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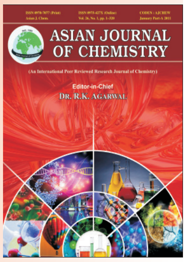

REVIEW

Use of Supercritical CO_2 and R134a as Solvent for Extraction of β-Carotene and α-Tocopherols from Crude Palm Oil

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Supercritical fluid extraction of α-tocopherol and β-carotenoids from crude palm oil offers an excellent method over other existing conventional methods. Supercritical fluid extraction was developed in the early and mid-1980s to reduce the use of harmful organic solvent in the laboratory. The present paper reviews the applications of supercritical fluid extraction technology in extraction of palm oil using CO_2 and R 134a fluids. Carbon dioxide is non- toxic, having low critical pressure (74 bar) and temperature (32 °C) which minimize the thermal degradation of product but it has a limited dissolving power for solutes of high polarity and high molecular weight in the supercritical state and it also requires higher pressure of upto 500 bar and this high pressure operation requires high operation cost and high capital cost. R134a is an alternative low pressure, non-reactive, non-flammable, non-toxic, non-ozone depleting and has comparable solvent properties as that of CO_2. R-134a needs only its temperature or pressure to be controlled in order to control the conditions of extraction or product isolation. This allows simple control of the process.

Keywords: Supercritical fluid extraction, CO_2, R134a, β-Carotene, α-Tocopherols.

INTRODUCTION

With the use of variety of organic solvents in processing of natural product using different separation technology spoliation of final product takes place and finally it also affects human health and environment. Therefore every industry is looking for new processing technology of natural product so as to obtain natural compounds of a high degree of purity[1]. With increasing consumer demand for "natural"products as medicines[2-5], food additives[6], natural pesticides[7], pharmaceutical industries[8] and also in the fields of solid material extraction and fractionation of liquid mixtures (plants and animals) where it is essential to obtain final products of a high degree of purity. These natural products are present in small concentration, therefore it is necessary to develop more effective and selective extraction technique with better selectivity for recovery of these compounds from the raw materials as these are present in very small concentration. Supercritical fluid extraction is considered to be a promising or convenient alternative technique because it provides practical and significant economic advantages over other traditional separation techniques (distillation, evaporation, extraction with "traditional" solvents) for extracting oils and other materials offers a number of advantages, including a lack of solvent residue and aromatic compounds are retained better[9-14].

Supercritical fluid extraction is an efficient extraction technique for solid materials extraction and fractionation of liquid mixtures for separation of active compounds from plants[15-17]. Other conventional methods were carried out at high temperature which leads to the abolition of valuable substances. Supercritical fluid extraction is an advanced separation technique based on the enhanced solvating power of gases above their critical pointand it is also environment friendly over other conventional methods for the recovery of natural products that uses supercritical fluid as the solvent.

Every fluid is having its own critical point which can be defined in terms of critical temperature and pressure. Above the critical temperature fluids cannot be liquefied even on applying pressure, but its density may reach close to liquid state. A pure compound is said to be supercritical fluid if its temperature and its pressure are higher than the critical values (Tc and Pc, respectively). Carbon dioxide and 1,1,1,2-tetrafluoroethane (R134a) are most frequently used supercritical fluids which makes it an ideal solvent for extracting thermally sensitive materials. Carbon dioxide is non-hazardous and non-inflammable. The extraction by supercritical fluid extraction can be enhanced by changing temperature and pressure above their critical points.

Supercritical CO_2 extraction has been used to isolate minor constituents from various oilseed and other products. These include the isolation of tocopherol from soybean and canola oil deodorizer distillates[18,19], sterols and tocopherols from olive oil[20], squalene from olive oil deodorizer distillate and shark liver oil[21,22], phenol and tocopherols from olive leaves[23,24], phospholipids from soybean[25], sterols from canola, corn and cotton seed oils[24] and carotene, vitamin E, sterols, squalene from palm-pressed fiber and palm leaves[27-29].

Extraction of palm oil using supercritical CO_2 from its fruits is relatively rare as compared to supercritical CO_2 extraction ofother vegetable oils. There are few studies which reports the extraction of palm oil from the fleshy mesocarp[30-32] and some others carried out the extraction of palm oil from palm press fiber[33-35]. They mainly focused on the recovery of residual palm oil in palm pressed fiber since palm oil extraction from palm fruit which is best done by screw press technology. Current palm oil extraction industry leaves a residue containing 3-5 % of oil per fruit and high carotene content in the fruit fiber.

Palm oil is well-known to contain a high amount of carotenoids. Typical crude palm oil contains 500-700 ppm carotenes, which contribute to palm oil stability and nutritional value. Conventional processing of palm oil leaves about 3-7 % residual oil in the fruit fibre[29,34,35] and a high content of carotenoids in the pressed palm fibres. Residual fibres from palm oil production contain between 4000 and 6000 ppm of carotenoids, about six times higher than that found in crushed palm oil[29,33]. Until now, palm oil fibre residues that are rich in valuable carotenoids are treated as a waste product, which is destined to be burnt together with empty fruit bunches[32] or transported to the plantation for field mulching[34]. Effective recovery of the carotenes would therefore give a significant advantage to the palm oil industry. The present work reviews the application of supercritical fluid extraction for determination of α-tocopherol and β-carotene in crude palm oil using CO_2 and R134a as a solvent.

β-Carotene in palm oil: β-Carotene is a natural pigment. It is mainly synthesized by plant, microorganisms and animals obtain it from plant as a food. The chemical structure of β-carotene is shown in Fig. 1. Until now more than 600 carotenoids have been isolated from natural sources[36] which are responsible for beautiful colors of many birds, insects and marine animals, as well as the colors of many flowers and fruits. Carotene is a precursor of retinol (pro-vitamin A) which act as an antioxidant to prevent the development of a number of diseases such as xerophthalmia and a night blind disease in human and animal metabolism and it is used in the food processing industry for coloring purposes[37,38]. Crude palm oil contains 500-700 ppm carotenes which is usually lost during palm oil refining[39,40]. Conventional methods used during the processing of palm oil leaves about 3-7 % residual oil in the fruit fibre[29,34,35] and also leaves high content of carotenoids in pressed palm fibers. Residual fibres from palm oil production contain about 4000 and 6000 ppm of carotenoids which is about six times higher than that found in crushed palm oil[32]. Until now, palm oil fibre residues that are rich in valuable carotenoids are treated as a waste product, which is either transported to the plantation for field mulching[34] or burnt together with empty fruit bunches[32]. Carotenoids in crude palm oil give oxidative protection to the oil[41]. Carotene is non-polar in nature and freely soluble in oils and fats[42]. As it can be seen from the Table-1[43] that carotenes are having low vapour pressure which indicates that it has low volatility and high molecular weight shows its difficulty in extraction unless some special properties of the solvent, such as polarity, could only assist the recovery of the active compound[43].

TABLE-1
PHYSICAL PROPERTIES OF CAROTENES[43]

Specifications	
m.w.	536.88 kg/kmol
Density (at 20 ºC)	1000.15 kg/m^3
Vapour pressure (at 70 ºC)	3 × 10^{-6} kPa

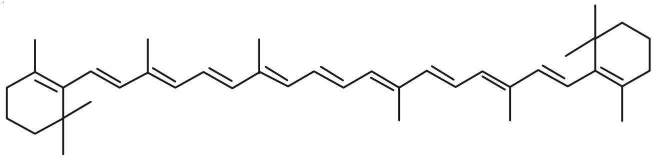

Fig. 1. Structure of β-carotene

Conventional methods based on the solvent extraction of carotene from natural products are time consuming since they require multiple extraction steps and need a large amount of organic solvents, which are often expensive and potentially harmful. Various methods for extracting carotenes from palm oil have been developed including saponification[44], adsorption[45-47], selective solvent extraction[48] and transesterification followed by molecular distillation[49]. However, only transesterification-distillation and transesterification-solvent extraction have been scaled-up to industrial use and these methods are energy-intensive processes since the esters must be vacuum-distilled and the carotenes may undergo thermal degradation. The total carotenoid content (TCC = 9.16-600 mg/100 g) in palm fruits and oils were determined spectrophotometrically[50-57]. Table-1 shows the physical properties of carotenes[43].

Supercritical fluid extraction technology using CO_2 and R134a as a solvent was introduced to prevent degradation of carotene during extraction. Usually extraction and recovery of the carotenes give a significant added value to the oil.

α-Tocopherols in palm oil: Tocopherol (molecular weight 430.7) present in palm fruits and crude palm oils are increasingly used in pharmaceuticals and food industries as a natural antioxidant compound, mainly due to consumer pressure and more demanding regulations regarding the use of artificial antioxidants. α-Tocopherol (Fig. 2) is an important antioxidants for good human health[58-60] and is the highest vitamin E activity for animals and humans[23,45,58] which acts as fat-soluble antioxidant. The main function of α-tocopherols is that of a radical-chain breaking antioxidant in membranes and lipoproteins, as well as in foods[61]. Due to its potential antioxidant property and various functions at the molecular level, it is believed to reduce the risk of cardiovascular diseases and of certain types of cancer, heart diseases, neurodegenerative

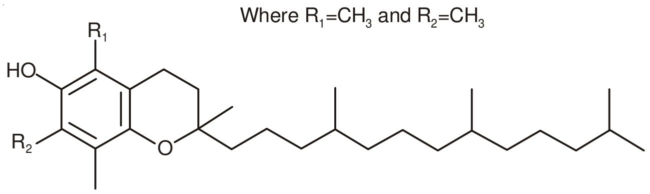

Fig. 2. Structure of α-tocopherol

diseases and diabetes[50,54,62,63]. Deficiencies of these compounds in the human body may cause many diseases such as anemia, neurological dysfunction and myopathies.

Degumming, bleaching and deodorization are the three major processes used during the physical refining method to remove undesirable components for other application[64] from crude palm oil. Different analytical extraction procedures such as pressurized liquid extraction[56], Soxhlet extraction[65,56] and solvent extraction using mixtures of methanol and water in different proportions[50,52,66], 60 % ethanol in water[54] were used for isolation of antioxidants from palm fruits and oils.

Supercritical fluid extraction technology using CO_2 and R134a as a solvent: Supercritical fluid extraction is an advanced separation technique based on the enhanced solvating power of gases above their critical point using supercritical fluid as a solvent. A schematic diagram of supercritical carbon dioxide instrument was given in Fig. 3. Tour in 1822 first reported the behaviour of supercritical fluids and later in 1879 Hannay and Hogarth demonstrated the solvating power of supercritical fluids for solids[67-69].

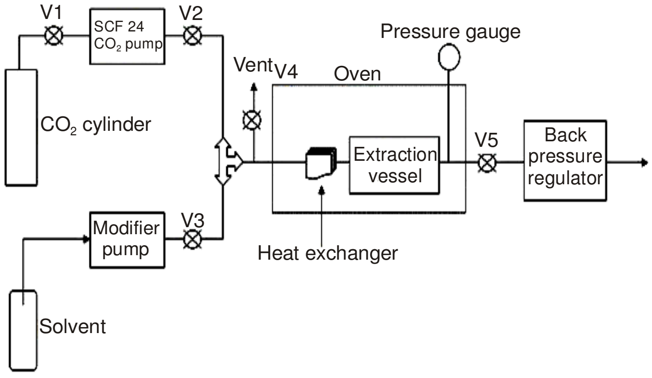

Fig. 3. Schematic diagram of supercritical carbon dioxide instrument

The critical point of a fluid is defined in terms of its critical pressure and temperature and above that temperature only one state exists, the supercritical fluid state. After attaining/reaching that state there is no distinction between the liquid and gas phase and the resulting supercritical fluid has one uniform density[70] similar to liquids and low viscosity similar to gases. Surface tension and heat of vaporization is relatively very low for supercritical fluids[71]. Above critical temperature and pressure fluid cannot be liquefied regardless of the applied pressure which makes it suitable for extraction process by slight variation in pressure and temperature near the critical point. The high density of supercritical fluids leads to greater solubilization of compounds without using multiple solvents while due to low viscosity of supercritical fluids it accelerates mass transfer of analytes and can be easily penetrate in different types of matrices and allows flow with less friction.

There are many factors which effects supercritical fluid extraction technology such as moisture content in the palm mesocarp, sample should contain less than 10 % of moisture for effective extraction[72-74], but it was found that in some cases high moisture content facilitates the removal of oil using supercritical fluid extraction[75,76]. Temperature is also an important parameter for successful supercritical fluid extraction, depending upon the pressure an increase in temperature of the fluid may increase, decrease or have no effect on solubility of the solute. However the density of the fluid decreases with temperature at constant pressure. At a fixed pressure with the increase in temperature the solvent density reduces and thus reducing the solvent power of the solvents. At higher temperature vapour pressure of the compound of interest also increases and thereby the tendency of the solute to dissolve in fluid phase also increases[77].

In general, at higher pressure solvent power is higher and extraction selectivity is smaller[45,77]. Particle size also plays an important role in extraction process, generally particles with diameters ranging 0.25 and 2 mm are used. Very small sized particle formed by grinding results in the loss of volatile compound. Extraction rate increases with decreasing particle size[71,78-80] and with increasing sample surface area to weight ratio[71].

Compounds, which have been used in their supercritical state as a supercritical fluids are listed in Table-2[78] along with their critical parameters. These are mainly consists of carbon dioxide, nitrous oxide, ethane, propane, *n*-pentane, ammonia, fluoroform, sulphur hexafluoride, water and R134a. For a maximum separation of natural product suitable solvent must be selected having low boiling point so that it will be removed easily after processing[81]. Table-3 shows the examples of supercritical fluid extraction technology employed in carotenoid analysis[29,34,35,45,56,82-86].

Usually, 90 % of all analytical supercritical fluid extraction is performed using carbon dioxide (CO_2) for several practical reasons, (a) low critical pressure (74 bar) and temperature (32 °C) which minimize the thermal degradation of product (b) solvent can be easily separated without leaving trace in the processed matrix after extraction (c) it is non-toxic and harmful so it is safe to be used in food production and processing (d) belongs to "clean technology" as there is no formation of secondary products and leaves no environmental hazardous waste so this technology is not harmful to the environment[77,87-92]. In the supercritical state, CO_2 has a polarity which is comparable to liquid pentane and is, therefore, best suited for lipophilic compounds (non-polar) but it lacks the polarity for the extraction of polar analytes[93] and thus has a limited dissolving power for solutes of high polarity and high molecular weight. Thus small quantity of liquid solvents (modifiers such as ethyl alcohol) are added which are easily solubilized by supercritical CO_2 and thus modify the solvent power of supercritical CO_2. The main drawback is that due to larger solvent power selectivity process is lowered and the co-solvent which is liquid at atmospheric pressure will also be collected along with the extracted compound in the separator and one of the advantages of the supercritical fluid extraction; *i.e.*, solventless operation is lost.

TABLE-2
PROPERTIES OF SOME SUPERCRITICAL FLUIDS AT C_p[78]

Supercritical fluid	T_c (°C)	P_c (bar)	Critical density (g/mL)
CO_2	31.0	72	0.47
N_2O	36.5	70.6	0.45
Ethane	32.3	47.6	0.2
Propane	96.7	42.4	0.22
n-Pentane	196.6	32.9	0.23
Ammonia	132.5	109.8	0.23
Fluoroform	26	46.9	0.52
SF_6	45.5	38.0	-
Water	374.2	214.8	0.32
134a	101.2	40.6	-

TABLE 3
SOME EXAMPLES OF SUPERCRITICAL FLUID EXTRACTION TECHNOLOGY EMPLOYED IN CAROTENOID ANALYSIS

Sample	Analyte	Solvent used	Ref.
Vegetable and plasma sample	Carotenoid	CO_2	82
Crude palm oil	β-Carotene	CO_2	51
Solid matrix (palm fruit)	Palm oil	1,1,1,2-Tetrafluoroethane (R134a)	83
Palm oil mesocarp	β-Carotene	134a	84
Pressed palm oil fibre	Carotene		29
Palm oil fruit and palm oil leaves	Tocopherols and carotenes	CO_2	32
Palm-pressed mesocarp	Carotene	CO_2	34
Screw pressed palm fibre	Palm oil	CO_2	35
Lotus bee pollen	carotenoid	CO_2	85
Crude palm oil	β-Carotene	CO_2	86
Pulp of the buriti fruit (*Mauritiaflexuosa*)	β-Carotene	CO_2	76

From the past 30 years, supercritical fluid extraction technology is known and established, however, commercialization of this technology is still limited[81]. Only few industries such as decaffeination of coffee and tea and extraction of hops for brewing have been reported which are using supercritical fluid extraction applications[94].

For satisfactory extraction using CO_2 as a solvent requires higher pressure of upto 500 bar[94] and this high pressure operation requires high operation cost and high capital cost. Attempts were made to use carbon dioxide at lower pressure (40-100 bar) but satisfactory results were not found[95-98]. This is the main reason for limited commercialization of supercritical fluid extraction process using carbon dioxide. Catchpole & Proells and later Wood & Cooper explore an alternative supercritical fluid extraction solvent which can be operated in less intense conditions and at more reasonable coststo fill the environmental and economic needs[99-101].

In contrast, R134a is an alternative low pressure[99,102] non-reactive, non-flammable, non-toxic, non-ozone depleting and has comparable solvent properties as that of CO_2[103] and can be used in the similar manner[78,100]. R134a is a naturally polar solvent and therefore the solubility of polar compounds in R134a is rather sufficient without the addition of any modifier and it can also address the shortcomings of supercritical CO_2 process[104,105]. R134a has a relatively lower critical pressure compared to other solvents and due to its high volatility and boiling point at atmospheric pressure of -26.2 °C, leaves negligible solvent residues in the products and can minimize the operating cost due to lower pressure required during the supercritical fluid extraction process. The use of R134a as an alternative to CO_2 for the extraction of palm from palm mesocarp oil was studied[83,84].

Conclusion

Supercritical fluid extraction is regarded as an alternative and promising technique over other extraction method due to its several attractive features such as supercritical fluids can penetrate more effectively in porous solids as compared to other liquid solvents and thus results in faster extraction due to low viscosity and high diffusivity. Thus by supercritical fluid extraction extraction time was much reduced with better recoveries. Solvation power of fluid in supercritical fluid extraction can be employ by changing temperature and pressure and there by high selectivity was achieved and was useful for extraction of complex samples. Sample concentration is a time consuming process and sometime it results in the loss of volatile component, supercritical fluid extraction does not requires preconcentration process. Thermally labile compounds can be easily studies as supercritical fluid extraction is usually performed at low pressure. In supercritical fluid extraction a very small amount of sample is needed (0.5-1.5 g) and requires only few milliliters of organic solvent. CO_2 can be recycled and reuse further in supercritical fluid extraction processes to minimize the waste generation. Extraction of palm oil (palm mesocarp) using CO_2 as a supercritical fluid gives higher yield as compare to R134a solvent due to its better mass transfer characteristic when operated below its critical point. Even though, in case of polar solutes extraction R134a gives better results as compared to CO_2.

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NOTE

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