Perfumer & Flavorist

A Continuous Steam Stripping Process for the Distillation of Essential Oils

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Natural essential oils are still believed by many to be of strategic importance to the flavor and fragrance industry. Despite continuing efforts and sophistication in aroma chemical manufacturing, essential oils remain absolutely necessary for fragrances, from the most sophisticated to most cosmetic, and even many household products.

Some 100 essential oils are commercially extracted for sale to the Flavor and Fragrance industry. Only a few oils are produced in the United States on a very large scale, such as peppermint and spearmint oils, citrus oils, pine oils and cedarwood oils. Some herb and spice oils are distilled only sporadically as the market demands. The majority of essential oils are still imported from foreign countries, mostly those with tropical and subtropical climates.

Typically, the producing countries have been using conventional and sometimes rudimentary methods of distillation. Apparently the essential oil industry has not done fundamental research on improved distillation technology.

A worldwide search of scientific literature conducted in 1981 revealed no significant new developments in the general methodology of essential oil distillation in the previous 40 years.

Continuous distillation of various aromatic plants has apparently been undertaken in the Soviet Union with some success, but no detailed scientific literature could be obtained. More recently some private and proprietary research on the

0272-2666/91/0002-0101$4.00/00—© 1991 Allured Publishing Corp.

Perfumer & Flavorist (ISSN 0272-2666)
An Allured Publication
Volume 16, Number 2
MARCH/APRIL 1991

subject is reported in France by Biolandes Techologies.

Historical Background

The application of an entirely new concept in essential oil distillation was first attempted in 1981 by the Boucard Group, long-time distillers of essential oils, and R. W. Serth, a chemical engineering professor at Texas A & I University.

The specific objective of the research underlying this new process design was to develop and test a method by which the essential oil of cedar (*Juniperus Mexicana*), containing diterpenes, sesquiterpene alcohols, fatty acids and rosins could be separated from heavily lignified plant tissues by continuous partial pressure distillation, using steam or heated inert gases, a mass transfer operation known as desorption or stripping.

After a year and a half of fruitful laboratory, and bench-scale testing at Texas A & I University, Texarome, Inc. was formed in 1982 by a group of investors, and a 4 tons/day pilot plant was set up in the Texas Hill Country near Kerrville to extract Cedarwood oil from the native Texas Cedar.

In 1984 the project was moved to Leakey, Texas, and the pilot plant was scaled up to a 12 tons/day commercial size plant, which has been operated around the clock to this date, with its production of cedarwood oil successfully marketed. Until now the process has been well guarded from the competition as a proprietary means by which to extract essential oils from aromatic plants on a continuous basis.

Due to the applicability of the process in areas of much greater importance and volume than essential oils, Texarome is also marketing the process for addressing the urgent problem of toxic solid waste clean up (soils contaminated with toxic volatiles), a separation task very similar to the production of essential oils.

Texarome's Continuous Steam Stripping Process

Superheated steam as a carrier gas can serve the purpose of vaporizing the high boiling volatile liquid present in the solids being conveyed, by virtue of the partial pressure effect. An elaborate and proprietary way of running the pipes within the conveying system allows a true countercurrent flow of the gas phase and the solid phase, to make the efficient mass transfer separation task possible.

Concept of the Novel Process—The concept came from the observation that when conveying very finely ground and sometimes overheated distillation material (80°C) with hot compressed air (65°C) by way of a pneumatic conveying system, much of the oil escaped at the top of the cyclonic separator (collector) along with the hot air.

The deduction was made that, were one to operate a pneumatic conveying system, using steam as a conveying gas (carrier gas) instead of air, the material could be exhausted of its oil content while being conveyed.

Indeed, while the word "pneumatic" refers to air as a carrier gas, the air in this case is being substituted with superheated steam which is, like air, a dry gas quite suitable for "pneumatic" conveying.

Table I. Values of Operating Parameters

Parameter	Value
Maximum throughput (t/d)	12.0
Steam to wood ratio (kg/kg)	1.0
Temperature range (°C)	120-220
Pressure range (bar)	0.07-0.2
Mean residence time (sec)	25
Average particle size (mm)	0.25

Principle of Operation—Since the object of the apparatus is not to separate the mixture of compounds in the liquid phase into fractions such as in a distillation, but rather to vaporize all of the liquid mixture, the task is considerably simplified. Hence, the operation is reduced to a simple *desorption* or *stripping* of the entire liquid phase from the inert carrier solid by partial pressure distillation.

Therefore, if the physical properties for the heaviest desired volatile are known, and the design criteria and steam conditions exist in which the heaviest volatile cannot exist in the liquid form, and is vaporized by the partial pressure phenomenon before leaving the continuous apparatus with the carrier solid, then the exiting solid will be free of that particular heavy volatile and all the lighter ones as well. This is not to say that one may not choose to leave some of the heavier and undesirable compounds in the solid, such as natural wood rosins, fatty acids, etc., by controlling the operating variables of the process, primarily the temperature, and steam/product contact time.

The Process

The main process variables and the values of the operating parameters in Texarome's continuous distillation plant are discussed below.

Table II. Texas Cedarwood Oil Analysis

Component	Sample #1	Sample #2	Sample #3
α-cedrene	3.7	17.5	15.5
β-cedrene	-	3.7	4.1
thujopsene	36.1	33.5	33.6
cedrol	43.0	29.2	22.8
widdrol	6.0	4.6	2.6
Totals	**88.8**	**88.5**	**78.6**

Sample #1: Cedarwood oil crude from Texarome's continuous process.
Sample #2: Cedarwood oil from 24h batch process at atmospheric pressure.
Sample #3: Cedarwood oil from 6h batch process at ± 15 psig (1 bar).

Steam/solid ratio—The finally established ratio of 1:1 for steam versus solids to be processed, provides both sufficient carrier gas for the low pressure/dilute phase "pneumatic" transport of the solid through the continuous apparatus, as well as more than sufficient steam for the distillation during the very short period of transport. The most dramatic improvement of that technology over the previous ones, is that this ratio of 1:1 is one third the amount required for the distillation of cedarwood oil in conventional batch plants.

This, of course, has a direct effect on the capital cost of any distillery where, typically, the largest piece of capital equipment is the boiler. In instances where the boiler is fired with expensive fossil fuels, plant energy is usually the second highest cost item after raw material, so that a reduction of energy requirements by one third will have a significant incidence on the cost price of the finished product.

Temperature—The temperature of the superheated steam indicates the amount of sensible heat available to effectuate the distillation. It is important to note that only the sensible heat portion of the steam is available for the actual mass transfer separation process. Drawing on the steam's latent heat for heat transfer to the "cold" incoming solid would cause condensation of the carrier gas and the "pneumatic" transport of the solids through the system would stop.

In fact, the prevention of condensation at any point of the apparatus is one of the major problems that had to be overcome in developing this technology. The only limiting factor for the superheated temperature of the steam is the ignition point of the exiting biomass, which is approximately 230°C. Higher temperatures would require cooling or quenching of the exiting solid.

Contrary to popular belief, the high steam temperatures do not seem to affect adversely the composition and the odor characteristics of the oil, certainly not in the case of cedarwood oil. This is attributed to the complete absence of oxygen and the very short residence (distillation) time. Interestingly, the absence of "moisture" and air also seems to mitigate both corrosion and erosion problems normally anticipated in such system.

For instance, an entire apparatus built out of 16 gauge mild steel (1.5mm) has been operating around the clock for six years with only minor corrosion/erosion at elbows and tangential inlets. It is apparent that the high steam temperatures (dry steam) and the absence of "moisture" or "liquid water" prevents the leaching of organic acids from the aromatic biomass, which not only contribute to corrosion but also have a catalytic effect on the reduction of alcohols (especially tertiary alcohols) and the hydrolysis of esters. A typical example is the reduction (dehydration) of Cedrol to Cedrene in conventional batch stills, which obviously occurs to a much lesser degree in this system (Table II).

Pressure—One of the advantages of this process is that it operates efficiently at the very low pressures required for dilute phase pneumatic conveying, i.e., 1 to 5 psig (0.07-0.34 bar). Such low pressures simplify the inevitable sealing problems associated with the continuous feed of solids into pressurized systems. Nevertheless, special know how had to be developed and various experiments conducted to deal effectively with the sealing and condensation problems at the inlet and discharge of the continuous apparatus.

Residence time—Routing the pipes for a true counter-current flow and achieving sufficient residence time are the key elements of this process. Standard pneumatic conveying velocities are in the order of 5,000 ft/min (1524m/min). Hence some half a mile (0.8km) of pipe would be needed to obtain a 30-second residence time while suffering considerable pressure drop. In this system, the residence time is provided for by the unique geometric configuration of the apparatus. Laboratory tests backed up by pilot tests for cedar indicate that 25 to 30 seconds is the residence time required for this separation task, given a particle size of 35 mesh and under (average 0.25mm).

Particle Size—Theoretically, the finer and the more non-porous the solid, the less residence time and the less stages are required to achieve the same yield. Given sufficient temperatures, these two variables, particle size and porosity, have been found to be the only main constraints in the way of an instant flash separation of the liquid phase from the solid phase. For instance, excellent yields in excess of 90% of the theoretical maximum can be obtained at 200°C with a particle size gradient of 35 mesh and under (aver. 0.25mm), for heavily lignified biomass in which the essential oil droplets are literally encapsulated by the cell walls and the lignin deposits, such as in the case of Texas cedarwood.

The Processing at Texarome

Texarome's continuous processing plan has a nominal capacity of twelve metric tons per day (t/d). Cedar logs are fed manually (one-man operation) into the first stage, a chipper, which has a nominal capacity of four tons per hour (t/h) and is driven by a 75 kw engine. The chipper feeds directly into the second stage, a 50 kw hammermill which transports the wood chips pneumatically to a storage bin. The chipper and mill have a much greater capacity than required, because it is impractical to scale down a chipper which must accept logs ranging in size up to 2.5 meters in length and 30 cm in diameter. For that reason, the primary grinding equipment operates only three to four hours per day, allowing for some material handling and work scheduling flexibility.

From the storage bin, the wood chips are delivered at a controlled rate of 500 kg/h into a third stage, a 40 kw pulverizer where the chips are reduced in size to 35 mesh and under (average 0.25mm). The pulverized wood is pneumatically transported to the inlet of the second stage continuous distillation unit. The finely ground material enters the distillation unit through a plug feeder and a rotary valve. In the distillation unit, the wood is contacted with superheated steam from the boiler. The steam and the suspended wood particles flow through the distillation unit in a co-current flow pattern. However, other flow patterns, e.g., counter-current or cross-flow are part of the wood's itinerary to the exit point, after a total residence time of 25 to 30 seconds. The contacting occurs in discrete stages with interstage heat addition. In each stage, the wood is dispersed in the steam, so that intimate contact between the gas and the solids is attained. The entire distillation apparatus is enclosed in an insulted plenum (2.5m tall, 1.2m wide and 9m long), through which passes the boiler stack gas with a temperature reading of 300-400°C, so as to prevent any form of condensation inside the distillation unit.

Before exiting the continuous distillation unit, the steam and wood suspension goes through a cyclonic vessel in order to separate the steam/oil vapors from the exhausted wood. The steam/oil vapors inevitably entrain a dilute stream of very fine dust and must be channeled through a special five micron steam filter before entering a water cooled shell and tube heat exchanger for condensation of the distillate. The condensate is sent to a decanter where the oil separates from the water by virtue of its lower specific gravity. Because the oil flow rate is quite small, the oil is drawn from the decanter periodically (rather than continuously) and transferred to $0.2m^3$ drums for shipment. The condensed steam which contains traces of dissolved oil, is drawn continuously from the bottom of the decanter and chemically treated for re-use as boiler feedwater. The plant's water consumption and effluents are, therefore, minimal.

The spent cedar exits the distillation unit through a rotary valve, and is divided into a waste stream and a fuel stream. The latter is pneumatically transported to a specially designed wood-burning furnace which supplies all the energy for steam generation. The spent cedar constitutes an excellent fuel, as any other spent aromatic biomass would, due to its small size and low (approximately 4%) moisture content. These characteristics permit the use of an inexpensive suspension-type furnace as opposed to a dutch oven or fluidized bed furnace required to burn larger wood chips and other coarse, wet biomass. The flow rate of wood to the furnace is controlled automatically by a baffle, the position of which is dictated by the boiler pressure sensor. The boiler is a two-pass return tubular type, rated at 680kg/h, which provides steam for the continuous distillation process, as well as for other refining operations of the plant. The stream of waste wood is pneumatically carried to a dual chamber incinerator for disposal.

The operation of the plant is completely automated except for the chipper feeding operation and the removal of the product oil from the decanter.

Product Quality and Yield

The Texas cedarwood oil produced in Texarome's continuous distillation plant is a reddish, viscous liquid which crystallizes at room temperature and is referred to as crude oil. The crude oil is then redistilled, fractionated or blended by Texarome to produce the variety of grades required by the fragrance and aroma chemical industry. An analysis of the crude oil is presented in Table II. For comparison, analyses are also given for the oil of a commercial batch plant operating stills at atmospheric pressure and 24h cycles, and for the oil of a commercial batch plant operating stills at 15 psig (1.03 bar) and 6h cycles. After studying the analytical results, it is obvious that the composition of Texarome's oil most resembles nature's original composition of the oil as it must occur in the wood of the mature tree.

As evidenced by the high cedrol content and low cedrene content of Texarome's oil, it is apparent that the extremely short distillation time of 25 seconds has, despite the high steam temperatures, the most gentle and forgiving effect on the natural chemistry of this oil, as it probably would for any other oil. Since the dehydration of cedrol to cedrene is not known to be reversible under the conditions of any steam distillation process, conventional or continuous, it follows that the oil with the highest cedrol owes its cedrol content not to the oxidation of α-cedrene to cedrol, but rather to lesser rate of reduction of cedrol to α-cedrene and ß-cedrene, and must, therefore, be the one closest to the natural composition of the oil before extraction.

The average yield of oil from this continuous process is 2.8% by weight. This value is significantly higher than the average yield of 2% reported by most of the commercial plants processing Texas cedarwood. It must be noted that the higher yield is accompanied by a slight increase in the rosin content of the oil. This is believed by some to increase the fixative properties of the oil, other users request the removal of all wood rosins by vacuum distillation. The net yield of oil is still considerably higher than in the conventional batch process. Further increase in cedrol yield and organoleptic quality could be achieved by continuously stripping the water phase immediately downstream of the separator with the novel continuous liquid/liquid extraction method described recently by Fleisher.[2]

Cedar is not an aromatic material which requires solvent extraction after steam distillation. However, a number of very valuable fragrance and especially flavor materials fall in that category and are marketed as extracts and oleoresins. This apparatus is readily adaptable to a liquid/solid design using solvent pumps, rotary valves and hydroclone separators. Such a unit could be installed downstream of this gas/solid distillation unit and efficiently extract the oil and/or the dry, pulverized material exiting the distillation unit in a continuous manner as well. Again, a third gas/solid unit could strip the remaining solvent from the exiting solvent-wet material. Unlike batch systems, a continuous system such as Texarome's apparatus can be dramatically scaled up, but it can also be scaled down to a table top size machine while maintaining considerable daily throughputs for expensive, small volume raw materials.

Concluding Remarks

An entirely new process for distilling essential oil-bearing materials has been successfully demonstrated in a pilot plant and in a scaled up commercial plant for the on-going production of Texas cedarwood oil. It should be noted at this point, that the system was briefly tested with vetivert roots (vetiveria zizanoides) and eastern cedar (juniperus virginiana). As expected, the apparatus made no distinction between one pulverized aromatic material and the others. The operational results were excellent, although like Texarome's cedarwood oil, the composition of these oils showed a higher proportion of heavy, oxygenated sesquiterpene constituents than the commercial grades. In particular, it has been shown that a high yield of a superior-quality oil can be obtained under field conditions for some of the most difficult raw materials. Other features of the process which have been demonstrated in the existing plant include a significant economy of steam and a high degree of automation, with a correspondingly low manpower requirement; a high degree of reliability; ease of operation; minimal environmental impact; and uniformity of product quality resulting from steady-state operation at tightly controlled processing conditions.

Finally, this particular continuous distillation technology opens the door for the next generation of essential oil extraction equipment, which is envisioned as a "distilling combine harvester." Essential oil bearing row crops, such as the mints, lavender, coriander, lemongrass and others may one day be extracted by a combine, in which the waste heat and the purified carbon dioxide exhaust of the combine's engine will strip the oil from the crop being harvested. A compact cryogenic cooling system will recover the oil from the CO^2 gas stream (most of Texarome's laboratory research was conducted with various hot inert gases), while the spent biomass will be spread back in the field or brought in for cattle feed.

Acknowledgment: *The author is deeply indebted to the shareholders of Texarome Inc. who financed this research, particularly to Prof. Dr. R. W. Serth of Texas A & I University, College of Engineering, Kingsville, Texas, a great teacher and mentor; to Dr. K. E. Harwell for his edifying contribution to the understanding of cedarwood oil chemistry; and last but not least, to a fine crew of dedicated plant workers, who helped turn theory into working reality.*

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