

Research article

Enhanced Oil Recovery Using Supercritical Carbon Dioxide With and Without Co-solvents

Shahid Hussain

Graduate in Oil and Gas Technology, Aalborg University Esbjerg · Niels Bohrs Vej 8 · DK-6700
Esbjerg, Denmark.

E-mail: hussainshahid56@gmail.com



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Abstract

Supercritical Fluid Extraction using pure and modified supercritical carbon dioxide has been investigated for the extraction of crude oil at different pressures ranging from 20 to 60MPa with interval of 5MPa at constant temperature (60°C) for 20 minutes of the extraction process. Supercritical Fluid Extraction could successfully extract oil using carbon dioxide which is friendly to the environment and efficient method, consuming less quantity of solvent, time and feasible to recover solvent. Extraction yields and initial extraction rates increased with increasing pressure. Results showed that addition of co-solvents had increased both extraction rates and extraction yields of extracting oil as compared to the extraction process without co-solvent. The co-solvents used to enhance this extraction process were methanol, ethanol, propanol and brine. The selection of these co-solvents were due to their capability to form hydrogen bonding with solutes and enhance the density of supercritical carbon dioxide complying higher miscibility and solubility of solvent in solutes of crude oil. Extraction rates and oil recovery were larger at high density conditions (high pressures for this isothermal extraction). While the addition of co-solvent attributed higher extraction rates and oil recovery even at softer operating pressures. The effect of co-solvents on both variables was following this order Ethanol> Methanol> Propanol> Acetone > Brine. Best operating conditions were selected to obtain higher extraction rates and extraction yields on the basis of results. These conditions were P=60MPa at 60C using ethanol as a co-solvent with concentration of 0.075 g of ethanol / g of crude oil. The weight of crude oil used in every experiment was 40 grams.

To study the effect of concentration of co-solvent on extraction rates and oil recovery, concentration of each co-solvent used was 0.125 g of co-solvent / g of crude oil at operating conditions of 20 to 60MPa at constant temperature (60°C) for 20 minutes of extraction time. Results showed that both extraction rates and oil recovery were increased using higher concentrations of co-solvents. Results were also showing that selecting of specific operating conditions could extract selective components (hydrocarbons) of interest by using supercritical carbon dioxide extraction which could not be feasible with other solvent extraction operation.
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Keywords Supercritical Fluid Extraction, co-solvent, hydrogen bonding, oil recovery, miscibility, solubility

1 Introduction

The use of supercritical carbon dioxide has taken much attention for separation processes in recent years. It has been used in many separation processes in industries including tea decaffeination, coffee decaffeination, and extraction of fatty acids from spending barley, pyrethrum, hops, spices, flavors, fragrance, corn oil and color from red peppers. Other applications include polymer fractionation, polymerization, particle formation for military and pharmaceutical use, and cleaning of electronic and machine parts, textile dyeing etc.

A supercritical fluid contains physical properties intermediate between liquid and gas with liquid like density to lead high loading of solutes, and the pressure-dependent solvating ability of supercritical fluid enables it an excellent solvent for separation processes. Low surface tension, low viscosity and high molecular diffusivity of supercritical fluid enables it excellent mass transfer solvent allowing its better penetration into the sample matrix than liquid solvents. Carbon Dioxide is an excellent fluid to use as a supercritical fluid due to its non-flammability, non-toxicity, lack of chemical residual problem and low critical temperature [1].

Recently, most familiar application of supercritical extraction has been the use of supercritical carbon dioxide in the enhanced oil recovery in order to recover crude oil that cannot be recovered from conventional methods used for recovering of oil from porous reservoir rocks. Total oil discovered in the United States, from 400 billion barrels approximately 300 billion barrels could not be recovered from standards methods [2].

Successful and effective application of any oil recovery technique significantly depends upon its capability to overcome the forces that are trapping the oil in the pores of reservoir rocks. When the reservoir is discovered, oil will be driven to surface automatically due to natural pressure forces in the formation as well as from the expansion of dissolved gas in the oil. For a simple light-oil reservoir, primary production can recover 10 to 20 % of oil in place while for a heavy and viscous oil reservoir; the recovery is even less. Secondary methods can recover additional 20 to 50% of oil in place for light-oil reservoirs, but still not sufficient enough for oil recovery [2].

Water flooding during secondary method causes numerous interfaces in the reservoir rocks due to its immiscibility with oil as displacement proceeds. These oil-water interfacial tensions result capillary forces which trap the oil in the small diverging and converging channels that make up pore space in reservoir rocks. The effects of these capillary forces can be reduced by using enhanced oil recovery specially injection of such solvents which are effectively miscible with oil. When these light hydrocarbon solvents are injected into the reservoir, they finger into the oil, and spread by dispersion or diffusion into the reservoir to decrease the viscosity of oil through the process of dilution. This enhances the production while solvents are recovered and recycled. To obtain significant extraction rates, solvent must have maximum solubility in the oil. However, selection of solvents can also be chosen on behalf of diluting more light ends having more value as compared to heavy ends. If light hydrocarbon solvent has a sufficient concentration in the diluted oil, then it causes the disputing process and reducing the viscosity to a greater extent.

It is observed that the increase in molecular weight of the solvent decreases the amount of asphaltene deposit. While mixing of low molecular weight solvent decreases the average molecular weight of oil and it tends to precipitate asphaltenes because asphaltenes are less stable in low molecular weight system [3].

Unfortunately, solvents effectively miscible with oil are hydrocarbons that are product of recovered crude oil in the first place, and significantly more expensive than the displaced crude oil. From recent studies, it is observed that supercritical CO₂ is significantly miscible with oil and also cheaper as compared to hydrocarbon solvents. So, supercritical CO₂ can be used in enhanced oil recovery to recover maximum oil from the reservoir.

Higher molecular weight (non-volatile) organic compounds have relatively less solubility in supercritical carbon dioxide. To increase the solubility of these compounds in the supercritical carbon dioxide, high operating conditions (pressure and temperature) are required that increase capital cost of this commercial-scale process. Although carbon dioxide is widely used in SFC but it has a drawback of lack of polarity and associated deficiency of specific solute-solvent interactions that will render to high loading or to select polar organic compounds. It is investigated that the addition of a small amount of another solvent called co-solvent to supercritical carbon dioxide can dramatically increase the solvent power of supercritical carbon dioxide [4].

Recently, progressive work has been made to understand solute-solvent interactions in supercritical mixtures. It is found that the cluster formation of solvent molecules around relatively large solute molecules form a local density higher than the bulk density. While the addition of co-solvent is making this more complex due to difference of local and bulk compositions [4].

There is also a change of composition in the region of the vicinity of solutes in a modified SFC with co-solvent. This additional interaction between the co-solvent and solutes is responsible to increase the solubility which is the main function of co-solvent addition. These possible interactions and other mechanisms contributed by co-solvent will generally increase the density of the mixture. This increase in mixture density will increase not only overall solubility but will also cause enhancement in physical interactions such as dipole-induced dipole, dipole-dipole, and induced dipole-induced dipole interactions. Nevertheless, use of polar co-solvent for polar solutes will increase overall solubility due to specific chemical interactions such as complex formation due to charge transfer or hydrogen bonding [5].

In this paper, effect of several co-solvents such as methanol, ethanol, propanol, acetone and brine, on the oil extraction by supercritical carbon dioxide in the broad range of pressure such as 20 to 60MPa was investigated to improve the efficiency of pure solvent (CO₂). Accordingly, in this paper, general trends in yields of supercritical extraction and extraction rates with modified CO₂, operating variable's effect on the recovered oil composition, optimum process conditions for efficient separation, are discussed. Different concentrations of co-solvents such as 7.5 % and 12.5% for ethanol, methanol, propanol, acetone and brine are also used to study the effect of concentration of co-solvent in the extraction process. The pure supercritical carbon dioxide is also used to compare extraction process without and with co-solvent.

2 Materials and Methods

2.1 Materials

The crude oil for the experiments was supplied by the Maersk oil company from the North Sea oilfield. This crude oil was dead, intense dark brown almost black in color. The pure carbon dioxide (99.9%) was supplied by Strandmollen A/S, Denmark. Methanol, acetone, ethanol and propanol of 99.9% of purity were purchased from the VWR Pro - lab. Brine was taken from the North sea.

2.2 Preparation of the Sample

Before inaugurate an experiment, a dry towel and an empty beaker were weighed. The sample consists of towel of 5 grams and crude oil (40 gram). The towel was soaked in crude oil for at least 72 hours to achieve proper saturation. The excess oil was stripped from the towel, and the towel saturated with crude oil was weighed. Then

the sample was put into the extractor carefully so that crude oil from the sample should not leak from the bottom of the extractor. Additionally, mass of co-solvent (either 7.5 % or 12.5%, depending upon experiment) was added into the extractor carefully.

The extractor vessel containing the sample was inserted into the SFE to commence the experiment.

2.3 Apparatus and Experimental Procedure

The experiments of extraction process were performed in Supercritical Fluid Extractor (SFE). Figure 2.3 shows the flow sheet diagram for the supercritical fluid extractor. It is numbered from 1 to 13 to identify every part of the apparatus.

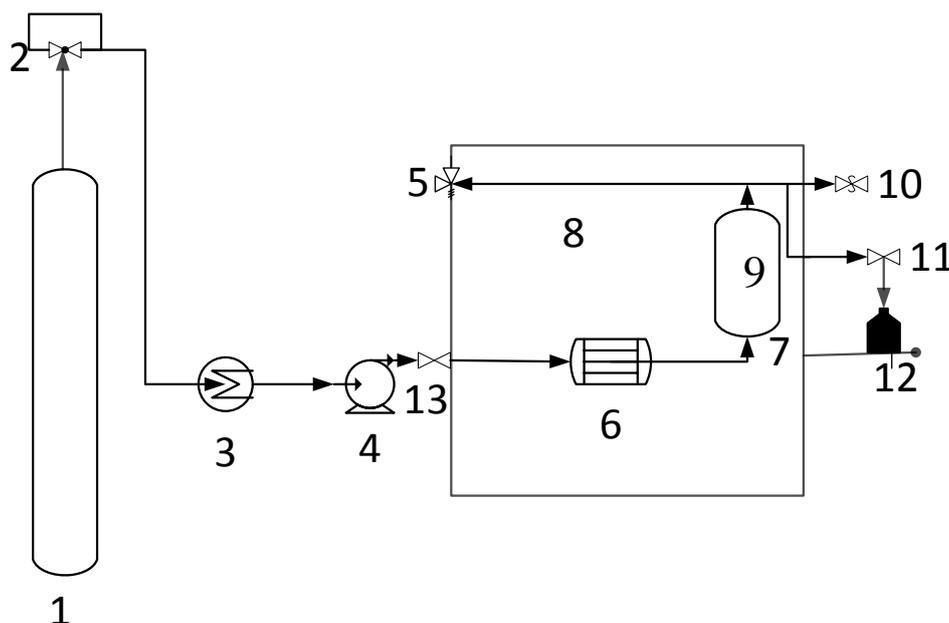


Figure 2.3 Flow sheet diagram for supercritical fluid extractor: (1) - CO₂ Storage Tank, (2) - CO₂ Outlet Valve, (3) - Cooler, (4) - Pump, (5) - Vent Valve, (6) - Pre-Heat Coil, (7) - Extractor Vessel, (8) - Oven, (9) - Sample (Towel saturated with oil), (10) - Outlet Valve, (11) - Exit Valve, (12) - Sample collector, (13) - Inlet Valve.

After introducing the sample into the extractor vessel (7), all outlet valves (5, 10, 11) and inlet valve (13) were closed tightly. When the required temperature of 60°C in the oven (8) and pressure (4) were achieved, outlet valve of CO₂ (2) and inlet valve (13) were opened so that CO₂ could flow inside the extractor vessel (7). The system was left for 20 minutes to equilibrate. A pump (4) was used to maintain the required pressure. Carbon dioxide was continuously supplied to the system by this pump (4). Meanwhile, empty test tubes were weighed to collect the extracted liquid. After 20 minutes, outlet (10) and exit valve (11) were opened to collect the extracted oil in the test tube. The test tubes having extracted crude oil were weighed again to calculate extracted oil.

The extraction of crude oil was ended when it was visually observed that no more extracted crude oil was collected in the test tubes. Afterwards, the inlet valve (13) was closed and outlet valves (10, 11) were opened till CO₂ from the system vanished completely. The towel with un-extracted crude oil was taken out from the extractor vessel (7) and weighed for calculations. The extractor vessel was cleaned properly to make it ready for the next experiment. A constant flow rate of CO₂ was maintained throughout the experiment.

Each experiment was conducted three times to minimize experimental errors, and reproducibility of the data was good.

This apparatus is equipped with High Pressure Alarm (HPA) and High Temperature Alarm (HTA) to ensure the safety. It has also latest control systems to control temperature, flow and pressure resulting in more precise experimental work.

2.3 Calculations for Recovered Oil

The extracted oil was calculated as weight difference of empty tubes and tubes containing extracted oil as described below:

$$W_{\text{ex o}} = W_{\text{ex tube}} - W_{\text{e tube}}$$

Where

$W_{\text{e tube}}$ = the weight of the empty test tube, gram

$W_{\text{ex tube}}$ = the weight of the test tube with extracted oil after experiment, gram

$W_{\text{ex o}}$ = the weight of the extracted oil, gram

The oil recovery was calculated by using following formula

$$R_o = W_{\text{ex o}} / W_s * 100$$

Where

R_o = Percentage oil recovery, %

$W_{\text{ex o}}$ = Weight of extracted oil, gram

W_s = Weight of crude oil (40gram)

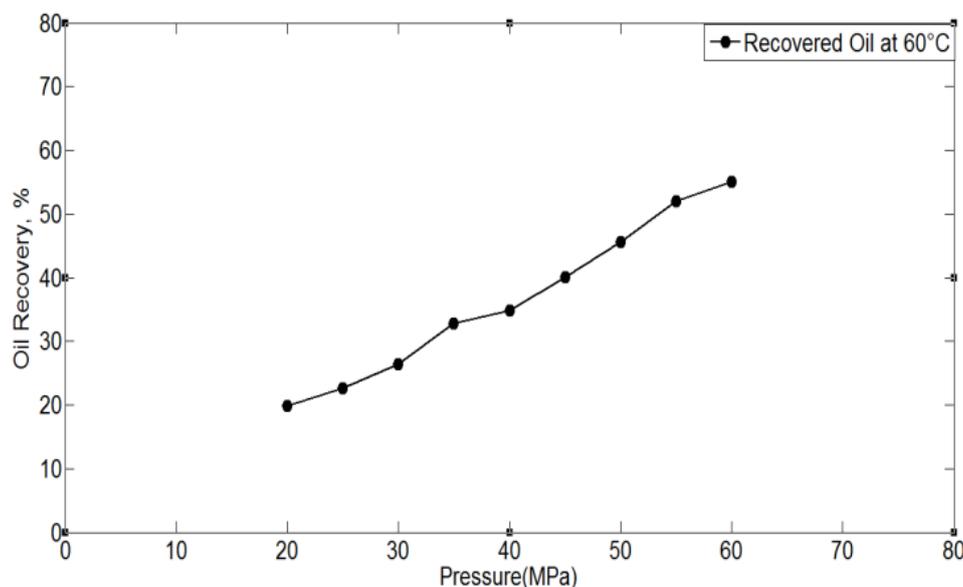
The experimental results were plotted as percentage recovery of extracted oil versus other parameters to explain the extraction process.

3 Results and Discussion

The composition of crude oil is very complex containing highly volatile components like methane to highly non-volatile hydrocarbons. The aim is to obtain maximum recovery of oil possible at optimum operating conditions, and efficient co-solvent.

3.1 Extraction Yields Using Pure Supercritical CO₂

The percentage yield of recovered oil from the extraction process using pure supercritical CO₂ at different pressure range for isothermal process is shown in the figure3.1 (a).



a

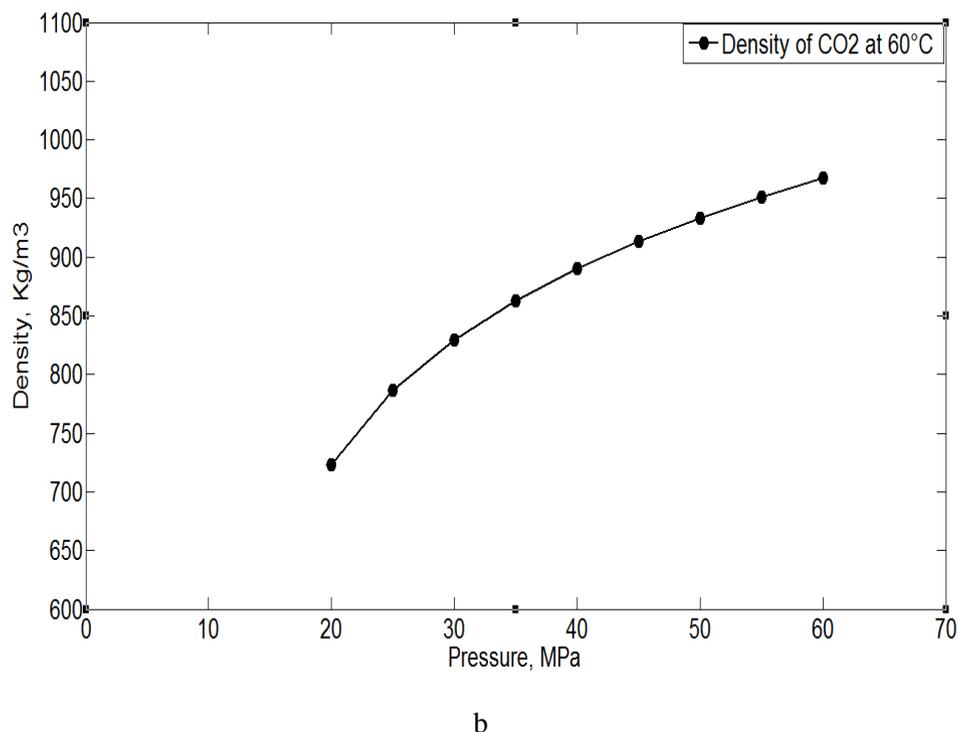


Figure 3.1 (a) Extraction yield obtained with pure Supercritical CO₂, (b) Effect of pressure at a density of CO₂ at 60°C.

Figure 3.1 (a) shows that extraction yield is increasing with increase in pressure for this isothermal extraction process. It is also observed that extraction yield is low under low density conditions (low pressure and high temperature), while the extraction rate is high for high density conditions (high pressure and low temperature).

Figure 3.1 (b) is showing an increase of density of carbon dioxide with operating pressures (supercritical conditions). Density data for CO₂ at operation conditions were taken from NIST (National Institute of Standards and Technology).

It is isothermal process, so high or low density conditions are depending on high or low pressure respectively. This result has agreed with the results obtained from other authors [7-11].

3.2 Extraction Yields Using Modified Supercritical CO₂

Previous result in 3.1 sections showed that extraction yield using pure supercritical carbon dioxide significantly depends upon operating conditions. But it was found from the literature that recovery of oil could be further increase sufficiently using co-solvent with supercritical carbon dioxide. Different co-solvents such as methanol, ethanol, propanol, acetone, and brine are used to investigate the effect of co-solvent addition on oil recovery as well as to know which co-solvent can effectively increase the recovery of oil. Figure 3.2 is showing the cumulative yields of collected crude oil by using the modified supercritical carbon dioxide extraction of crude oil (40 g) under different operating conditions.

These results showed that the percentage yield (solubility and miscibility) of extracted crude oil by supercritical CO₂ significantly depend upon the pressure. At the higher pressure, solubility and miscibility of crude oil are higher rendering the high extraction rate and extracted yields of crude oil. From these results, it was strongly observed that under high density conditions (low temperature and high pressure), the extraction rate and extracted yields of crude oil were high, while under low density conditions (low pressure and high temperature) the extraction rate and extracted yields of crude oil were lower. Thus, for isothermal system, increase in pressure increases the solvent (CO₂) density, crude oil miscibility, solubility and extraction yield.

However, for isobaric system, decrease in temperature increases the solvent density, crude oil solubility and extraction yield. These results were in good agreement with previously reported results [7-11].

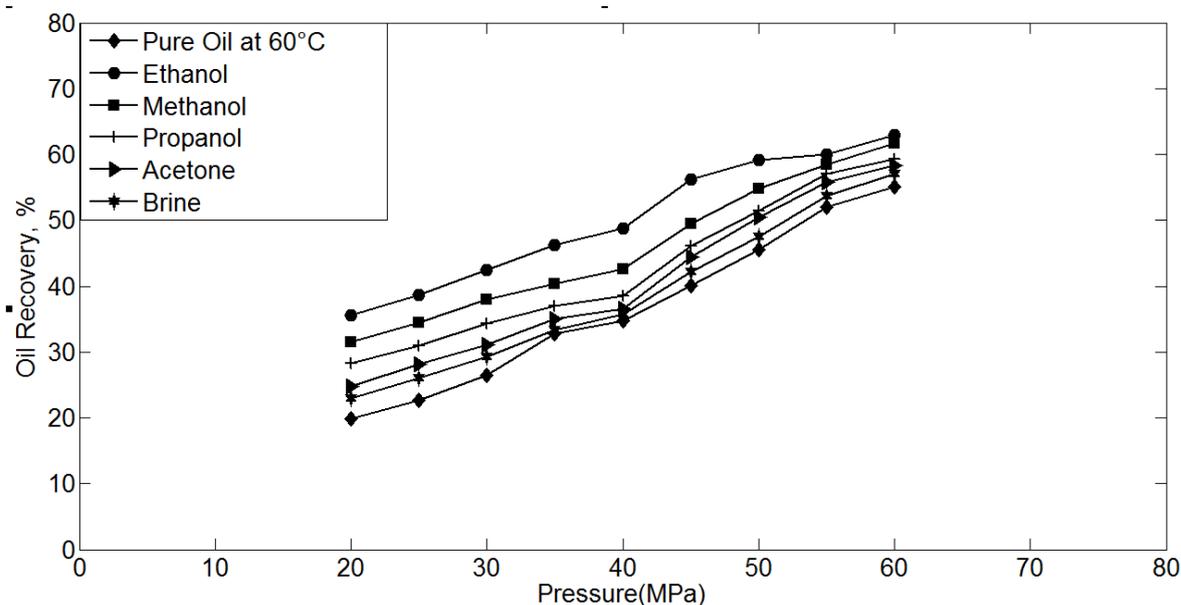


Figure 3.2 Percentage of oil recovery using co-solvents.

It was also observed from figure 3.2 that the extraction rate at early stages was constant, but as the extraction proceeded, it decreased gradually. It could be due to the solubility difference between low molecular and high molecular weight components.

The addition of co-solvents (ethanol, methanol, propanol, acetone, and brine) had modified the supercritical CO₂ behavior for crude oil extraction that is investigated here. The co-solvent addition increases the density of pure solvent (CO₂) and enlarges the interactions between solutes and modify CO₂ resulting appearance of new specific interactions. Similar results were obtained by other authors [12-14].

Figure 3.2 is clearly showing that percentage recovery of extracted oil has significantly increased with the addition of co-solvents at the same operating conditions.

It is investigated that extraction yields are approximately the same at pressure range of 55 and 60MPa using all co-solvents as compared to a pressure range of 20 to 50MPa as shown in figure 3.2. This is due to rapid increase in density of supercritical CO₂ in 55 and 60MPa as shown in figure 3.1 (b) resulting in high miscibility and solubility of solutes in solvent.

The effect of co-solvents in oil recovery is due to their participation of interaction between solutes and solvents with their capability to form hydrogen bonding specially. Table3.2 is showing the parameters of hydrogen bonding for a different co-solvents. Unfortunately, these parameters are not available for every co-solvent used in this extraction process.

Table1 3.2. Cosolvent Solvatochromic Parameters [15].

Cosolvent	π^*	α	β
Methanol	0.60	0.93	0.62
Ethanol	0.54	0.83	0.77
Acetone	0.71	0.06	0.48
Water (brine)	1.09	1.17	0.18

The β , α , and π^* represent the tendency of a solvent to be a hydrogen bond acceptor, to be a hydrogen bond donor, and the polarizability or polarity of a solvent, respectively. The values β , α , and π^* for brine are also not available, so these values are assumed to be close to water. These parameters contribute significant role to study interference of a co-solvent to form the specific interactions between solutes and solvent for the extraction process. Complete miscibility can occur only if the degree of hydrogen bonding is comparable for the components [16].

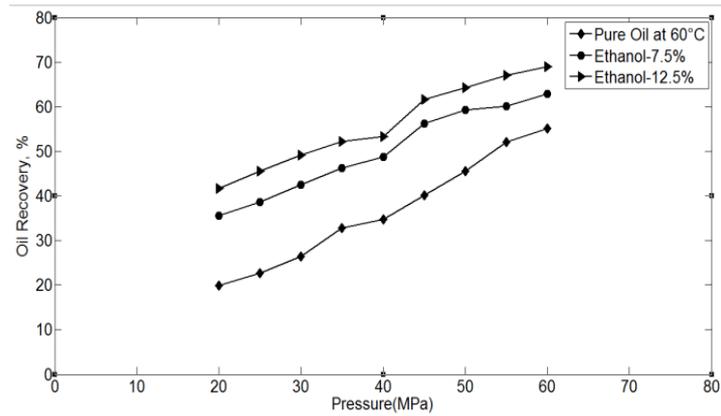
From Table 3.2, It is observed that methanol with $\alpha=0.93$ and $\beta=0.62$ is a good hydrogen bond donor while it is a worse hydrogen bond acceptor as compared to ethanol. Thus, ethanol has higher contribution in hydrogen bonding due to its high hydrogen bond acceptor capability that increases its co-solvent effect in crude oil extraction. While acetone with $\alpha=0.06$ and $\beta=0.48$ has less capability to form hydrogen bonding as compared to ethanol and methanol, but still it can better participate in hydrogen bonding than brine. On the other hand, brine with $\alpha=1.17$ and $\beta=0.18$ is the best hydrogen bond donor and the worse hydrogen bond acceptor that renders it less effective co-solvent in crude oil extraction as compared to methanol, ethanol and acetone. Brine as a co-solvent contributes its density and high polarity effects (solvent Dipole-solute induced Dipole and Dispersion forces) more prominent in the extraction process as compared to its hydrogen bonding.

Researchers [17, 18] have investigated that polar components with near about similar molecular sizes are extracted easier than non-polar components by addition of co-solvents. It is due to strong potential of polar components to form hydrogen bonding with co-solvents, and its magnitude depends upon hydrogen bonding nature of co-solvents (weak, moderate or strong hydrogen bonding capability). Co-solvent Solvatochromic Parameters [15] in table 3.2 are showing that larger polar components of crude oil will be extracted by ethanol as compared to methanol, acetone and brine. This is attributed to ability of ethanol to participate in hydrogen bonding, as prominent hydrogen bonding acceptor molecules as compared to methanol, acetone and brine, which increases the number of ways in which these molecules can interact with the polar components of crude oil.

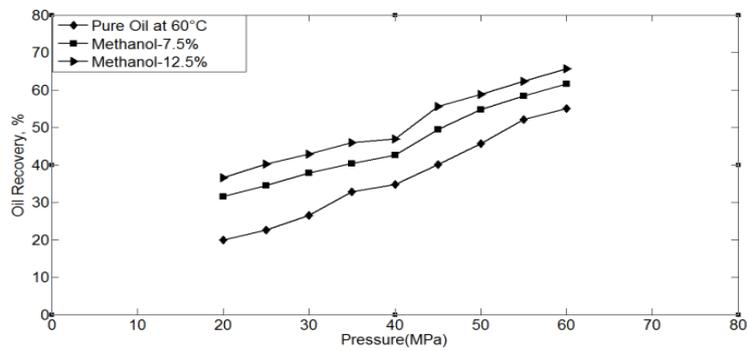
During experiments it was observed from color of extracted crude oil that higher molecular weight hydrocarbons were extracted using ethanol and methanol respectively at same operating conditions as co-solvents with supercritical CO₂ as compared to brine. This is due to the existence of preferential intermolecular forces among the different polar hydrocarbons and co-solvents. This greater affinity of ethanol and methanol is due to their hydrogen bonding ability with components of crude oil. But still brine can extract higher molecular weight hydrocarbons due to their high polarizability that interact with higher molecular weight hydrocarbons by dipole-induced dipole interaction. However, results indicated that the magnitude of this interaction is smaller than hydrogen bonding. Similar results were investigated by other authors [17, 18].

3.3 Extraction Yields Using Different Concentrations of Co-solvents with Supercritical CO₂

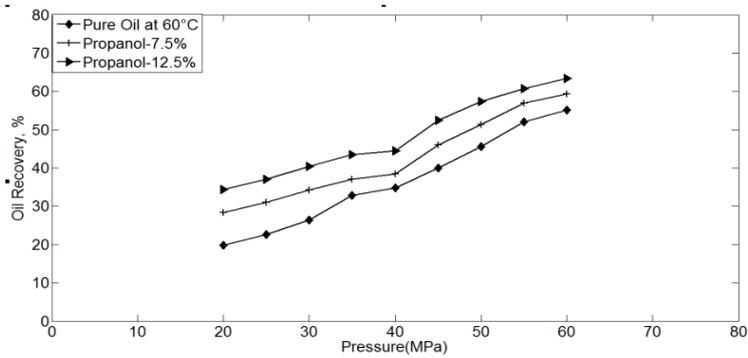
In section 3.2, different co-solvents with concentration of 7.5% were analyzed with their variable effect on recovery of extracted oil due to their participation of interaction between solutes of oil and Supercritical carbon dioxide. To study the effect of concentration of these co-solvents, Concentration of each co-solvent has increased to 12.5% instead of 7.5%.



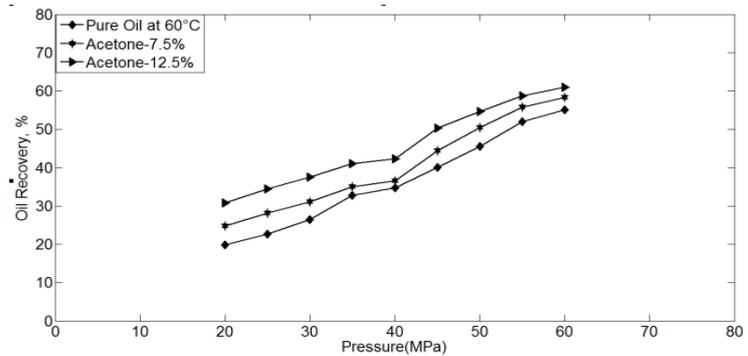
(a)



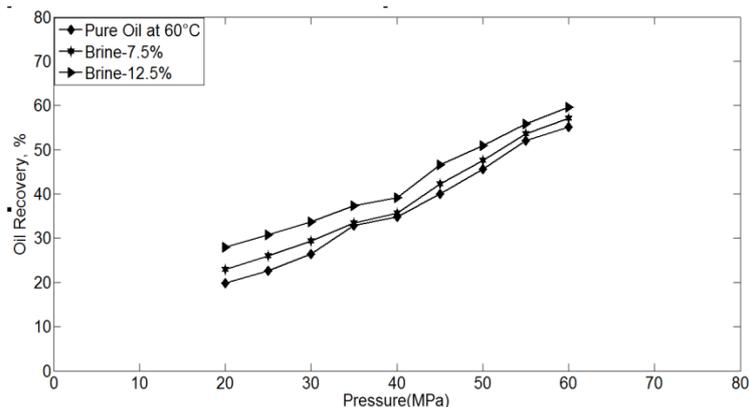
(b)



(c)



(d)



(e)

Figure 3.3 (a), (b), (c), (d), and (e), is showing comparison of oil recovery (%) using 7.5% and 12.5% concentration of ethanol, methanol, propanol, acetone and brine respectively with oil recovery (%) using pure supercritical CO₂ in pressure ranges of 20 to 60MPa at 60°C.

The percentage recovery of crude oil has increased with increasing the concentration of co-solvents as shown in the figures 3.3 3 (a), (b), (c), (d), and (e). In fact, increase of co-solvent concentration has increased the CO₂ density that has in turn enhanced its solvent power. Consequently, increased concentration of co-solvent has enlarged the interaction capability between the solutes of crude oil and CO₂ due to formation of new interaction. Similar results were concluded by other authors [12-14].

Consequently, this co-solvent's effect is due to specific interactions between their functional groups and those of crude oil components. The increase in solubility and miscibility due to co-solvent addition is mainly from the formation of specific interactions between the solutes and co-solvent molecules, so largest effect of co-solvent is found from ethanol due to its strongest interactions with solutes as compared to other used co-solvents (stronger hydrogen bonding).

4 Conclusions

Extraction of the crude oil by using super critical carbon dioxide with and without co-solvents were investigated. Crude Oil can be successfully extracted using supercritical carbon dioxide. Maximum Percentage recovery is achieved at 60MPa at 60°C with and without co-solvent. Results are showing that desired and valuable components (hydrocarbons) can also be extracted by selecting relevant operating conditions for this supercritical fluid extraction which is not achievable with the other solvent extraction process.

The operating conditions of this extraction process are taken as pressure range from 20 to 60MPa with interval of 5MPa at 60°C (constant temperature) for an interval of 20 minutes of extraction time after considering literature and similar works done by other authors. All co-solvents used in this extraction process have effectively participated to enhance the oil recovery, but ethanol has increased the extraction yields higher than all other co-solvents.

The effect of co-solvent is following this order;

Ethanol > Methanol > Propanol > Acetone > > Brine

This general trend is also explained qualitatively with physical property of co-solvents with their Solvatochromic Parameters following the same order as mentioned above. It is found that specific interactions between the molecules of solutes and solvent with a co-solvent contribute an important role in the extraction operation.

Extraction rate and yields have increased significantly with an increase in concentration of a co-solvent (from 7.5% to 12.5%) because a larger amount of a co-solvent has increased its capability to form a complex molecule clustering between the molecules of solvent and solutes (intermolecular interactions) resulting in larger local density than the bulk.

Solvatochromic parameters (β , α , and π^*) of co-solvents ranging from weak hydrogen bonding like brine to the strong hydrogen bonding like ethanol showed their physical significance in the extraction process.

It is also concluded that at high density conditions (high pressure) a co-solvent extracted larger amount of heavy (non-volatile) components than at low density conditions (low pressure) and vice versa, this is observed realistic from visual analysis of extracted oil.

References

- [1] Park, Hyong Seok ; Lee, Hee Jin ; Shin, Min Hye ; Lee, Kwang-Won ; Lee, Hojoung ; Kim, Young-Suk ; Kim, Kwang Ok ; Kim, Kyoung Heon, Effects of cosolvents on the definition of green tea of Supercritical CO₂, Food Chemistry, 2007, Vol.105 (3), p. 1011-1017.
- [2] F.M. Orr and J.J. Taber, Use of Carbon Dioxide in Enhanced Oil Recovery, Science, New Series, Vol. 224, No. 4649 (May 11,1984), pp 563-569.
- [3] A.A. Dehghan, R. Kharrat. M. H. Ghazanfari, Visualization and Quantification of Asphaltinic-Heavy Oil Displacement by Co-Solvent at Different Wettability Conditions, Petroleum Science and Technology, 28:2,176-189.
- [4] S.S.T. Ting, S.J. Macnaughton, D.L. Tomasko and N.R. Foster, Solubility of naproxen in supercritical carbon dioxide with and without cosolvents. Ind. Eng. Chem. Res., **32** (1993), p. 1471. | View Record in Scopus | Full Text via CrossRef | Cited By in Scopus (117).
- [5] Frye, S. L.; Yonker, C. R.; Kaikwarf, D. R.; Smith, R. D. Application of Solvatochromic Probes to Supercritical and Mixed Fluid Solvents. ACS Symp. Ser. 1987,329, 29.
- [6] <http://webbook.nist.gov/cgi/fluid.cgi?ID=C124389&Action=Page> (11-10-12).
- [7] Kwon & Yoon, 1996 Y.A. Kwon and S.K. Yoon, Concentration of medium chain fatty acids from coconut oil by supercritical CO₂ extraction. Foods and Biotechnology, 5 (1996), pp. 229–233.
- [8] Shishikura et al., 1986 A. Shishikura, K. Fujimoto, T. Kaneda, K. Arai and S. Saito, Modification of butter oil by extraction with supercritical carbon dioxide. Agricultural and Biological Chemistry, 50 (1986), pp. 1209–1215.
- [9] Yoon, 1993 J. Yoon, Extraction of EPA and DHA from tuna oil using supercritical carbon dioxide. Korean Journal of Food Science and Technology, 25 (1993), pp. 288–294.
- [10] Gu'c,lu'-U' stu'ndag', O.; Temelli, F. Correlating the solubility behavior of fatty acids, mono-, di-, and triglycerides, and fatty acid esters in supercritical carbon dioxide. Ind. Eng. Chem. Res. 2000, 39, 4756.
- [11] Gu'c,lu'-U' stu'ndag', O.; Temelli, F. Solubility behavior of ternary systems of lipids in supercritical carbon dioxide. J. Supercrit. Fluids 2006, 38, 275.
- [12] Gu'c,lu'-U' stu'ndag', O.; Temelli, F. Solubility behavior of ternary Systems of lipids, cosolvents and supercritical carbon dioxide and processing aspects. J. Supercrit. Fluids 2005, 36, 1
- [13] Kim, S.; Johnston, K. P. Effects of supercritical solvents on the rates of homogeneous chemical reactions. ACS Symp. Ser. 1986, 42, 329.

[14] Foster, N. R.; Singh, S. L.; Yun, J.; Tomasko, D. L.; Macnaughton, S. J. Polar and nonpolar cosolvent effects on the solubility of cholesterol in supercritical fluids. *Ind. Eng. Chem. Res.* 1993, *32*, 2849.

[15] Kamlet, M., J.; Abboud, J.-L.M.; Abraham, M.H.; Taft, R. W. Linear Solution Energy Relationships. π^* , α and β , and Some Methods for Simplifying the Generalized Solvatochromic Equation. *J. Org. Chem.* 1983, *48*, 2877.

[16] Barton, A. F. M. *CRC Handbook of solubility parameters and other cohesion parameters*; CRC Press, Inc.: Boca Raton, FL, 1985, pp249.

[17] Yates, R. A.; Caldwell, J. D. Regeneration of oils used for deep frying: a comparison of active filter aids. *J. Am. Oil Chem. Soc.* 1993, *70* (5), 507.

[18] Takeoka, G. R.; Full, G. H.; Dao, L. T. Effect of heating on the characteristics and chemical composition of selected frying oils and fats. *J. Agric. Food Chem.* 1997, *45*.