PATCHOULI ALCOHOL ENRICHMENT FROM PATCHOULI OIL USING MOLECULAR DISTILLATION UNIT

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ABSTRACT

Molecular distillation in the vacuum pressure about 10^{-3} mbar, temperature in the range of 80 °C – 100 °C, and wiper rate between 60 – 80 rpm was used for the separation and purification of patchouli oil. Patchouli oil has a large composition of patchouli alcohol which is usually used as a fixative for perfumery, cosmetics and pharmaceuticals. A large composition of patchouli alcohol usually is in the residue due to the high boiling point of this compound than others. Purification of patchouli alcohol increased with wiper rate and temperature but decreased with the feed rate. The temperature of liquid entering to the unit, feed rate and the wiping element bears on the distillation cylinder due to the centrifugal force and rolls over it bringing the liquid on the distillation cylinder into an intensive complex movement were the important technological parameters that determined distillation operation. Pursuant to condition above produced purification of patchouli alcohol due to the patchouli oil about 73.37%.

***Keywords:* Patchouli oil, molecular distillation, patchouli alcohol, separation, purification.**

INTRODUCTION

Patchouli oil is one of several kinds of essential oils. Vegetable oils and essential oil are the most interesting products proposed for its purification using molecular distillation unit. Molecular distillation technology has many advantages due to its characteristic vapor pressure of each substance. The vapor pressures difference dictates how easily a complex compound can be separated into its constituent components (Joddy and Wuryaningsih, 2002). The patchouli oil could be woody, earthy, and sweet smell that is pungent and persistent. It is stimulate in small amounts and sedates when use more generously. The patchouli oil is anti-inflammatory and anti septic. The oil can be applied directly to relieve skin condition such as burns, cracked skin, allergies, acne, herpes and eczema. In aromatherapy it is used for tiredness, tension, dandruff and oily skin or scalp (Ketaren, 1985).

Molecular or short-path distillation is characterized by a short exposure of distilled liquid to elevated temperatures, high vacuum in the distillation space and a small distance between the evaporator and the condenser. The short residence time of the liquid on the evaporating cylinder is guaranteed by distributing the liquid in the form of a thin film of even consistency (Cvengros, 2000). Feed temperature influences on the efficiency of a molecular evaporator were explored by Cvengros *et al.* (1999). Cvengros *et al.* (2000) studied film wiping in the molecular evaporator. Heat and mass transfer in the evaporating film of molecular evaporator were presented by Lutisan *et al.* (2001). Wiped-Film Molecular or molecular distilla-tion, can be used where the distillate has high boiling point, then processed in high vacuum (to 0.001 torr) and a close proximity internal condenser are utilized in molecular distillation. This design is selected when a significant percentage of moderate or low boiling distillate, medium vacuum (to 0.1 torr) and a high surface area external condenser are utilized (Anonimous, 2004).

A fundamental process attribute of molecular distillation is ability of evaporate high molecular weight materials without heat degradation and to strip off solvent into very low residual levels. This is accomplished by (i) reducing system pressure, to a minimum of several microns, (ii) lowering the boiling temperature of the distillate, and (iii) creating a very thin film on the distillation unit wall. The thin liquid film is produced by slotted wiper a blade propels across heated surface in a few seconds. A high degree of film turbulence is also created, to minimize the temperature difference between the wall and the evaporating surface of the liquid. All of these factors are combined to allow operation at the lowest possible temperature, thus preserve product stability.

Since the molecules of all matter are in constant motion in varying degrees, depending upon the chemical composition of that matter and the temperature and pressure applied to it, molecules near the surface have tendency to escape into the surrounding atmosphere. As temperature increases and pressure decreases, this escaping tendencies usually increases and the substance evaporate. The force generated by these escaping molecules is referred to as the vapor pressure of that material at a particular temperature and pressure. It is the relative difference in vapor pressure of substance which dictates how easily a complex compound can be separated into its constituent compounds (Anonimous, 2004)

The aim of this study is to investigate the effect of different wiper peripheral speed, and heating mantle temperature on purification of patchouli oil. The analysis of product included a purity of patchouli oil, and physical-chemistry properties e.g. density, refractive index, solubility in ethanol, acid value, ester value, fatty acid and kruing contents, optical rotation, and viscosity.

EXPERIMENTAL PROCEDURE

Apparatus

The experimental assembly consisted of a two stage molecular distillation with pilot plant scale.

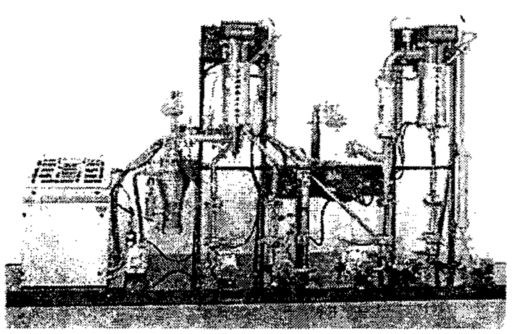

Figure 1. 6" two stage molecular still system.

The unit, as pictured, is completely self-contained (including full instrumentation) and ready for evaporation and/or molecular distillation operation by itself, or in combination with existing processses. Continuous or batch, single or two-stage modes, may be selected. The 2.36 sq. ft (0.22 sq. m) 316 stainless steel still bodies (glass or Hastelloy also available) allow feed rates from 2 to beyond 50 liters/hour, depending upon the application. Vacuum levels from 300 torr to better than 0.001 torr, and temperatures from ambient to beyond 400 degrees Celsius are possible. There are contain two side of heating mantle in one stage, thus we can adjust the temperature between up and down in one stage among the conditions. These could be affected the mass and heat transfer inside the column. The Pope Wiped-Film Still consists of a heated body into which a fluid system requiring some degree of separation is continuously fed. The fluid is spread into a thin film by a rotating wiper blade assembly driven at a predetermined speed. The film, while being forced into turbulent flow by the wiper blades, progresses down the inside body wall aided by gravity and the slots in the wiper blades.

It remains a primary means of separation for many applications, including:

- Solvent stripping
- Stripping monomer from polymer
- Stripping free fatty acids from fats and oils
- Distilling fats and oils
- Concentrating or distilling heat-sensitive pharmaceuticals, nutraceuticals and biomaterials
- Distillation of polymers
- Distillation of petroleum fractions
- Reactives from solid catalysts
- Concentration of fruit juices
- Isolation of aromatic compounds
- Deodorization of oils
- Removal of colors
- Separations of waxes or silicones
- Foods and flavors purifications

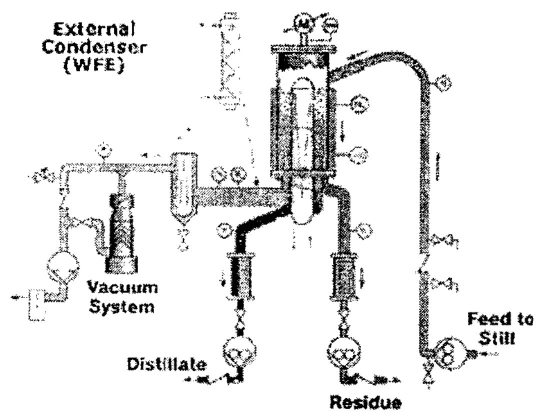

Figure 2. Basic Process Description for molecular system.

Feed liquid is admitted into still under vacuum, immediately spread into a very thin film and forced quickly down the evaporation surface. Heated walls (orange) and high vacuum (yellow) drive the more volatile components (distillate) to the closely positioned internal condenser as the less volatile components (residue) continue down the cylinder. The resulting fractions, thus separated, exit through individual discharge outlets. Depending on application, the desired product is either the distillate or the residue fraction.

Experimental

The effect of different wiper peripheral speed, and heating mantle temperature on purification of patchouli oil resulting a highly purity of product was specifically analyzed, therefore these three variable was assumed as dependent variable. Different wiper speed was carried out at 60, 70 and 80 rpm. While another dependent variable is heating mantle temperature was carried out at 80, 90 and 100°C. Independent variable in this study was carried out with the following:

1. Vacuum pressure : 10^{-3} mtorr.
2. Feed temperature : 50°C.
3. Feed rate : 1000 mL/h.

RESULT AND DISCUSSION

The wiped-film still consists of a heated body into which a fluid system requiring some degree of separation was continuously fed. The fluid was spread into this film by a rotating wiper blade assembly driven at predetermined speed. The film, while being forced into turbulent flow by the wiper blades, progresses down the inside body wall aided by gravity and the slots in the wiper blades.

During the course of flow through the heater system, some degree of evaporation took place depending upon the characteristics of the feed material and the inside wall temperature, in addition to the system pressure. The non-evaporated fluid forming the bottom product flowed out of the system continuously while the vapor was condensed either inside or outside the system. Thin film are created for a variety of reasons, (1) turbulence created by a rapidly moving wiper or controlled clearance blade greatly assists in heat transmission, thereby lowering the temperature required on the inside evaporator wall for a given system pressure, (2) a maximum surface area per unit volume of flow was generated facilitating rapid evaporation, (3) the liquid exposure time to the elevated wall temperature was controlled within second or less. This minimize product degradation of heat sensitive materials by controlling the wiper assembly speed, (4) the units slotted wiper blades promoted plug flow with little back mixing. These minimize dwell time distribution, ensuring that material flowing through the system has a uniform exposure to produce conditions.

Table 1shows the result of physical properties analysis of crude patchouli oil raw material. There was an indication that patchouli oil did not consist of fat in the form of fatty acids or kruing oil content, so the patchouli oil used in this study was not contaminated by other compounds. For this reason, process dissociation was directly conducted without doing purification early from crude of patchouli oil as raw material. The result also showed that optical rotation was a negative number, this mean that patchouli oil had a levo-rotation optic, specification of its deals with patchouli oil specification in the market, especially used for pharmaceutical and cosmetics grade. Patchouli oil had a rich component inside it which was patchouli alcohol. The content of patchouli alcohol in the crude patchouli oil raw material for this study was 19.37% analyzed by GCMS method. This raw material came from Bengkulu. It was seen that the concentration of patchouli alcohol was very low hence this product will have low value if sold. Therefore for this requiring look for a technology which could produce a highly concentrated of patchouli alcohol.

The experiment showed that separation of product was divided into two shares. One is the product rich of volatile component or has a low boiling point called Distillate, and another is the product which has a poor volatile component or highly boiling point called Residue. Figure 1 shows yield of product in both distillate and residue from this experiment, this determining that the product became two shares, distillate and residue which has a % yield for each share.

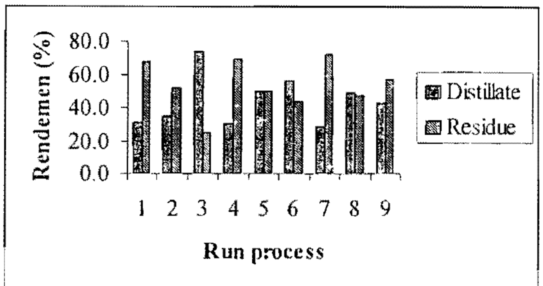

Fig. 1. Yield of both distillate and residue products.

The fundamental process attribute of molecular distillation was the ability to evaporate fairly high molecular weight materials without heat degradation. This was accomplished by reducing system pressure, to a minimum of several microns, thereby lowering the boiling temperature of the distillate, and by creating a very thin film on the evaporator wall. The thin liquid film produced by slotted wiper blades propels the liquid across the heated surface in a few second, minimizing exposu-res to the elevated temperature. A high degree of film turbulence was also created, minimizing the temperature differences between the wall and the evaporating surface of the liquid. All of these factors combine to allow operation at the lowest possible temperature, thus preserving product stability.

In order to sustain the lowest operating temperature, it was necessary to provide uniform heat flow and effective temperature control of the heated surface.

Table 1. Physical properties analysis of crude patchouli oil raw material.

No.	Analysis	Result
1.	Density 20°C/20°C (g/ml)	0.9597
2.	Refractive index ($_nD^{26}$)	1.5486
3.	Solubility in ethanol 90%, At T = 20°C ± 3°C	Soluble in any comparison
4.	Acid value (mg KOH/g sample)	9.2618
5.	Ester value (mg KOH/g sample)	4.5549
6.	Another compound	
	Fatty acids	(-)
	Kruing oil	(-)
7.	Optical rotation T = 20°C	-9.660
8.	Viscosity (cp)	14.4490

Heat energy was transferred from one body to another by one of three fundamental mechanism, conduction, radiation and convection. The rate at which heat was transferred is proportional to a driving force, which is a function of the temperature difference between the heat source and its ultimate destination, and a resistance to the heat flow. For heat transfer by conduction, the resistance to heat flow was a function of material thickness and its thermal conductivity. Obviously, the thicknesses of the evaporator wall and its construction material had a significant bearing on the rate of heat conduction. Heat transfer by radiation increased by the difference of the fourth power of the absolute temperature of the emitting body and receiving body, while the resistance to flow was related to a factor called emissivity. Emissivity is a measure of the surface's ability to absorb or reflect radiation of given wave lengths. A very highly polished metal surface reflects most of the radiant heat while a dull black body, such as an asphalt road, absorbs most of the radiant energy. Convective heat transfer is most complex and depends upon the turbulence created in the fluid regime and the mixing of molecules of different energy levels. The turbulence created depends upon the fluid velocity, fluid viscosity, the mass flow rate and the configuration of the restraining physical barriers. The driving force causing distillation was measured by the temperature difference (ΔT) between the heating element and distillation temperature of the process fluid inside the distillatory body.

The resistance to heat flow to the process fluids was the sum of the resistances through each solid materials, the convective film resistance on each side of the distillatory wall and the resistance created by the fluid film flowing across the distillatory wall. Decreasing all of these resistances to heat flow decreases the response time between the application of heat and its transmission to the evaporating fluid. It also minimizes the driving force required, the temperature difference between the heating source and distillation temperature of the fluid inside the system.

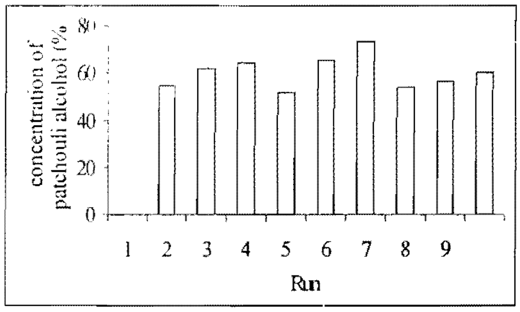

Fig. 2. Concentration of patchouli alcohol at the residue fraction analyzed by GCMS.

In molecular distillation, the desired final product is usually the distillate. In this experiment the desired product was in the residue. This means that, patchouli alcohol was contained in the residue. Patchouli alcohol has a molecular weight about 222.37 g/mol and boiling point about 280.23 °C (553.37 K). This boiling point was higher than other components in the patchouli oil, therefore, patchouli alcohol was in the residue fraction. Figure 2. Showed the result of patchouli alcohol concentration was in the residue fraction as a main product. The higher concentration of patchouli alcohol which remains from this experiment has 73.37% from 19.37% of patchouli alcohol concentration which remain in crude of patchouli oil found at 6th run process. Thereby the concentration of patchouli alcohol increased a lot as effect of wiper rate and heating mantle temperature. the operation condition at 7th run process are wiper rate at 70 rpm and heating mantle temperature at 100°C, meanwhile this is the optimum condition to produce a highly purity patchouli alcohol in patchouli oil. Figure 3. showed a GCMS analyzing for that optimum process condition, and Figure 4 show a GCMS analysis for patchouli oil raw material.

Figure 3. Showed that still there are impurity in patchouli oil, but the result of GCMS indicate that content of impurity pursuant to result of process at optimum condition decreased and concentration of patchouli alcohol increased. Therefore, further research is required to eliminate content of impurity in product after conducted a purification using distillation molecular. In this case, effect of wiper rate and mantle heating temperature play an important role in purification of patchouli oil. In

general, the strand regime of the liquid movement on the evaporator can be considered favorable from the point of view of molecular distillation. The liquid dwells on the distilling cylinder for a prolonged period of time, and there was enough liquid to be mixed during the increased hold-up on the distilling cylinder. Moreover, the wiper mixes the liquid at the high rate in the direction of the distillator's axis in the strand regime, what enables to enrich also lower distillator's parts with the volatile component. Table 2. and 3. showed the physical analysis of patchouli oil in residue and distillate fraction respectively.

CONCLUSION

A variable wiper rate and heating mantle temperature gave an effect on to purification process of patchouli oil using distillation molecular method. The result showed optimum condition at a wiper rate and mantle heater temperature of 70 rpm and 100°C, respectively. GCMS analysis indicated that optimum condition produced high purity of patchouli alcohol enough about 73.37%. As a seen that result of GCMS there are still quite a lot of component of impurity, but after the purification process the impurity content decreased. This matter required to get a special attention in terms to produce patchouli alcohol with high purity

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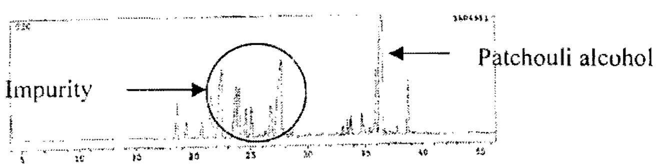

Fig. 3. The result of GCMS analysis of patchouli oil raw material.

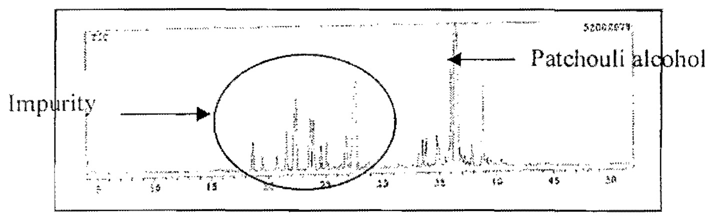

Fig. 4. The result of GCMS analysis for patchouli oil at the optimum process condition

Table 2. The result of physical analysis of patchouli oil in residue fraction, rich patchouli alcohol.

Operating condition		Density 20°C/20°C (g/ml)	Refractive index (nD^{26})	Acid value (mg KOH/g sample)	Ester value	Optical rotation T=20°C	Viscosity (cP)
Vw_1=60 rpm	T_{HM}=80°C	0.9809	1.5088	14.689	11.289	-10.380	35.086
	T_{HM}=90°C	0.9817	1.5092	17.481	12.714	-10.864	35.364
	T_{HM}=100°C	0.9862	1.5112	20.052	17.659	-11.252	39.957
Vw_1=60 rpm	T_{HM}=80°C	0.9965	1.5091	17.489	13.144	-10.454	31.971
	T_{HM}=90°C	0.9884	1.5111	17.847	14.838	-10.976	30.668
	T_{HM}=100°C	0.9865	1.5112	17.617	16.254	-11.037	29.191
Vw_1=60 rpm	T_{HM}=80°C	0.9776	1.5094	15.285	11.291	-10.608	21.669
	T_{HM}=90°C	0.9848	1.5099	18.071	12.703	-10.746	27.312
	T_{HM}=100°C	0.9867	1.5119	18.434	14.833	-10.924	31.488

Note : Vwl = wiper rate, rpm.
T_{HM} = heating mantle temperature, °C.

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