THE JOURNAL OF THE American Oil Chemists' Society

Volume 29 MARCH, 1952 No. 3

Improvements in the Simple Distillation of Fatty Acids by Continuous Methods[1]

RALPH W. BERGER, WURSTER & SANGER INC., Chicago, Illinois

RECENT developments in the fractional distillation of fatty acids and the consequent emphasis of this process in the literature have overshadowed the older but nevertheless highly important process of simple distillation or stripping. Improved results in the simple distillation of fatty acids are obtained by continuous operation under high vacuum and relatively low temperature with indirect heating by Dowtherm vapors. Continuous vacuum drying and deaerating of the crude feed stock, efficient removal of entrainment from the vapor stream, continuous cooling of the distilled product prior to discharge to the atmosphere, and automatic discharge of the residue also contribute to improved results. Although the successful operation of most continuous processes depends upon long periods of operation on a feed of constant composition, the continuous fatty acid distillation plant is designed with a low hold-up that permits relatively short runs on feed stocks of different compositions with little interruption of the process and a minimum of mixing of the products. The continuous process is as flexible as the batch process in this respect. Automatic instrument control aids in quickly re-establishing equilibrium after a change in feed stock.

Although these principles are generally recognized, some continuous fatty acid distillation processes have been only partially successful because one or more of these features have been overlooked or inadequately handled. Another factor contributing to the unsatisfactory operation of some processes is the designer's ignorance or disrespect for the heat sensitivity of fatty acids. In such cases only the very low vapor pressure of the fatty acids is considered, and the resulting designs are merely adaptations of processes for petroleum stocks with similar vapor pressure.

The following discussion applies specifically to a recently developed design embodying both generally recognized principles and a true appreciation for the heat sensitivity of fatty acids. This process has been in highly successful, full-scale commercial operation since 1946.

Drying and Deaerating

In operation, following the general diagram in Figure 1, the crude fatty acids are fed to the dryer and deaerator, Item 2. A dry feed stock is essential for the uniform operation of the distilling equipment. On account of the high specific volume of water vapor under the conditions of operation of the distilling equipment, the introduction of varying amounts of moisture and perhaps slugs of water in the feed stock would cause entrainment due to surging and may lead to instability in the vacuum equipment. The effect of the latter is to vary the distillation rate and condenser load as well as the respective operating temperatures. A deaerated feed stock is desirable to reduce oxidation losses in the subsequent distillation. Both of these objectives are most satisfactorily achieved by heating the crude feed stock to a temperature of 150° to 200° F. while under a vacuum of 26″ to 28″ Hg. This results in a considerable improvement over the widely used method of holding the crude stock at a temperature of 220° F. for prolonged periods in open tanks with air agitation (3), thereby contributing to oxidation losses. Another method in use consists of preheating the crude stock and subsequently passing it into an evacuated flash drum, but the disadvantage of this method lies in the application of heat prior to deaeration.

Distillation

The dried and deaerated fatty acids are fed to the still, Item 4, in which the volatile components are stripped from the nonvolatile. Because of the heat sensitivity of the fatty acids it is essential that this operation be conducted at as low a temperature as is economically feasible and that the duration of the operation be reduced to a minimum. The effects of temperature and time on this operation are to cause losses of fatty acids by polymerization to tars which appear in the residue and by cracking to hydrocarbons which may contaminate the distilled product. These effects are minimized by conducting the stripping operation in a single pass through a bubble tray column at an absolute pressure of about 5 mm. Hg, maintaining a low liquid level on the trays and applying the heat through the surfaces of the trays themselves by means of condensing Dowtherm vapors within the trays, as illustrated in Figure 3.

With this unique heating method it is practical to distill fatty acids without heating above the vaporization temperature. Thus the maximum operating temperature, when distilling a mixture of fatty acids with a vapor pressure of 5 mm. Hg at 400° F., actually would be about 400° F. The maximum surface temperature to which the fatty acids are exposed is in the range of 525° to 625° F., and the time of exposure varies from less than a minute for the fatty acids vaporized on the top tray to about 30 minutes for the residue leaving the bottom of the still. Most of the fatty acids are held at distillation temperatures for 10 to 15 minutes.

This is a considerable improvement over the direct fired pot type still, in which distillation temperatures reach 500° to 550° F., surface temperatures approach

[1]Presented at 25th fall meeting, American Oil Chemists' Society, Chicago, Ill., Oct. 8-11, 1951.

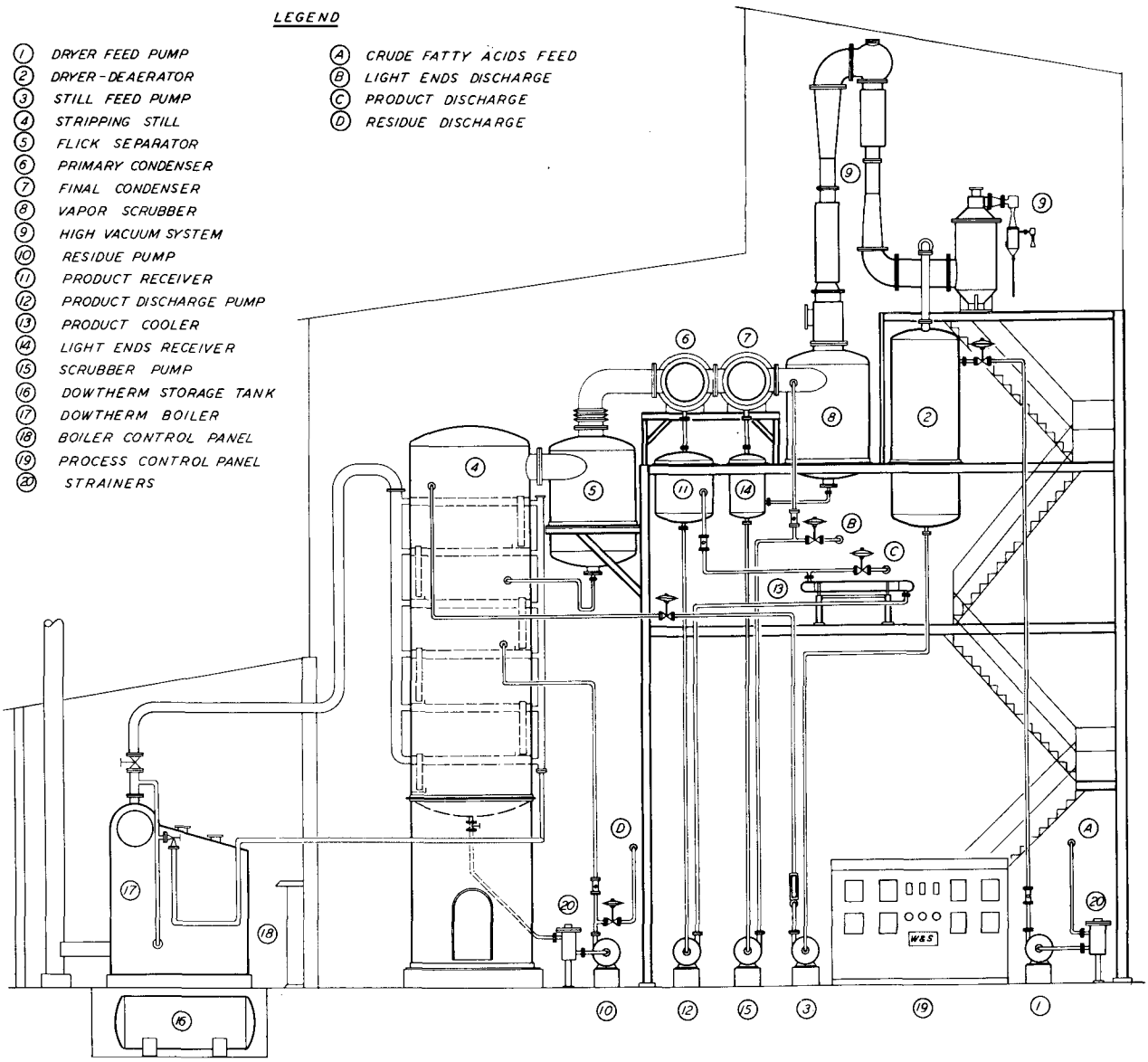

FIG. 1. General arrangement of equipment.

that of the furnace, and exposure time for some of the fatty acids extends from 24 to 48 hours (3). It is also an improvement over the flash type still, in which it is necessary to preheat the crude fatty acid feed to 575° F. to provide heat for subsequent vaporization upon entering the still (2), and the recycling usually required increases the exposure time of the fatty acids to elevated temperatures. Previous applications of the bubble tray column to fatty acid distillation have also employed high temperature preheating and consequently have the same disadvantage as the flash type still in this respect. Of course it is possible to transfer heat by means of pipe coils on the bubble trays (1), but this arrangement may seriously interfere with tray performance, cleaning, and maintenance.

Entrainment Separation

The fatty acids vaporized in the stripping still carry entrained crude fatty acids which, if not removed from the vapor stream, will be collected with, and contaminate, the distilled product. In the case of conventional pot still equipment an attempt is made to handle this problem by providing a disengaging space of sufficient cross sectional area to reduce the vapor velocity and of sufficient height to allow some of the entrained particles to drop out of the vapor stream; however this inefficient method is not directly applicable to high vacuum operations because the high specific volume of the vapors would require an uneconomically large disengaging space. The requirements of compact economical equipment, efficient entrainment removal, and low pressure drop through the equipment are successfully met by the Flick centrifugal entrainment separator, Item 5. This equipment is designed to separate the entrained particles from the vapor stream by centrifugal force and to collect and remove the material thus separated by means of stationary baffles. The fatty acid vapors leaving the separator usually contain less than 0.03%, by weight, of entrainment. Because of the ample vapor passages in the separator the pressure drop usually is less than 1 mm. Hg.

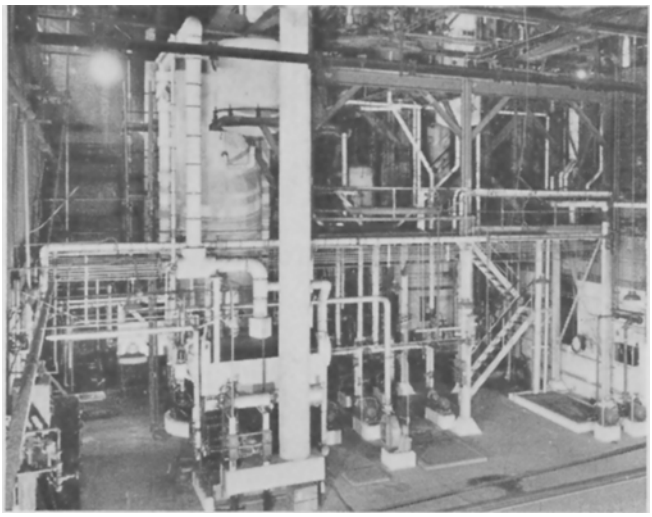

FIG. 2. General plant view.

Condensing

The purified fatty acid vapors from the entrainment separator are condensed in surface type condensers, Items 6 and 7. For economical usage it is desirable to employ the lowest temperature cooling water readily available. However, fatty acid mixtures that solidify at temperatures of 110° to 130° F. are frequently encountered, and careful condenser design and temperature control are required to employ cooling water temperatures significantly below the solidification point of the fatty acid condensate. For economical usage of the vacuum equipment it is desirable to minimize the pressure drop through the condensers. Both of these objectives are achieved through the use of two tube and shell type condensers illustrated in Figure 4. These condensers have ample vapor passages and are connected in series, with an individual automatic temperature controller on each condenser. Cooling water temperatures as low as 70° F. may be used directly without tempering, and the pressure drop is less than 1 mm. Hg per condenser.

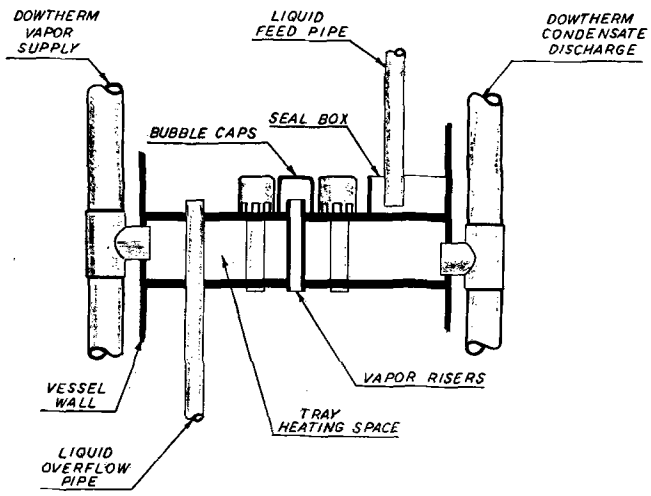

FIG. 3. Dowtherm heated bubble tray.

For comparison, the condensing equipment used on pot type stills of conventional design, many of which are in operation today, consists of tube bundles suspended in tanks to which the flow of water must be manually regulated, by means of an external circulating and tempering system, to maintain an inlet temperature of 100° to 130° F.; and, because of inefficient design, temperature control throughout these condensers is so poor that solidification of the fatty acid condensate will result if lower temperatures are used (3).

In addition to the improvements afforded by the foregoing basic features of the condensing system, special provisions may be made for the separation of the most volatile components from the distillate, thereby improving the odor and stability of certain stocks. Usually the amount of the fraction thus separated is in the range of 5% to 15% of the total distillate. In the case of smaller plants of under 2,500 pounds per hour capacity, the function of the second condenser may be combined with the vapor scrubber; and it is not usual to provide for the separation of a fraction of the distillate because of the small quantity involved.

Cooling

To avoid oxidation it is necessary to cool the condensed fatty acids before discharge to atmospheric storage tanks. By cooling continuously in counterflow tube and shell type equipment, Item 13, under automatic temperature control, cooling water temperatures as low as 70° F. may be used directly without tempering. Usual stocks may be discharged at a temperature of 150° F., which is sufficiently low to avoid darkening, but lower temperatures are practical for more sensitive stocks. For comparison, the cooling system used on conventional pot type stills consists of water tanks in which the condensate receivers

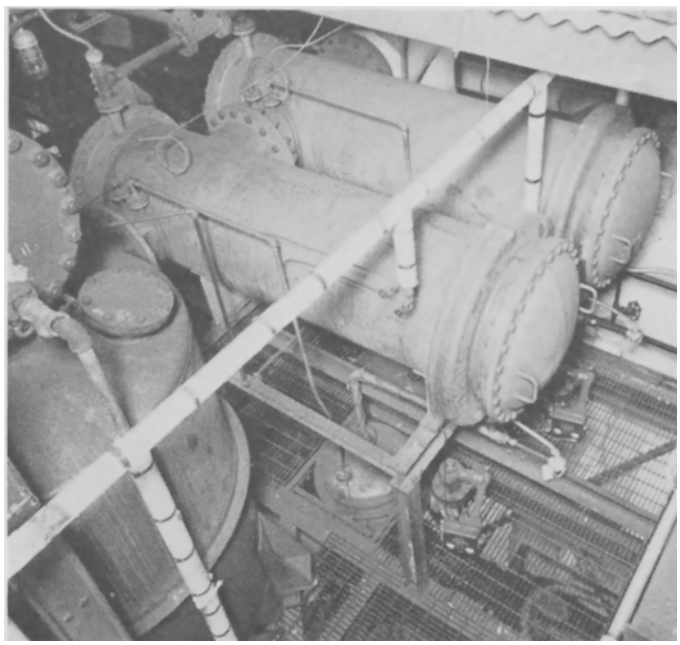

FIG. 4. Fatty acid condensers.

are mounted, and, due to inefficient distribution, the water temperature must be maintained above the melting point of the fatty acid condensate.

Exhaust Vapor Scrubbing

The vapors leaving the final condenser consist predominantly of stripping steam and non-condensable gases. However, some of the fatty acids condense in the form of a fog which is carried out of the condenser with the exhaust vapors.

If this entrainment is not removed from the vapor stream, it will pass out of the system and collect as a heavy clabber in the vacuum system hotwell. Recovery of the fatty acids from this hotwell clabber is laborious and inefficient. Furthermore it is necessary to operate the vacuum system condensers at 115° F. to 140° F. to avoid blockage by solidified fatty acids. Therefore even if the nuisance of hotwell clabber and the losses of fatty acids were tolerable, a high vacuum system operated in this temperature range would be very uneconomical.

Obviously there is a need for the removal of entrainment from the exhaust vapors; but the problem is complicated by the small size of the entrained particles which are not efficiently removed by conventional separators. A real improvement in this respect is the wet scrubber, Item 8 and Figure 5, in which the exhaust vapors are passed through a coarse spray of fatty acids, coalescing the fog type entrainment to drop size particles which may be removed efficiently by a Flick centrifugal entrainment separator. Here, as in other parts of the vapor system, low pressure drop is essential and usually is less than 1 mm. Hg. In the case of smaller plants it is practical to circulate sufficient fatty acids through the scrubber to permit its operation as a direct contact type final condenser in addition to its entrainment separation function.

FIG. 5. Vapor scrubber and vacuum system.

Residue Handling

The nonvolatile materials present in the crude fatty acid feed stock appear in the bottom of the still as residue, which is automatically pumped out to storage. The unsplit fat is the most important component in the residue and may amount to 50% of its weight. The residue also usually contains 15% to 25% free fatty acids; and, although it is possible to strip the residue down to less than 5% free fatty acids, such vigorous stripping is accompanied by an increase in the color and the unsaponifiable content of the distilled product. Because of the mild operating conditions and short residence time within the still, polymerization is minimized, and the residue is quite fluid compared to that obtained in direct fired pot still equipment.

The general practice is to store the residue in a tank equipped with steam coils until a sufficient quantity has accumulated to charge the Twitchell tank or batch autoclave in which splitting is conducted according to the usual procedures. The split residue may be distilled in a direct fired pot still if such equipment is available. This operation is conducted to produce a distillate containing not over 10% unsaponifiable material and a solid tar or pitch. The distillate may be marketed as a low grade product or blended with crude fatty acids and fed to the continuous still for the production of high grade distilled fatty acids. The tar may be marketed if conditions are favorable or burned under the plant steam boilers.

Except for large capacity plants or unusual economic conditions the installation of special tar still equipment is not justifiable. The split residue, either alone or preferably in combination with lowest quality crude fatty acids, may be rerun in the continuous still. The distillate so obtained corresponds to a "single-distilled" grade of fatty acids and is marketed as such or redistilled. The second residue may be burned under the plant steam boiler or otherwise disposed.

Instrumentation

The uniformly high quality of the products and the high operating efficiencies characteristic of the continuous process are dependent upon the automatic control of all the critical process variables. Of these the distillation temperature is the most important. With the feed temperature and rate controlled at constant values the distillation temperature is dependent upon the Dowtherm temperature. A proportioning type burner with a 5 to 1 turndown ratio is most satisfactory for firing the Dowtherm boiler, and the temperature may be regulated with a simple high-low floating controller on the smaller units or a more elaborate wide band proportional controller with automatic reset on the larger units. The feed temperature is easily maintained with a narrow band proportional controller regulating the steam supply to the dryer and deaerator. The feed rate is also easy to control because of the constant head pumping conditions.

The fatty acid condenser temperatures are difficult to control, probably because of the low heat transfer coefficients, and wide band proportional controllers with automatic reset are required. After the fatty acids have been condensed, however, higher heat transfer rates are attainable, and the fatty acid cooler temperature is easily controlled with a narrow band proportional controller.

The dryer and deaerator feed, product discharge, and residue discharge are regulated by liquid level controllers mounted in the bottom of the dryer and deaerator, product receiver, and still. In these ap-

plications smooth flow rates are much more important than accurate liquid level, consequently wide proportional band controllers are used.

Key temperatures not recorded elsewhere are recorded on a strip chart as a check on plant operations. An absolute pressure recorder for the vacuum system is also useful for this purpose. Manual control of the vacuum system barometric condensers usually is satisfactory since adjustment is required only to compensate for changes in the water supply temperature. However automatic control of the pressure of the steam supply for the ejectors is usually necessary because plant steam pressure seldom is sufficiently constant for this purpose.

With the process under automatic instrument control the operator may be lulled into a false sense of security; therefore it is desirable to provide an alarm system to warn him of any serious departure from normal operating conditions and a signal light system to direct his attention to the source of the difficulty. Such a system may include excessive Dowtherm pressure, flame failure of the burners in the Dowtherm boiler, abnormal liquid levels, flow stoppages, excessive temperatures, and loss of vacuum.

Grouping the instruments and controls on a central panel, as illustrated in Figure 6, contributes to the efficiency of the plant operation.

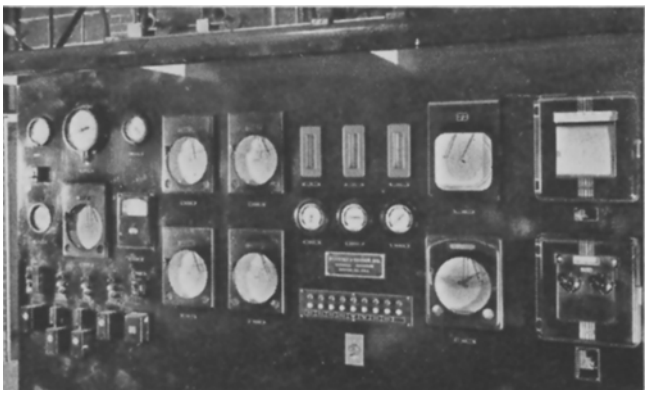

FIG. 6. Central instrument and control panel.

Materials of Construction

The surfaces of all equipment, piping, pumps, and instruments contacted by the fatty acids are constructed of stainless steel to assure freedom from metallic contamination in the products and to provide a satisfactory service life for the plant. A.I.S.I. type 316 stainless steel is the most generally applicable alloy, and this analysis is available in both the wrought form for welded fabrication and the cast form for valve and pump parts. Except for relatively thin sections however, the intergranular precipitation of carbides at welding temperatures destroys the corrosion resistant properties of this alloy. Since the equipment involved is quite complex, a carbide resolubilization heat treatment after fabrication is not practicable. At present the best answer to this problem seems to be the use of a special extra low carbon grade of this alloy so that the small amount of carbides precipitated during welding do not significantly affect the corrosion resistance.

Stainless steel does not satisfactorily withstand the attack of the acids of crude tall oil at elevated temperatures; and, if distillation of this material is contemplated, the equipment operated at high temperature should be constructed of "Inconel." Both stainless steel and "Inconel" are attacked at high temperatures by mineral acids or their organic compounds in the crude fatty acids even though present only in trace amounts, and careful testing of all feed stocks is recommended. To minimize corrosion from this source a short boil with dilute soda ash usually is sufficient for well settled stocks.

Operating Requirements

The operating requirements for the improved continuous process listed in Table I (A) are approximately correct for plants in the capacity range of 2,500 to 5,000 pounds per hour; and the requirements for smaller plants are somewhat higher per unit of output. The operating requirements for conventional pot still equipment are listed in Table I (B), based upon double-distilled product for comparison. The operating economies of the continuous process are obvious, and the savings in building space are equally important. In one instance a 4,000 pounds per hour continuous plant was installed in approximately the same space previously occupied by a single pot still unit capable of an average output of only 500 pounds per hour of double distilled product.

TABLE I

Operating Requirements for 1,000 lbs. of Distilled Fatty Acids

	(A) Improved continuous process	(B) Conventional pot stills
Steam, @ 100 p.s.i.g. pressure	475 pounds	1,200 pounds
Water,		
@ 72° F. for barometric condensers	4,350 gallons	9,000 gallons
@ 85° F. for surface condensers	3,000 gallons	9,000 gallons
Fuel oil, #6 @ 150,000 B.t.u./gallon	3½ gallons	15 gallons
Electrical power, A.C.	2⅔ kw. hrs.	.....
Compressed air, @ 40 p.s.i.g. pressure	25 cu. ft.	.....
Labor, semi-skilled	¼ manhours	3 manhours

In the operation of the continuous process, steam consumption is slightly less at higher pressures. Surface condenser water consumption is not appreciably affected by variations in temperature over the usual range of 75° to 95° F.; however increasing the barometric condenser water temperature significantly increases the water consumption and also may increase the steam consumption. It is usually economical, therefore, to install a cooling tower for the barometric condenser water if the supply temperature is expected to exceed 85° or 90° F.

The Dowtherm boiler may be arranged for firing with most grades of fuel oil or gas. If justified by economic conditions, electrical heating of the Dowtherm boiler is also possible. Firing with solid fuels is not practical however because control would be inadequate for the process.

Operating Results

Material balances and yields are given in Tables II through VIII for commercial operation with stocks of both animal and vegetable origin. Product qual-

TABLE II

Distillation of Tallow Fatty Acids

	Feed		Distillate		Residue		Remarks
Free fatty acids, MW 272	945#	94.5%	925#	99.5%	14#	20%	Yield, as FFA in distillate = 98% of FFA in feed.
Unsaponifiable material	7#	0.7%	5#	0.5%	14#	20%	Yield, including rerun of distilled split residue = 960# distillate + 40# tar.
Unsplit stock	48#	4.8%	0#	0.0%	42#	60%	Color of distillate = 1R/5Y to 2R/15Y (1" Lovibond).
Total	1,000#	100.0%	930#	100.0%	70#	100%	

TABLE III

Distillation of Hydrogenated Tallow Fatty Acids

	Feed		Distillate		Residue		Remarks
Free fatty acids, MW 275	965#	96.5%	917#	99.7%	28#	35%	Yield, as FFA in distillate = 95% of FFA in feed.
Unsaponifiable material	9#	0.9%	3#	0.3%	28#	35%	Yield, including rerun of distilled split residue = 955# distillate + 45# tar.
Unsplit stock	26#	2.6%	0#	0.0%	24#	30%	Color of distillate = 1R/5Y to 2R/15Y (5¼" Lovibond).
Total	1,000#	100.0%	920#	100.0%	80#	100%	

TABLE IV

Distillation of Red Oil

	Feed		Distillate		Residue		Remarks
Free Fatty Acids, MW 283	980#	98.0%	970#	99.5%	10#	40%	Yield, as FFA in distillate = 99% of FFA in feed.
Unsaponifiable material	20#	2.0%	5#	0.5%	15#	60%	Yield, including rerun of residue = 985# distillate + 15# tar.
Total	1,000#	100.0%	975#	100.0%	25#	100%	Color of feed = 7R/40Y (1" Lovibond) Color of distillate = 1R/8Y (5¼" Lovibond)

TABLE V

Distillation of Coconut Fatty Acids

	Feed		Distillate		Residue		Remarks
Free fatty acids, MW 209	965#	96.5%	937#	99.7%	3#	5%	Yield, as FFA in distillate = 97% of FFA in feed.
Unsaponifiable material	5#	0.5%	3#	0.3%	30#	50%	Yield, including rerun of distilled split residue = 955# distillate + 45# tar.
Unsplit stock	30#	3.0%	0#	0.0%	27#	45%	Color of distillate = 1R/8Y to 3R/12Y (5¼" Lovibond)
Total	1,000#	100.0%	940#	100.0%	60#	100%	

TABLE VI

Distillation of Soyabean Fatty Acids From Split Soapstock

	Feed		Distillate		Residue		Remarks
Free fatty acids, MW 280	910#	91.0%	843#	99.2%	45#	30%	Yield, as FFA in distillate = 93% of FFA in feed.
Unsaponifiable material	40#	4.0%	7#	0.8%	60#	40%	Yield, including rerun of distilled split residue = 915# distillate + 85# tar.
Unsplit stock	50#	5.0%	0#	0.0%	45#	30%	Color of distillate = 2 to 4; 4 to 7 after 500 heat test (Gardner 1933).
Total	1,000#	100.0%	850#	100.0%	150#	100%	Iodine value of distillate = 125 to 135.

TABLE VII

Distillation of Cottonseed Fatty Acids From Split Soapstock

	Feed		Distillate		Residue		Remarks
Free fatty acids, MW 277	910#	91.0%	690#	98.5%	150#	50%	
Unsaponifiable material	50#	5.0%	10#	1.5%	114#	38%	Yield, single distilled, as FFA in distillate = 76% of FFA in feed.
Unsplit stock	40#	4.0%	0#	0.0%	36#	12%	Color, single distilled = 5R/30Y (5¼" Lovibond).
Total	1,000#	100.0%	700#	100.0%	300#	100%	

TABLE VIII

Redistillation of Single Distilled Cottonseed Fatty Acids

	Feed		Distillate		Residue		Remarks
Free fatty acids, MW 277	690#	98.5%	660#	99.2%	21#	60%	Yield, double distilled, as FFA in distillate = 73% of FFA in crude.
Unsaponifiable material	10#	1.5%	5#	0.8%	14#	40%	Color, double distilled = 1R/5Y (5¼" Lovibond).
Total	700#	100.0%	665#	100.0%	35#	100%	Over-all yield, double distilled, including rerun of distilled split residue = 800# distillate + 200# tar.

ity and residue composition also are given since a figure for distillate yield alone is meaningless because, by severe stripping in some processes, it is possible to drive almost all of the feed overhead to produce a poorly colored distillate high in unsaponifiables and a highly polymerized unrecoverable residue, thereby showing an excellent distillate yield although the products are next to worthless. The over-all yields given include recovery of free and combined fatty acids from the residue as previously described.

These data are generalized to the extent that they represent average conditions of a number of runs on similar stocks, and they are not instantaneous values obtained under optimum conditions of operation. It should be recognized however that even stocks of the same general type are widely variable in composition, and it is impossible to make completely general statements regarding their behavior. For example, it should not be assumed that every lot of fatty acids of the tallow type would behave exactly as indicated in Table II.

As would be expected, it is necessary to take more residue when distilling low split stocks or stocks high in unsaponifiables if the quality of the product is to be maintained. And, since residue recovery is not as efficient as straight distillation, the over-all yields are lower. Higher distillate yields are possible with any stock if the attendant sacrifice of product quality with respect to color and unsaponifiables is acceptable.

Losses due to polymerization are low as indicated by the exceptionally fluid residues obtained and by the high iodine value of the distilled soyabean fatty acids. Losses in straight distillation usually range from 1% to 2% of the free fatty acids in the feed and are represented by a gain in unsaponifiables: however it is important to observe that the unsaponifiables are concentrated in the residue and, for the most part, kept out of the distillate.

Crude fatty acids from acidulated cottonseed soap stock are one of the most difficult materials confronting the distiller. The combination of solvent extraction of the cottonseed and continuous refining of the oil are primarily responsible for the presence of large amounts of extremely persistent coloring matter in the soap stock. Tables VII and VIII illustrate the behavior of the more difficult of such stocks which require a double distillation to produce a high grade product. The composition of these crudes however is widely variable, and many can be handled satisfactorily with a single distillation. It should be noted that the crudes were prepared by conventional acidulation of the soap stocks followed by high pressure hydrolysis, and no special treatments were used at any time.

In the distillation of animal fatty acids with conventional pot still equipment, a 90% yield of double distilled product with a color of 4 Red/35 Yellow (1″ Lovibond) may be obtained, depending primarily upon the skill and technique of the operators. With the same stock distilled by the improved continuous process described, a 96% yield of product with a color of 1 Red/5 Yellow (1″ Lovibond) may be obtained. In the distillation of vegetable fatty acids it is difficult to make a quantitative comparison of processes because with conventional pot still equipment the results are so unsatisfactory with respect to yields and product quality that it is not usually attempted.

Summary

In the simple distillation of fatty acids, improved results with respect to product quality, uniformity, and yields are obtained with reduced plant operating and space requirements because of the following factors:

1. Continuous operation under automatic instrument control.
2. Deaerating the feed stock before heating.
3. Vacuum drying the crude fatty acids at low temperatures.
4. Indirectly heating the fatty acids to distillation temperatures by means of condensing Dowtherm vapors within the bubble trays of a stripping still. With this unique heating system the fatty acids are not exposed to high temperatures until they are under the high vacuum within the still itself, thereby eliminating the preheating of feed stocks far above distillation temperatures before they are under high vacuum.
5. Low hold-up and minimum heat transfer surface temperature in the stripping still.
6. Removal of entrainment from the distillate vapor stream before condensing the product.
7. Efficient product condensing system and cooling the product before discharge to the atmosphere.
8. Wet scrubbing of the exhaust vapor stream after condensing the product.
9. Residues which are easily handled for the recovery of free and combined fatty acids.
10. Freedom from metallic contamination, because of all stainless steel construction wherever contacted by the fatty acids.

Acknowledgment

The cooperation of Frank Fedowitz, chemical engineer, and the other members of the technical staff of A. Gross and Company, Newark, New Jersey, in obtaining the foregoing plant operating data is sincerely appreciated.

REFERENCES

1. Barnebey, H. L., J. Am. Oil Chem. Soc., *26*, 493-502 (1949).
2. Mills, V., Fatty Acid Distillation, U. S. Patent 2,274,802 (1942).
3. Wurster, O. H., "Operation of Fatty Acid Distillation Plants," Chem. and Met. Eng., Vol. 25, No. 14 (1921).

[Received October 29, 1951]