Design and construction of a prototype extractor of aromatic plant essences with supercritical fluids

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Abstract—This project contributes to the research for obtaining essences applied in different areas of the industry such as food, medical and cosmetology, among others. The method of extracting essences by supercritical fluids is a method that optimizes extraction time, although its application is more costly than conventional methods. The prototype extraction plant has a booster gas pump, extractor and separator, the high pressure cylinders are manufactured in steel 304 for its applications in the food industry, under the ASME standard section VIII division 1 in the design of the high pressure cylinder (extractor), for the stage of separation of the essence is used a vertical biphasic separator. Tests are carried out on the prototype plant with emphasis on plants native to the Ecuadorian Andean region such as molle and paico; the extraction point was determined by experimentation, using supercritical CO₂ obtained essences without solvent residues and in less time of processing than conventional methods, emphasizing that the extraction by supercritical fluid is a process of high initial investment, but with low cost of operation for the facility with which its operating conditions can be controlled.

Index Terms—Supercritical fluid, extractor, two-phase separator, essence, extraction point.

I. INTRODUCCIÓN

The tendencies to acquire healthy products, with lower fat content or containing oils, for applications in the different industrial areas such as food, pharmaceuticals, cosmetics, etc., involving a great deal of the extraction efficiency of each method, thus evidencing an increase in the cost associated with non-conventional separation methods; one of them is supercritical fluid extraction, which entails a high cost in its construction due to the slow kinetics of the process, since high pressures are normally used, and mechanical agitation is difficult to apply to the system[1].

The present research seeks to find the efficiency of the supercritical fluid extraction method, based on a specific system designed and built, which is detailed results obtained in the evaluation of the extraction of essence from molle and paico plants that are typical plants of the Andean region of Ecuador, through CO₂ in supercritical conditions. A critical point of extraction of such plants is proposed through trial and error experiments to determine whether the design is really efficient.

Supercritical fluids are based on a periodic process in which the fluid in supercritical parameters is used as solvent[2]. If the pressure and temperature of a fluid is adequately raised to a point called a critical point, the density of the liquid and gaseous states is equalised, at this point the distinction between gas and liquid is blurred, leaving only one supercritical phase. The critical point takes place at a specific temperature called Critical Temperature, Tc, and at a specific pressure called Critical Pressure, Pc.

Carbon dioxide is one of the ecological solvents, called Green Chemistry [2]-[3], because it is inexpensive, non-toxic, non-flammable and easy to administer, which acquires supercritical fluid conditions at a temperature of 304.1 °K (31.1°C) and at relatively moderate pressure of 7.39 MPa (1072 psi) [3]-[4]-[5] as shown in figure 1.

FSC CO₂ has the ability to penetrate through the solid matrix and dissolve the desired extract due to its dual gaseous and liquid characteristic[6], has a great solvent power due to its similar density to that of liquids, likewise it has a good mass transfer due to its viscosity similar to that of gases, which makes it the most effective solvent.

The principle of supercritical fluid extraction depends on the solvent power of the fluid according to the operating pressure and temperature [7], since by modifying each variable the fluid can be presented in various solvent states, as shown in figure 1, for the solvent CO₂.
II. PROTOTYPE DESIGN

A. Sample

Two aromatic plants from the Andean region are chosen:

The paico, (Dysphania ambrosioides) or (Chenopodium ambrosioides) its leaves are the part of the plant most used in folk medicine, as antihelminthic and also as antifungal[8].

Molle (Schinus molle) is important in terms of the possibility of exploitation of natural resources because of its special metabolism[9].

The two plants were purchased in a local market, each sample was appropriately purified and dried at room temperature to remove natural waste.

B. Supercritical extraction process

Figure 2 shows the supercritical fluid extraction process, which is divided into four essential stages[2]:

1. Feeding: The raw material (sample) and solvent fluid must be stored in the extraction chamber.

2. Pressurization: The solvent fluid undergoes an increase in pressure until it reaches its critical point (Pc), where when it becomes supercritical fluid it comes into contact with the matrix to be processed (aromatic plants), and thus drags the solution of interest.

3. Depressurization: Decrease in pressure until atmospheric pressure level is reached.

4. Separation: The essence extracted from the supercritical fluid is separated by pressure and temperature changes in a biphasic separator.

The equipment necessary for the extraction with supercritical CO2 is selected based on the thermodynamic states that the equipment will work in its different stages, keeping in mind the conditions of the CO2 critical point as a basis for the extraction of molle and paico leaves.

Figure 3 shows the diagram Pressure vs Entalpia of CO2 in which the states of the extraction process with supercritical CO2 from the prototype are located.

State 1: Supply of a compressed CO2 cylinder, the state of the substance in this stage is defined by the following parameters \( P_1 = 700 \text{ psi}, T_1 = 15 \degree \text{C}, \text{gaseous} \).

Status 2: When CO2 is in the gaseous state, the pressure is increased so that the fluid subsequently enters the extraction chamber. The pressure is increased from stage 1, by means of a compression pump for high-pressure gas, the temperature will also increase. Process 1-2 is an isoentropic
compression, because CO2 meets the following parameters: 
P₂ = 1100 psi, T₂ = 23 °C.

State 3- In this state, the pressurized fluid deposited in the extractor rises to a temperature higher than Tₖ= 31.1°C so that the CO₂ can reach its supercritical state and in this way the essence of the sample can be extracted. The extractor vessel is completely airtight, the fluid volume will remain constant and in conclusion there will be an increase in pressure. In process 2-3, an isocoric temperature increase is highlighted because at this stage the state of the substance is defined by the following parameters: P₃=1674 psi, T₃= 35°C.

State 4- In this state the process is based on depressurizing the CO₂ in such a way that it leaves the supercritical state for the subsequent separation of the essence. It is known that for pressures of less than 72.5 psi CO₂ passes into a state that forms a solid gas mixture which would make the separation process difficult[13]. So we must depressurize it to a value that keeps the CO₂ in a gaseous state and be able to separate it from the essence. Process 3-4 will be an isenthalpic expansion such that the state of the substance at this stage is defined by: P₄=500 psi, T₄=31.3°C, superheated steam.

C. Supercritical CO₂ pilot plant

The pilot plant must be designed in a complex manner capable of operating at high pressures and is therefore expensive compared to conventional methods [5].

The pilot plant was designed and built, capable of handling high pressures that exceed the critical point of CO₂, maintaining temperature and pressure variables without exceeding the safety factor of the material; this is composed of the following elements, figure 4:

V1, V2, V4, V5, Ball Valves, 1/4 NPT stainless Steel.

V3 and V6, Expansion valves, CO₂ when depressurized loses its supercritical properties and it is possible to separate the solvent extract, 1/4 NPT stainless steel needle valve was used.

M1, M2 and M3, 304 stainless steel pressure gauges with 3000 psi capacity, 1/4 NPT inlet.

The CO₂ cylinder (1) stores carbon dioxide in a gaseous state from a cylinder which can be acquired commercially.

The pump (2) compresses the gas to a pressure above the critical pressure of CO₂, using the Air Driven Gas Booster, model STA40, minimum inlet pressure: 217 psi, maximum outlet pressure: 4640 psi, inlet and outlet connection: 1/4 NPT.

In the extraction chamber (3), the extraction cylinder is designed on the basis of ASME section VIII division I standard, with a capacity of 1/2 litre, this parameter is used to calculate the measurements of the extraction vessel, resulting in the dimensions of figure 5.

Length of high-pressure vessel:

\[
L = \frac{\pi V}{D} \quad (1)
\]

For the diameter of the pressure cylinder we use the minimum diameter required by the ASME standard (figure 5). The design of the high-pressure vessel must take into account a safety factor of 10%.

\[
P_{\text{diseño}} = 1,1 P_{\text{extracción}} \quad (2)
\]

The container shall bear the internal pressure as the only load, so it shall be subjected to 2 main types of stress.

Circumferential stress

\[
P = \frac{P + R}{S \cdot E - 0.6 \cdot P} \quad (3)
\]

\[
P = \frac{S \cdot E - 0.6 \cdot t}{R + 0.6 \cdot t} \quad (4)
\]
Longitudinal stress

\[ t = \frac{PR}{2 \times 5 \times E + 0.4 \times P} \]  \hspace{1cm} (5)

\[ P = \frac{2 \times 5 \times E + t}{K + 0.4 \times t} \]  \hspace{1cm} (6)

\[ Dg = \sqrt{\frac{4 \times Ag}{\pi}} \]  \hspace{1cm} (9)

Separator height

\[ H_t = H_g + H_l + H_d + 2 \times H_o \]  \hspace{1cm} (10)

**Fig. 4.** High pressure cylinder drawing.

The temperature regulator (4) consists of a TCM-RR4 thermostat and a J-type thermocouple.

The two-phase separator (5) is capable of separating CO2 in the gaseous state at high temperature at the top while the extract in the liquid state is extracted at the bottom.

The design of a vertical separator shown in Figure 6 details the inner diameter and length for separation.

Terminal speed

\[ V_t = \text{terminal speed (ft/s)} \]
\[ \rho_L = \text{liquid density (lbm/ft}^3\)) \]
\[ \rho_g = \text{gas density (lbm/ft}^3\)) \]
\[ K = \text{Souder Brown constant} \]

\[ V_t = K \times \sqrt{\frac{\rho_L - \rho_g}{\rho_g}} \]  \hspace{1cm} (7)

Design flow rate

\[ Q_{g} = \text{Gas flow rate.} \]

\[ Q_{g}(diseño) = 1.2 \times Q_{g} \]  \hspace{1cm} (8)

Gas outlet diameter

\[ Ag = \text{Gas containment area} \]

At the Co2 outlet, once the separation of the essence has been carried out, the gaseous CO2 is collected for its subsequent reuse.

In the extract collector, the essence accumulates in the bottom part of the biphasic separator, by means of a valve the samples are collected.

**III. EXPERIMENTAL ANALYSIS**

Initially, 20 grams of plant is entered into the extraction chamber, we proceed to release the source CO2 and then power the booster gas, so that the sample is first saturated with gaseous CO2 that becomes supercritical when the pressure in the extractor increases to greater or equal to the critical (1100 Psi)[10], once the critical pressure is established, we proceed to reach the critical temperature, starting from the ambient temperature of 23.6 \(^\circ\) C to a temperature greater
than or equal to the critical (31.1°C), and we maintain it in supercritical state in a period of time of 15 to 20 minutes for extraction, because by subjecting the raw material to longer time in supercritical conditions the essence can be destroyed, consequently FSC is depressurized to pass to the biphasic separator and thus obtain the essence in liquid state.

Once the first cycle is completed, with the same initial sample, the procedure already specified is immediately restarted in order to obtain a second quantity of essence from the same sample.

In a third consecutive cycle no essence is obtained, immediately the separator is opened and it is observed that the raw material (plant) has a discolored and dried aspect. This makes it possible to conclude that 2 consecutive extraction cycles are sufficient to perform an extraction.

In order to determine the extraction point of the Molle plant with the above conditions the efficiency is low, it is decided to increase the conditions of pressure and temperature up to 1400 Psi for safety reasons it is established not to exceed 1500 Psi, so as not to subject the separating cylinder to an effort in which it can begin to show leaks, in the static analysis it can be observed that the threads of 1/4 in. are the first elements to yield before the stress of a high pressure.

IV. RESULTS

The extraction point of paico and molle cannot be calculated by means of an equation, the optimal extraction point can be obtained by means of experimental tests in which three variables are assigned as: critical pressure, critical temperature and extraction time.

In addition, the extraction efficiency is calculated in each case, given by the following equation [11]-[12].

\[ \eta = \frac{n_c}{n_i} \times 100 \]  

\( n_c: \) oil mass extracted in gr.

\( n_i: \) mass of the initial sample in gr.

A) Paico

To extract the essence, 20 grams of plant are entered, 2 cycles are carried out with the raw material.

\[ E_{(\text{Paico})} = \frac{n_c}{n_i} \times 100 \] (11)

B) Molle

For the extraction of essence of 20 grams, 2 cycles are carried out with the raw material, to then remove the used one and place new raw material in the extractor.
In the first extraction cycle 0.4 ml of crude essence is obtained, the second extraction cycle of the raw material (Schinus molle) is carried out at a critical pressure condition of 1350 Psi, obtaining 0.7ml of crude essence in a 2 cycle extraction period.

In this plant, minimum results are obtained and we can conclude that it needs a higher pressure and temperature, but for safety reasons it is established not to exceed 1300 PSI in order to avoid deformations in the high pressure cylinder.

**Fig 9. Molle, Pc and Tc behavior as a function of time.**

The process efficiency curve for molle extraction is shown in Figure 10, based on the set extraction point of 1300 psi.

**Fig 10.-Efficiency in the Molle extraction process.**

It is determined that the average extraction time is 15 to 20 minutes for both paico and molle.

V. CONCLUSIONS

- An extraction and separation system is built where, due to its operating conditions, instruments are necessary to define the state of the system, since this will allow the operator to make the decision of opening and closing the valves.
- High-pressure coated hoses are used at the inlet as well as on the inside of the system as they are capable of withstanding pressures up to 5000 psi for air, gas or liquid.
- The additional electrical resistances installed in the separator dilute particles of essence in the solidification process at the moment when the supercritical fluid is depressurized inside the separator, in this way liquid particles are added to the extract obtained.
- Of the equipment selected for the pilot plant, the most economically representative element is the gas booster pump for increasing CO2 pressure, representing 50% of the total estimated price for the construction of the pilot plant.
- Extraction of essences by supercritical CO2 results in solvent residue-free essences and shorter processing times, resulting in a raw sample with a higher percentage of purity than conventional methods.
- Supercritical fluid extraction is a high initial investment process, but with low operating costs due to the ease with which its operating conditions can be controlled.
- The efficiency of the extraction process depends directly on the critical point set in the separation chamber as a function of time.

REFERENCES


