DESIGN AND MANUFACTURE OF A SUPERCRITICAL CO_2 LIQUID EXTRACTION MACHINE FOR EXTRACTING BIOACTIVE COMPOUNDS
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DESAIN DAN PEMBUATAN MESIN EKSTRAKSI SUPERKRITIS BERBASIS CO_2 CAIR UNTUK MENGEKSTRAKSI SENYAWA BIOAKTIF

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ABSTRACT
Bioactive compounds in animal and plant cells have many benefits for human health, such as antioxidants, antibacterial, anti-inflammatory, and anticancer. Extraction and separation of bioactive compounds from other compounds is an important step, and commonly, conventional methods are used, but these methods have disadvantages, like producing unwanted compounds. Alternative methods can be conducted using supercritical fluid extraction, but this equipment is expensive and has a small capacity. So, this study aims to produce functional and structural designs and manufacture supercritical fluid extraction machines using carbon dioxide solvents (CO_2) operating with a semi-continuous system. This research succeeded in designing and manufacturing a supercritical fluid extraction machine using carbon dioxide (CO_2) solvent that operates in a semi-continuous system for the extraction of bioactive compounds, with main components including cover frames, supercritical extractor chamber, low and high-pressure CO_2 tubes, compressors and boosters, pipelines, direct valves, manometers, heating, cooler, expanders, reservoirs and automatic control. Moreover, the preliminary simulation test studies revealed that the supercritical extractor chamber could withstand an absolute pressure of 1000 bar, a temperature of 300°C, and a work capacity of 1 L. It indicated that the supercritical CO_2 fluid extractor system was performing well for the conditioning of the extractor chamber, which is generated using a booster and controlled by a one-way valve. Then, the extract is transferred to the separation chamber to separate the CO_2 gas. Then, CO_2 gas is returned to the low-pressure CO_2 tubes for recycling and reuse for the following process.

ABSTRAK
Senyawa bioaktif pada sel hewan dan tumbuhan memiliki banyak manfaat bagi kesehatan manusia, sebagai antioksidan, antibakteri, antiinflamasi, dan antikanker. Ekstraksi dan pemisahan senyawa bioaktif dari senyawa lain merupakan langkah penting, dan biasanya dilakukan menggunakan metode konvensional, namun metode ini memiliki kelemahan seperti menghasilkan senyawa yang tidak diinginkan. Metode alternatif dapat dilakukan dengan ekstraksi fluida superkritis, tetapi peralatan ini mahal dan kapasitasnya kecil. Maka, penelitian ini bertujuan untuk menghasilkan desain fungsional dan struktural serta pembuatan mesin ekstraksi fluida superkritis menggunakan pelarut karbon dioksida (CO_2) yang beroperasi dengan sistem semi kontinyu. Penelitian ini berhasil merancang dan membuat mesin ekstraksi fluida superkritis menggunakan pelarut karbondioksida (CO_2) yang beroperasi dalam sistem semi kontinyu untuk ekstraksi senyawa bioaktif, dengan komponen utama meliputi rangka penutup, ruang ekstraktor superkritis, rendah dan tabung CO_2 bertekanan tinggi, kompresor dan boosters, saluran pipa, katup langsung, manometer, pemanas, pendingin, ekspander, reservoir dan kontrol otomatis. Selain itu, studi uji simulasi awal mengungkapkan bahwa ruang ekstraktor superkritis dapat menahan tekanan absolut sebesar 1000 bar, suhu sebesar 300°C, dan kapasitas kerja sebesar 1 L. Hal ini menunjukkan bahwa sistem pada ekstraktor cairan CO_2 superkritis bekerja dengan baik untuk pengkondisian ruang ekstraktor, yang dihasilkan menggunakan booster dan dikendalikan oleh katup satu arah, dan ekstrak dipindahkan ke ruang pemisahan untuk memisahkan CO_2. gas dari ekstrak (senyawa bioaktif). Gas CO_2 dikembalikan ke silinder CO2 bertekanan rendah untuk didaur ulang dan digunakan kembali untuk proses selanjutnya.

INTRODUCTION

The appropriate extraction method for bioactive compounds from various herbal plants has been widely studied. Conventional extraction methods are commonly used to extract bioactive compounds from these commodities. However, the disadvantage of this method is that it produces unwanted residues, and extract results can change because of the oxidation process during solvent separation (*Chen and Ling, 2000; Khanyile et al., 2022*). The extraction process using fluid at temperatures and pressures above the critical point is an alternative method to prevent the oxidative process of extraction results. Compared to conventional methods, such as liquid organic solvents, supercritical fluids have properties of a higher value of diffusivity and lower values of density, viscosity, surface tension, and the nature of the supercritical fluid can vary widely by regulating operating conditions (*Jha and Sit, 2021; Markom et al., 2012*), so that there is better flavor and fragrance obtained (*Dias et al., 2021*), and also conservation of its biological properties in their extracts *(Hämäläinen and Ruusunen, 2022; Tita et al., 2021*). Typically, carbon dioxide is a type of solvent used in supercritical fluid extractions due to the fact that the CO_2 gas has non-toxic and non-burned properties, is safe for the environment, has abundant material sources, has relatively low prices with high purity, and is suitable for extracting dangerous materials against low heat, low volatility, and polarity (*Santo et al., 2021*), and is easily separated from the material extracted (*Patil et al., 2021*). Carbon dioxide (CO_2) turns into a supercritical state when it is at critical pressure and temperature of 7.28 MPa and 304.1°K respectively *(Jin et al., 2021*).

The high-pressure gradients during pressure release can produce extracts free from living creatures and spores with a longer shelf life than the extract using standard solvents (*Santo et al., 2021*). The main drawbacks of Supercritical Fluid Extraction (SCFE) are the use of critical pressure that prevents system development for production scale, expensive equipment price, and low dielectric constants (*Fikri et al., 2022*). CO_2 and extractor storage tanks must be isolated correctly and equipped with a safety system. Supercritical CO_2 is nonpolar and lipophilic, whereas extraction capability is limited by polarity (*Dauber et al., 2022; Roy et al., 2022*). Co-solvent is recommended to improve the solubility of target compounds and or to increase extract selectivity by carrying out operations at lower pressure (*Pratiwi et al., 2018*). The decision of whether co-solvent can be considered a suitable companion solvent must be examined by cases. Supercritical Carbon Dioxide (SC-CO_2) shows good potential as a solvent to extract bioactive compounds from natural products *(Lefebvre et al., 2021*). However, until today, the scale of SC-CO_2 is still a small capacity, with a high price *(Larocca et al., 2023*). SC-CO_2, with a large capacity, needs to be adequately designed and manufactured with low operating costs to support the extraction of bioactive compounds from nature and their industrialization in the future. Therefore, this study developed a semi-continuous supercritical fluid extraction machine to reuse CO_2 solvents continuously and for the re-utilization of CO_2 in supercritical fluid extractions by flowing CO_2 gas and separating from the expander tube to the CO_2 tube at low pressure, with the hope of saving process time and producing high-quality extract materials.

MATERIALS AND METHODS

1. Basic Design of Machine

The design extraction machine is based on the theory of the supercritical method, where it is assumed that the solvent in supercritical conditions has high diffusivity so that the extract will have a very high purity value. In this case, the solvent to be used is carbon dioxide (CO_2). Carbon dioxide (CO_2) used in supercritical fluid extraction must be in supercritical conditions above the critical point (a specific temperature and pressure), so that it undergoes a phase change to become a gas. Carbon dioxide (CO_2) as a solvent has relatively moderate critical properties, around 31.1 °C for temperature and 73.8 bar for pressure (Laboureur et al., 2015). Thus, it is necessary to design a machine that can create supercritical conditions for carbon dioxide (CO_2), reacting carbon dioxide (CO_2) with materials, separating carbon dioxide (CO_2) from materials, and repeating the carbon dioxide (CO_2) cycle.

2. Engine Power Design

The supercritical reactor was built using a commercial compressor of the "lakoni" brand type "Imola 125", with electrical power specifications of 500 W for 2 hours (1 time supercritical CO2 production phase). The temperature controller uses the brand "Omron" type "e5scl" with electrical power of 3.5 W for 2 h for water heating and 2 h for 1 time supercritical CO_2 production. The heater uses the brand "Dernord" type "DN25", with electric power specifications of 750 W for 2 h for water heating and 2 h for 1 time supercritical CO_2 production phase. This study used 2 boosters, 2 temperature controls, and 2 heaters. So, the total engine power for 1 time of supercritical CO_2 generation was 2.507 kWh.

3. Design Calculation

3.1. Calculation of chamber capacity

The planning capacity for the extraction chamber was 1 L, and 37.5 liters for the separator. CO_2 cylinders use a capacity of 2 m^3 based on the following calculation:

$$P.V = n.R.T \tag{1}$$

$$V = \frac{n.R.T}{P} \tag{2}$$

$$V_1 = \frac{n.R.T}{P} \tag{3}$$

$$V_2 = \frac{n.R.T}{3P} \tag{4}$$

$$V_2 = \frac{1}{3}.\frac{n.R.T}{P} \tag{5}$$

$$V_2 = \frac{1}{3}V_1 \tag{6}$$

where: V_1 - volume of CO_2 gas cylinder (m^3); V_2 -volume of supercritical CO_2 cylinder (m^3);

Q - pressure; n - number of moles; R - common gas constant (8.31 J/mol.K); T - temperature (Kelvin)

3.2. Piping planning and calculation

The pipes used are made from SS304 stainless steel. The pipe used is calculated using the Barlow formula (Equation 7) to calculate the wall thickness of the pipe used (*Isaac and Sylvanus, 2022*).

$$t = \frac{P\ x\ D}{2\ x\ F\ x\ S\ x\ E} \tag{7}$$

where: P - pressure (kPa); S - minimum yield strength (kPa); t - pipe thickness (mm); D - pipe outside diameter (mm); F - design factor; E – magnitude of the longitudinal joint

3.2.1. Connecting pipe

The connecting pipe is used to connect the components of the supercritical extraction machine. The connecting pipe will be added with connectors such as tee, elbow, flange, and water nuts to connect components with a 0.5-inch or 21.34 mm in diameter. The data for the connecting pipe on the supercritical extraction machine are as follows: the pressure (P) of 12000 kPa, minimum yield strength (S) of 293000 kPa, outside diameter of the pipe (D) of 21.34 mm, and design factor (F) of 0.72 for pipes, and magnitude of longitudinal joints (E) of 1 for seamless pipes, and these data were calculated using Barlow Formulas as presented Equation 7 to get the pipe thickness. Based on the calculation, the pipe thickness is ≥ 0.607 mm. According to the *National Institute of Standards and Technology (NIST)* of stainless steel pipe thickness table (2023), the pipe thickness was equivalent to 1.65 mm.

3.2.2. Screw pipe

A screw pipe on the supercritical extraction machine is used to heat CO_2 gas and press it. The threaded pipe in supercritical extraction machines is 0.25 inch or 13.72 mm in diameter. The smaller pipe thickness will shorten the heating process. Screw pipe data on the supercritical extraction machine are as follows: the pressure (P) of 5000 kPa, minimum yield strength (S) of 293000 kPa, outside diameter of the pipe (D) of 13.72 mm, and design factor (F) of 0.72 for pipes, and magnitude of longitudinal joints (E) of 1 for seamless pipes, and these data were calculated using Barlow Formulas as presented Equation 7 to get the pipe thickness. Based on the calculation, the pipe thickness is ≥ 0.163 mm. Pipes with a thickness above 0.163 mm were categorized as pipes with schedule 10, namely SS304 pipes with a size of 0.25 inches.

3.3. Planning and calculation of the flow rate

The formula used in planning is the flow rate formula, then the Reynolds formula to determine the flow type. The data used in this study was a flow rate (Q) of 3.34 x 10^{-4} m^3/s, pipe diameter (D) of 0.0213 m, and π of 3.14. Then, the cross-sectional section (A) was found using Equation 8, and the average speed of the flow (v) was calculated using Equation 9, and the average flow speed was obtained at 9.382 m/s. After which, the Reynold number was calculated using Equation 10. Based on the calculation, the Reynolds number was 349,714.05, identified as turbulent (*Cantwell, 2019*).

$$A = \frac{\pi D^2}{4} \tag{8}$$

$$Q = A\ v \tag{9}$$

$$Re = \frac{D\ x\ v\ x\ \rho}{\mu} \tag{10}$$

Where:

Re: Reynolds number; ρ: fluid density 350 kg/m^3; μ : fluid viscosity= 2 x 10^{-5} kg/ m.s

RESULTS AND DISCUSSION

1. Working Principle of Machine

The supercritical fluid extraction process was carried out gradually, placing the raw material in the extractor chamber and filling pure CO_2 into a low-pressure tube. The compressor produced compressed air, and then the booster used the air to pump and compress CO_2 gas from a low-pressure tube to a high-pressure tube into a supercritical CO_2 fluid. Then the supercritical CO_2 fluid was flowed into the extract tube until reaching the supercritical condition and was left in a certain period of steady state to carry out the extraction process. After reaching a certain period, the pipe's faucet connecting the extractor tube and the separator tube was opened to move a mixture of supercritical fluid with extraction material into the separator. The air pressure in the separator chamber was controlled at 1 bar to change the CO_2 supercritical fluid phase to gas *(Alvarez-Henao et al., 2022; Salinas et al., 2020*). Followed by the separation between CO_2 fluids and extracted materials, CO_2 gas is returned, flowing into a low-pressure CO_2 tube to be reused. At the same time, the extract results are held in the separator chamber. The schematic of the working of the supercritical extraction machine is illustrated in Figure 1.

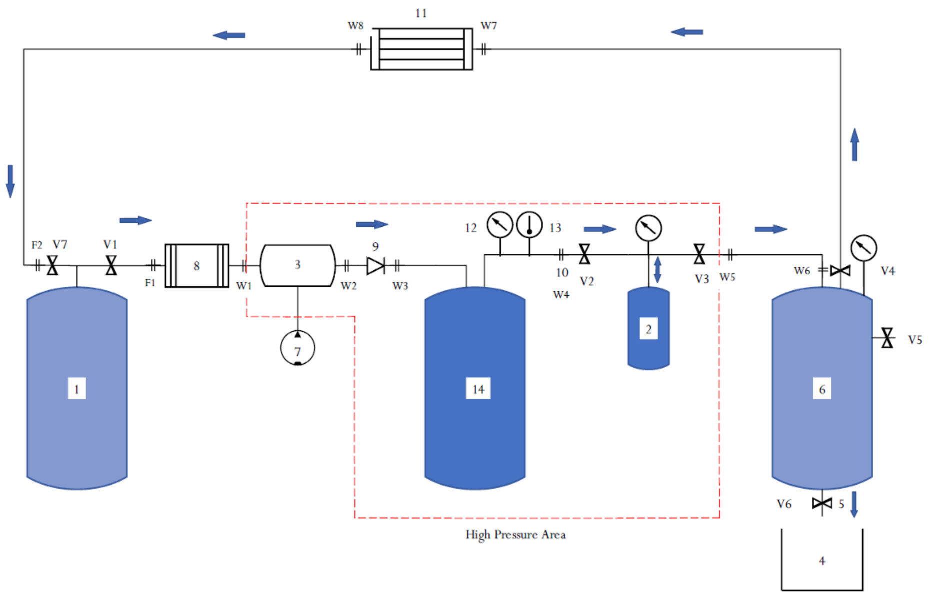


Fig. 1 - The schematic of the working of the Supercritical Fluid Extraction Machines

1. CO_2 tube; 2. extractor chamber; 3. Boosters; 4. Product output tank; 5. Valves (V1-V7); 6.Separator; 7. Compressor; 8. Heaters; 9. Check valves; 10. Water mur (W1-W8);11. Condenser; 12. Pressure gauges; 13. Thermometers; 14 Supercritical fluid storage tubes; 15.Flanges (F1-F2)

2. Functional and Structural Design and Realization

Functional design determines the layout and requirements of machine components and temperature control systems of supercritical fluid extraction. The functional design planning of the supercritical Fluid extraction machine, in the form of technical drawings, is presented in Figure 2. The specification from each component based on planning design and calculations is presented in Table 1, and also the construction realization of the supercritical CO_2 fluid extract machine is presented in Figure 3.

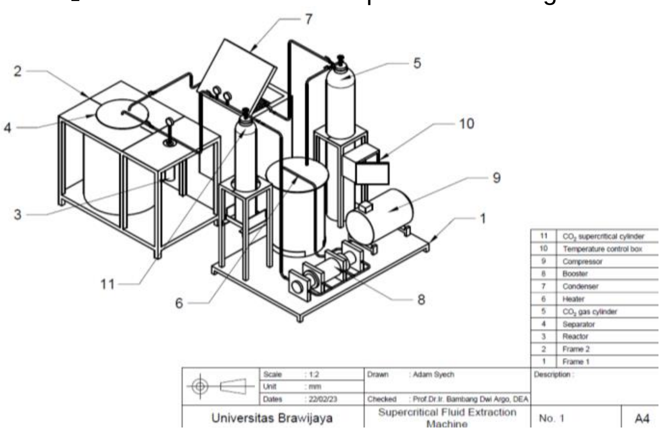


Fig. 2 - The Functional and Structural Design of Supercritical Fluid Extraction Machines

Table 1

Supercritical extraction machine specifications

Machine Component	Specifications
Supercritical extraction machine dimensions	1600 mm x 2600 mm x 1600 mm
Reactor	SS316, 1 L
Separator	SS316, 37.5 L
CO_2 gas tube	6000 mm^3
Supercritical CO_2 tube	2000 mm^3
Heater	90 L, module merk OMRON E5CSL
Condenser	Modified top-door refrigerator from “Midea” brand (commercial brand)
Framework 1 dimension	1400 mm x 970 mm x 900 mm
Framework 2 dimension	1080 mm x 680 mm x 860 mm
Compressor	Compressor brand "Lakoni" type Imola 125, with specifications of 1 hp, 145 L/min, 8 bar, 220 V/ 50 Hz
Thermometer`	100 °C
Pressure gauge	Diameter of 2,5 inches, 280 bars
Booster pump	Booster brand "suncenter" Type DGT40
Valve	Sankyo, SS304, 2,5 inches, max pressure 1000 WOG
Flange	SS304 JIS 10k dan ANCI 300 ½”
Water mur	SS304, 0.5 inches
Check valve	SS304, max pressure 250 bar, 0.5 inches

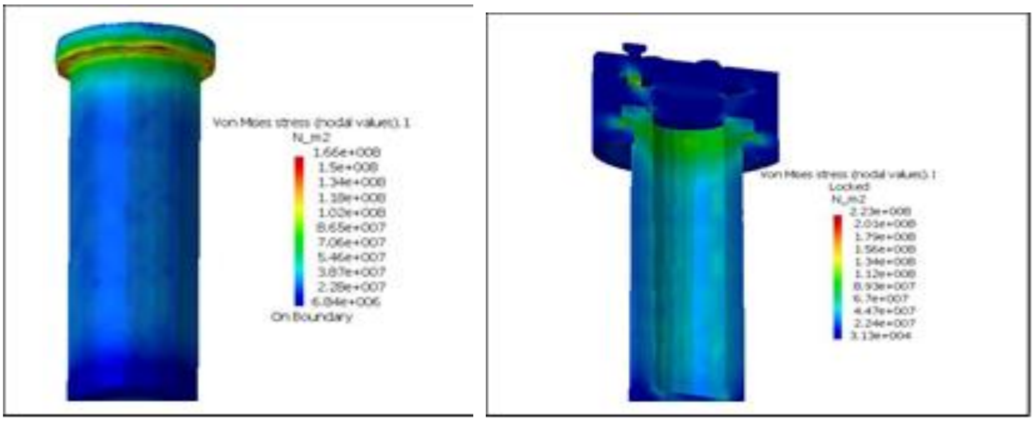
Based on Table 1, Figure 2 and Figure 3 showed that the supercritical CO_2 extractor has a maximum capacity of 1L and can retain the pressure up to 1000 bar using a semi-continuous system with extraction tube material, separator, and a piping system made of SS-316 material.

Fig. 2 - Supercritical fluid extraction machine realization

This extractor engine has a compression and booster capacity of 8 and 350 bar, respectively. Initial test results show that achieving supercritical fluid conditions requires a relatively long compression time of 2.5 h. Efforts to speed up the compression of supercritical fluids are carried out by manipulating the velocity of the airflow produced by the compressor or by increasing the ratio of high and low-pressure CO_2 tubes. In this study, the best ratio of CO_2 gas tube (high pressure) and supercritical CO_2 tube (low pressure) volumes was 3:1 (Table 1).

In addition, the compression speed of the supercritical fluid has an impact on increasing the capacity of the booster, which is 1:40, which means that an increase in pressure of 1 bar in the compressor will increase 40 bar in the booster. Furthermore, the release and termination of the supercritical fluid flow are still done manually using a faucet, which is installed between the CO_2 gas tube and the high-pressure CO_2 tube to ensure that the supercritical conditions in the extractor tube are according to the specified conditions. In the future automatic control is needed to ensure that supercritical conditions are appropriately achieved. However, one of the advantages of this machine is being able to recycle CO_2 gas coming from the separator tube to be accommodated in a low-pressure CO2 tube to be reused in the following process, thereby can reduce operational costs economically. Additionally, controlling the temperature of the CO_2 fluid in the cooling and heating unit on this machine uses an on-off control system, with the resulting control results according to the expected set point, with a deviation value in the range of 1°C than the specified reference value.

In this machine, a cylindrical extractor chamber (Figure 2 and Figure 3) made of SS-316 steel with a wall thickness of 2.5 cm can withstand pressure up to 1000 bar based on the simulation test in Figure 4. Figure 4 shows that the design of the supercritical extractor chamber structure can accept loads at high pressures of up to 1000 bar, indicating that the extractor chamber can be operated under CO_2 supercritical conditions, with a pressure and temperature of 73 bar and 31°C, respectively. The chamber extractor also has a safety factor of more than 14, which indicates that the supercritical fluid extraction machine is technically safe to operate.



(A) (B)

Fig 4 - The pressure distribution simulation in the outer chamber wall (A) and inner chamber wall (B) of the supercritical reactor lid

During the construction of the supercritical fluid extraction machine, there were many problems occurred, especially leakage in between pipe joints, as experienced by other researchers, such as (*Kwartiningsih et al., 2019)*, and various steps were taken to overcome them so that the supercritical extractor could function properly and safely during the extraction process. The list of problems and solutions for solved problems during construction are listed in Table 2.

At the end of the performance test, given the ongoing process of improving the design of the equipment, the supercritical extraction machine was successfully performed from 72.8 to 90.0 bar pressure and temperature of 31.1-35.5˚C, with a capacity of 1L with no leakage. However, reporting on research activities only extends to process simulation and tool design, then the process of separating bioactive compound components will be carried out in the next research phase.

Table 2

The list of problems and solutions for solved problems during construction

Problem	Solution
Leaks in the flange	To overcome the problem of joint leaks, it replaced flange-type connections with watermur seal rubber.
Inadequate installation and lack of Thread Seal Tape (TBA) provision	Ensuring that all installations are tight until the connection rotation is maximized and ensuring that there is a lot of TBA
Leaks at the joints due to damage to the seal rubber and leaks in the check valve	Changes with the new watermur seal rubber

CONCLUSIONS

This study was successful in designing and manufacturing a supercritical fluid extraction machine that uses carbon dioxide (CO_2) solvent and operates in a semi-continuous system for the extraction of bioactive compounds, with primary components such as frames, supercritical extractor chamber, low and high-pressure CO_2 tubes, compressors and boosters, pipelines, direct valves, manometers, heating, cooler, and expanders, and reservoirs and automatic control. Furthermore, the early testing simulation showed that the supercritical extractor chamber could resist an absolute pressure of 1000 bar, a temperature of 300°C, with a capacity of 1L.

The supercritical extraction machine performed well from 72.8 to 90 bar pressure and 31.1-35.5°C at the end of the preliminary performance test. The supercritical CO_2 fluid extractor system was working well for conditioning the extractor chamber, which is created by a booster and regulated by a one-way valve. The extract is then passed to the separation chamber, where the CO_2 gas is separated. The CO_2 gas is then recycled and reused in the following step by returning it to the low-pressure CO_2 cylinder.

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