings, Berl saddles, and short rods. The sizes are indicated in footnote^{α} of Table II and in Table IV; the latter also shows areas, free space, etc., which were determined in the laboratory. (Several runs are shown that were made with no packing in the tower. These runs gave poor results as indicated by plotting on the equilibrium curve and on the chart or as shown by the E. T. S. and % Eff.)

		TAI	BLE IV.	DATA O	N PACKING			07 E.a.a
Packing	$\stackrel{\frown}{{}}{}^{\mathrm{D}}_{In.}$	imensio B In.	$\int_{t}^{2ns^a}$	Surface Area per Piece Sq. ft.	Pieces per Cu. Ft. Packed Space	Surface per Cu. Packed space Sq. ft.	e Area Ft. of: Free space Sq. ft.	Space When Packed (Voids)
No. 1 No. 2 No. 3 No. 4 No. 1 + 2 mixed No. 2 + 3 mixed	$\begin{array}{c} 0.50 \\ 0.35 \\ 0.28 \\ 0.25 \\ 0.16 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\ 1 \\$	$\begin{array}{c} 0.75 \\ 0.50 \\ 0.43 \\ 0.38 \\ 0.37 \\ \end{array}$	$\begin{array}{c} 0.04 \\ 0.04 \\ 0.04 \\ 0.04 \\ 0.02 \\ \end{array}$	$\begin{array}{c} 0,01595\\ 0,00751\\ 0,00498\\ 0,00369\\ 0,00251\\ 0,01173{}^{b}\\ 0,00624{}^{b} \end{array}$	$\begin{array}{c} 12,000\\ 25,500\\ 58,400\\ 75,200\\ 148,500\\ 16,600\\ 35,900 \end{array}$	191.5191.8291.5277.5372.0195.0224.0	$\begin{array}{r} 245.5\\ 270.0\\ 520.0\\ 475.0\\ 586.0\\ 275.0\\ 346.0 \end{array}$	$\begin{array}{c} 78.0 \\ 71.0 \\ 56.0 \\ 58.5 \\ 63.5 \\ 70.8 \\ 64.7 \end{array}$
Glass rod: No. 6 No. 7 Stoneware rings Porcelain rings	$\begin{array}{c} 0.31 \\ 0.18 \\ 0.38 \\ 0.38 \end{array}$	${}^{0.41}_{0.25}\\{}^{0.38}_{0.38}$	0.06 0.08	$\begin{array}{c} 0.00398 \\ 0.00133 \\ 0.00528 \\ 0.00533 \end{array}$	31,400 162,200 26,200 27,200	$125.0 \\ 217.0 \\ 138.3 \\ 143.8$	$\begin{array}{c} 831.0 \\ 613.0 \\ 210.0 \\ 222.5 \end{array}$	$37.8 \\ 35.4 \\ 65.9 \\ 64.6$
Carbon rings: ^{1/2} in. ^{1/4} in. Stoneware saddles	0.50	0.50 0.25	0.06	$\begin{array}{c} 0.01043\\ 0.00240\end{array}$	10,800 90,300	$\substack{112.5\\216.5}$	$\begin{array}{c} 147.0\\ 451.0\end{array}$	$\begin{array}{c} 76.5 \\ 48.0 \\ \end{array}$
$\frac{1 \text{ in.}}{\frac{1}{2} \text{ in.}}$	1.00 0.50	$1.00 \\ 0.50$	$\begin{array}{c} 0.06 \\ 0.06 \end{array}$	$0.03780 \\ 0.00998$	$1,660 \\ 11,700$	$\begin{array}{r} 63.2 \\ 117.0 \end{array}$	$85.7 \\ 162.5$	$\begin{array}{c} 75.6 \\ 72.0 \end{array}$
• t) A +	_		† ₿ ↓			the lo ⁻ and pa Table be han size or

Glass cylinders or rings were used for the majority of the runs. In general they produced good results and had the advantage of allowing greater throughput of oil than equivalent sizes of other packings except saddle shapes, and for experimental work they allowed excellent visibility. Glass rods were used only in connection with graded ring packing and were not studied separately. Unglazed stoneware rings gave approximately the same results as glass and allowed better throughput of oil than did carbon packing. Unglazed stoneware Berl saddles allowed very high throughput of oil and gave poorer results than the same size glass and carbon rings.

Carbon rings showed low % Eff., fairly high E. T. S., and very low oil throughput.

Packing Size

Relatively few sizes of packing were tested (Table IV). The experiments showed that better extraction was obtained with small packing than with large. Good distribution, good dispersion, and very little channeling on the wall were characteristic of all packing sizes of 1/2 inch or less. Much

	Rate of Oil Input above Whic Flooding Occurs at a Solvent Rat of 2.0 Gal per Hr							
Packing	Packing No.4	No recirculation	Recirculation 3.8 gal. per hr.					
Glass rings: $\frac{1}{2}$ in. $\frac{1}{4} \times \frac{3}{5}$ in. Graded	B C G	1.00 0.36 0.33	$1.90 \\ 1.10 \\ 0.70$					
Stoneware rings, 3/s in.	н	0.30	1.00					
Carbon rings: $\frac{1}{2}$ in. Graded	$_{ m L}^{ m K}$	0.20 0.10	0.75 0.65					
Saddles: 1 in. 1/2 in.	\mathbf{I}	>10.50 3.00	>10.50 3.50					

greater throughput was obtained with large packing than with small (Table V). The oil used in this work had a viscosity of approximately 9500 seconds at 50° F., the temperature of

ely 9500 seconds at 50° F., the temperature of operation. Because of this high viscosity large packing was necessary in order to allow high rates of oil flow. As extraction took place, the viscosity of the raffinate decreased and smaller packings could be used to handle the same quantity of oil. Graded packing, as described under "Extraction Tower," gave a good balance between large-size packing for optimum throughput and small-size packing for best extraction. It was therefore found advisable to use different sizes of packing in different portions of the tower, and the experiments with graded packing gave better results than comparable experiments with uniform packing.

For any size of packing there was a definite limit to the amount of oil which could be handled by it, thus limiting the throughput. When oil was fed at too rapid a rate, a cross section of the tower would become filled or flooded with oil and gradually a layer of oil would be formed, filling

the lower part of the column until the oil short-circuited and passed out of the tower by way of the extract outlet. Table V gives the maximum rates of oil feed which could be handled by the different packings. The effect of packing size on the maximum oil feed rate is exceedingly important for the nitrobenzene-oil system used when comparing installation and operating costs for commercial packedtower equipment with batch mixing and settling equipment as commonly used.

No difficulty was experienced with flooding the column with solvent from the top, and it was possible to use very small packing at the top of the column without any danger of short-circuiting or lowering the solvent throughput. One successful run was made using a solvent ratio of over 7.

Recirculation of Extract

It was found advantageous to recirculate part of the extract to the bottom inlet of the tower and to mix it with the oil feed. Such recirculation and mixing gave two desirable results: (a) The maximum allowable oil feed (and throughput) was increased for any one packing, and (b) the E. T. S. was increased. Table V gives the data for various packings and shows that the effect of recirculation on maximum oil rate is not uniform

for different packings. For example, there is very little effect with 1/2-inch saddles. From the standpoint of throughput it is quite advantageous to recirculate extract and to mix it with oil feed. The principal reason for this is that the viscosity of the bottom feed is greatly reduced, allowing better flow up through the packing.



FIGURE 8. EFFECT OF RE-CIRCULATION OF EXTRACT ON EQUIVALENT THEORETICAL STAGES AND ON PER CENT EFFICIENCY FOR GRADED GLASS PACKING

Experiments are reported in Table II, both with and without recirculation of extract and mixing with oil. In Figure 8 for graded glass packing, runs without recirculation are plotted with runs with recirculation (calculated to the same conditions as those without recirculation); for an increase of recirculation from 0 to 3.8 gallons per hour there is an increase in the E. T. S. of 20.0 per cent (or 0.42 stage), while the % Eff. is not affected.

Settling Sections

All runs shown in Table II were made with settling spaces $8^{1/2}$ inches high. This height was found to be ample for both the top and bottom sections. Observations indicated that larger settling spaces are required at the top, or in the raffinate phase, than at the bottom for this oil-solvent system.

Solvent Ratio

Most treats were made using solvent ratios of 0.95 to 3.70. With other variables substantially constant, data from runs using different solvent ratios are plotted in Figure 9. These curves show for three types of packing the slopes of the lines



FIGURE 9. EFFECT OF SOLVENT RATIO ON EQUIVALENT THEORETICAL STAGES AND ON OVER-ALL SOLVENT EFFICIENCY FOR THREE TYPES OF PACKING (55 INCHES)

are nearly the same, and thus the effect of solvent ratio on E. T. S. is the same for these packings. The effect on % Eff. is likewise the same for the different packings. It is notable that the effect is the same for glass and carbon. The intercept of each of these curves on any one solvent ratio is, of course, different and will vary for different sizes of packings.

For graded glass an increase of solvent ratio from 1.5 to

3.5 produces a decrease in E. T. S. of 18.4 per cent and a decrease in % Eff. of 5.5 per cent. The decrease in % Eff. is small and does not appear to be significant, since it is probably within the limits of the experimental error. The following relations are valuable for calculating data for graded glass packing from any treat as given in Table II to a given treat for comparison as in Table III. From Figure 9

E. T. S. =
$$2.85 - 0.0023 SR$$
 (9)
% Eff. = $97.2 - 0.033 SR$ (10)

Since the slopes of the carbon ring lines and uniform glass lines are the same, the same factors in Equations 9 and 10 can be applied to these types of packing also, the differences being in the constants of the equations.

Rate of Feed (Throughput)

The relation of size and type of packing to throughput and a method for increasing throughput by recirculation of extract have been discussed. The relation of throughput to extraction was determined by a series of experiments using graded glass, and is shown in Table II where variables other than oil feed rate were held substantially constant. When these data are plotted, the resultant curves are approximately



FIGURE 10. EFFECT OF INTERFACE LEVEL ON EQUIVALENT THEORETICAL STAGES AND ON OVER-ALL SOLVENT EFFICIENCY FOR TWO TYPES OF PACKING

straight lines, from which it is seen that, when the oil feed rate is increased from 0.1 to 0.6 gallon per hour, there is a decrease in the E. T. S. of 18.6 per cent and an increase in the % Eff. of 7.6 per cent. Again it is to be noted that the change of % Eff. is probably not significant. The following equations result from the curves:

E. T. S.
$$= 2.49 - 1.0 OR$$
 (11)

$$\% \text{ Eff.} = 79.5 + 12.2 \, OR$$
 (12)

Interface Level

The position of the interface level has an effect on the contacting and extraction; in other words, the fact that one phase or the other is continuous affects the extraction. The height of the interface level above the bottom of the packing is given as a percentage of the total height of packing in Table II. Data from comparable runs are plotted in Figure 10. Again there is striking similarity between glass and carbon packing. For an increase in level in graded glass packing from 25 to 75 per cent of packing height, the E. T. S. are lowered by 15.8 per cent and the % Eff. is lowered 7.6 per cent. Again, the decrease in % Eff. is hardly significant. The following equations derived from Figures 11 and 12 are



FIGURE 11. EFFECT OF PACKING HEIGHT ON E. T. S. (RECIRCULATION)

useful for relating graded glass packing extractions to the same conditions of interface level:

E. T. S. =
$$3.27 - 0.0096 L$$
 (13)
% Eff. = $92.8 - 0.136 L$ (14)

A much longer time was required for equilibrium conditions to be reached for low-level than for high-level runs, because (a) larger quantities of raffinate must be produced for lowlevel runs, (b) at lower levels extraction is better, (c) there are higher E. T. S., and (d) lower yields of raffinate are obtained (Figure 5 shows an increase in E. T. S.). Slightly higher throughputs of oil can be obtained by high interface levels, largely because of the lower resistance to flow of the less viscous extract layer.

Height of Packing

Most runs were made using 55 inches of packing; and when the number of E. T. S. is known, the average height of packing equivalent to one theoretical stage (H. E. T. S.) can be computed. However, it was desirable to know more definitely how packing height affected E. T. S. and also % Eff.; therefore runs were made as shown in Table II with lower heights of packing. Assuming that the same relations between solvent ratio, oil rate, interface level, E. T. S., and % Eff. hold for these lower heights of packing, the data were



FIGURE 12. EFFECT OF PACKING HEIGHT ON OVER-ALL SOLVENT EFFICIENCY (RECIRCULATION)

calculated to a uniform basis by the use of Equations 9 to 14, and are plotted in Figures 11 and 12. It is evident from the shape of the curves that for E. T. S. the uniform packing re-

TABLE VI. DATA ON WETTING OF PACKING								
$Packing^a$	Packing No. ^b	Raffinate Held on Surface Lb./sq. ft.	Wetting Ratio, Rafinate to Extract	Ratio by nal to Subse- quent wetting of raffi- nate sur- face by extract	Wt., Orig Final Subse- quent wetting of extract surface by raffi- nate	Ability for Raffinate Compared to Glass as Unity		
Glass rings, 1/2 in. Glass rings, 1/4 × 3/8 in. Stoneware rings, 3/8 in. Porcelain rings, 3/8 in. Carbon rings, 1/2 in. Saddles, 1/2 in.	B C H J	$\begin{array}{c} 0.00428\\ 0.00428\\ 0.00740\\ 0.00880\\ 0.02240\\ 0.00860\\ \end{array}$	$1.125 \\ 1.125 \\ 1.388 \\ 2.350 \\ 0.874 \\ 1.530$	$1.13 \\ 1.13 \\ 1.92 \\ 2.35 \\ 1.06 \\ 2.10$	$\begin{array}{c} 0.880 \\ 0.880 \\ 0.564 \\ 0.415 \\ 0.922 \\ 0.520 \end{array}$	1.001.001.732.055.222.02		
^a A low interface level gave the best over-all tower extraction performance in all cases.								

quires increasingly more packing height for additional stages and, although the same is true for graded packing, the increase in height required for additional stages is less than is the case with uniform packing. From these curves the H. E. T. S. can be determined for various conditions of solvent ratio, oil rate, and interface level. The curves in Figure 12 indicate that 100 % Eff. is approached rapidly as packing height increases and that an estimate of the height of packing necessary to produce 100 per cent efficient use of solvent (H. E. 100 % Eff.) can be made. It should be noted that uniform glass packing gives approach to 100 % Eff. more rapidly with respect to height of packing than does graded glass, which in turn is better than carbon. This follows from the slope of the lines in Figure 12, even though the sizes of packing are not comparable.

Wetting of Packing

Experiments were made to determine preferential wetting of the various packings used and to find a correlation between wetting, interface level, and % Eff. Table VI summarizes these data. Weighed amounts of packing were wet with ex-



FIGURE 13. RELATION OF EQUIVA-LENT THEORETICAL STAGES AND OVER-ALL SOLVENT EFFICIENCY WITH SOLVENT RATIO FOR TWO INTERFACE LEVELS AND THREE RATES OF OIL FEED

tract solution and with raffinate solution. The amounts of each, held on similar packing, were determined by weight after uniform draining of the samples at constant temperature. No valuable correlation was evident between these wetting data and the behavior of the packing in the column. For example, both glass and carbon have the same relation to change of E. T. S. and % Eff. with regard to solvent ratio and interface level, whereas glass is more easily wet with raffinate than with extract, and carbon is more easily wet with extract than with raffinate. Also, carbon holds more than five times the quantity of raffinate than does glass, but this does not seem to have an effect on the over-all extraction. If preferential wetting were an appreciable factor in this oil-solvent system, it would be expected that there would be some noticeable difference in behavior of glass and carbon with respect to a variable such as interface level.

Correlation of Principal Variables

Solvent ratio, oil rate, and interface level were the three principal variables evaluated from the standpoint of control of extraction and economics in design of equipment. By means of Equations 9 to 14 the data for runs with 55 inches of graded glass packing were calculated to various solvent ratios, oil rates, and interface levels (one set of which is given in Table III) and are plotted in Figure 13. These curves give a composite picture of the interrelation of these variables with E. T. S. and % Eff. Such plots are useful together with the chart (Figure 5) to determine optimum solvent ratio, oil rate, and interface level to be used with a given height of packing at a definite temperature to produce a given quality and yield of oil.

Summary

Countercurrent liquid-liquid extraction in a packed tower can be evaluated by laboratory equilibrium batch countercurrent extractions in terms of equivalent theoretical stages and over-all per cent efficiency of solvent. Economics of extraction by means of a packed tower are largely dependent upon the over-all per cent efficiency of the solvent.

A direct method for evaluating a tower extraction and relating it to equilibrium data of a three-component system has been discussed and given in the form of an extraction chart.

Experimental results were reproducible, and for a series of runs made over wide ranges of solvent ratio, oil rate, and interface level calculated to the same conditions, the results show an average deviation of approximately 6 per cent from the mean.

Data fundamental to the design of liquid-liquid packed tower equipment are presented for a particular solvent-oil system, and variables entering into the design of such equipment may be summarized as follows:

1. There is little difference in extraction using glass, stone-

 There is notice unreference in contraction of equivalent size.
 There is a limit to the amount of oil that can be handled by a given size of packing. Throughput is lower but extraction is

4. Recirculation of extract and mixing it with incoming oil increases the oil throughput and the E. T. S.

Increases the on throughput and the E. T. S.
5. Settling spaces required are small and are dependent upon the size of packing nearest the space.
6. For a given height of packing, as the solvent ratio increases the E. T. S. and the % Eff. both decrease. The effect is greater on the E. T. S.

7. For a given height of packing, as the oil rate increases the E. T. S. decrease and the % Eff. increases. The effect is greater on the E.T.S.

Low interface levels (or large continuous raffinate phase) 8. give highest E. T. S. and % Eff.

Preferential wetting of the packing seems to have no appreciable effect on the extraction.

Acknowledgment

The experimental work on which this paper is based was carried on with the coöperation of the Research and Development Department of The Atlantic Refining Company, in the Chemical Engineering Laboratory of the Drexel Institute of Technology.

Nomenclature

- B. C. C. = batch countercurrent
 - E= extract solution
- E/R = extract-raffinate solution ratio E. T. S. = equivalent theoretical stages
- H. \overline{E} . \overline{T} . \overline{S} . = height of packing equivalent to one theoreti-
- cal stage
- H. E. 100% Eff. = height of packing equivalent to 100 per cent efficient use of solvent L = interface level, per cent of packing height
 - above bottom of packing
 - $M = \min$
 - = naphthenic N
 - OR =oil rate, gallons per hour P =paraffinic

 - % Eff. = over-all per cent efficiency of use of solvent R= raffinate solution
 - S =stock to be extracted
 - SR = solvent ratio, by volume
 - T = raffinate oil U = extract oil
 - V. G. C. = viscosity-gravity constant
 - $\Phi = \text{solvent}$
 - $\Phi NO_2 = nitrobenzene$

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RECEIVED January 11, 1937.

Corrections

Recently it was called to our attention that an error occurs in the synopsis of our article on "Compressibility of Butane-Air Mixtures below One Atmosphere" [IND. ENG. CHEM., 28, 870 (1936)]. Through an oversight we gave the maximum deviation of the compressibility of butane-air mixtures as 23 per cent from that expected from the additive rule for gas mixtures; we should have said that the compressibility coefficient varied a maximum of 23 per cent. Actually the compressibility of the mixture in question showed a maximum deviation of 0.3 per cent.

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F. W. JESSEN

Our attention has been called to an error in our recent article On "Resinous Derivatives of Vinyl Alcohol" [IND. ENG. CHEM., 28, 1123 (1936)]. The product referred to by Regnault in the first paragraph is a substituted vinyl chloride (according to modern nomenclature, α -chlorovinyl chloride or unsym-dichloroethylene) and not vinyl chloride as commonly understood. The reference was made for the purpose of establishing the earliest date on record at which the polymerization of a vinyl compound had been described, and we feel sure that the import of our comments is not materially changed by this correction which, nevertheless, is made with pleasure for the sake of accuracy.