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Sub and Supercritical Decontamination of Oil-Based Drill Cuttings: A Review

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Abstract

Drill mud is a fluid which is used in oil extraction industries in order to cool and lubricate the drill bit. Due to containing numerous toxic components, it is considered as a hazardous waste which must be treated before discharging to the environment. Current separation techniques for drill cutting treatment can be categorized into three main categories of physical (dewatering), physicochemical (solvent extraction, surfactant enhanced washing, and supercritical CO_2 extraction), and thermal methods (desorption and microwave heating). In this research, the effectiveness of superheated steam extraction for drill mud recovery is studied. Super-critical fluid extraction is an innovative process in the field of contaminated soil treatment. Extraction with super-critical fluid is a simple and rapid extraction process which uses supercritical fluids as solvents. In order to enhance the decomposition percentage, supercritical extraction is accompanied by oxidation process using H_2O_2. Using superheated steam extraction process at 2.3MPa and 225 °C, 78.56% and 83.09% of total organic carbon were removed from the drill mud sample. In the combined system of supercritical extraction and oxidation with H_2O_2, more than 99.9% of polycyclic hydrocarbons in the drill mud mixture were decomposed. It shows that combination of supercritical extraction with an advanced oxidation process can significantly enhance the efficiency of the remediation process. The great advantage of this hybrid process is being eco-friendly due to using water as the solvent in the extraction process.

Keywords: Oil contaminated soil, Drill mud, polycyclic aromatic hydrocarbons, Supercritical extraction, advanced oxidation.

Introduction

Drill fluids, also called drill mud are specific liquids used in drilling operations for oil and gas exploration and extraction which performs several drilling functions(Chen et al., 2019; B. Ma, Wang, Ni, & Wang, 2019), including cooling and lubricating the drill bits, balancing the down-hole formation pressures, stabilizing well bores, cleaning the borehole bottom, and transporting rock cuttings to the surface(Zhang, He, Li, Fan, & Li, 2019) Most of the drill fluid in the returned

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mixtures is separated for reuse, while a small amount of drill fluid and rock cuttings forms the drill waste, which is generally called as drill cuttings (Ball, Stewart, & Schliephake, 2012). Drilling fluids can be broadly categorized into three main groups of water-based, synthetic-based, and oil-based depending on the base fluids (Ball et al., 2012). Among these, in the oil-based fluids, heavy oil (such as diesel, white oil, and crude oil) is used as the base fluids, which are costly and hazardous, but deliver excellent drilling performance(Chen et al., 2018).

Large quantities of oil-based drill cuttings (OBDC) are produced worldwide and is a type of hazardous organic waste with toxic, mutagenic, and carcinogenic properties (B. Ma et al., 2019; J. Ma et al., 2016). Considering the challenges of safe treatment and disposal of drill cuttings, the combination of two or more technologies instead of a single-step method is currently favorable (Kogbara, Ayotamuno, Onuomah, Ehio, & Damka, 2016; J. Ma et al., 2016; Yan, Lu, Guan, Zhang, & Zhang, 2011). The treatment process consists of a group of primary pretreatment technologies and focuses on separating the organic contaminants from the drill cuttings (Chen et al., 2017). Current separation techniques for drill cutting treatment can be categorized into three main categories of physical (dewatering (Committee, 2011)), physiochemical (solvent extraction (Ball et al., 2012), surfactant enhanced washing (J. Ma et al., 2016), and supercritical CO_2 extraction (Khanpour, Sheikhi-Kouhsar, Esmaeilzadeh, & Mowla, 2014)), and thermal methods (desorption (Okparanma, Araka, Ayotamuno, & Mouazen, 2018) and microwave heating (Pereira et al., 2014; Robinson et al., 2009; Shang, Snape, Kingman, & Robinson, 2006)).

Water in high temperatures and under high pressures has attracted the attentions of researchers in various fields because it is an eco-friendly green material with interesting properties (Akizuki, Fujii, Hayashi, & Oshima, 2014; He et al., 2018; Machida, Takesue, & Smith Jr, 2011; Teo, Tan, Yong, Hew, & Ong, 2010). Interestingly, the dielectric constant (ε) of liquid water (subcritical water) will decrease noticeably with an increase in temperature, and within the range of 150 to 300°C, it reaches to the values similar to common non-polar organic solvents at room temperature, such as methanol, ethanol, acetone, and dimethyl formamide (DMF) (He et al., 2018; Khanjari, Eikani, & Rowshanzamir, 2016). Furthermore, the ε of gaseous water (superheated steam) is as low as 1 to 4 in the same temperature range and is close to that of n-hexane at 20 °C. The intensive changes in ε offers this possibility that water at high temperatures and pressures may behave as a natural non-polar solvent. It should be noted that superheated steam has been utilized to remove volatile components from lignite (Hoadley et al., 2015), sludge (Tahmasebi et al., 2012), wood, food (Mujumdar, 2014), and to extract essential oils from raw materials (Liu, Zang, Xu, Wang, & Li, 2017; Rouatbi, Duquenoy, & Giampaoli, 2007). In addition, subcritical water has been successfully applied for the remediation of soils contaminated with various complex organic pollutants, such as polycyclic aromatic hydrocarbons (Khanjari et al., 2016), 2, 4, 6 Trinitrotoluene (Islam, Shin, Jo, & Park, 2015), pesticides (Islam, Jo, Jung, & Park, 2013), diesel (Islam, Park, & Park, 2015) and phthalate esters (Chang, Shen, Yang, & Wu, 2011). In this study, the potential of using supercritical fluids for cleaning drilling mud has been investigated and the optimum extraction conditions have been presented and various methods and techniques for this method implementation have been reported.

Material and methods

Several methods have been used to extract contamination from solid matrices. Essentially, they can be classified into two subcategories(Chen et al., 2019):

(a) Processes using pressurized fluids; (b) Atmospheric pressure processes.

Processes using pressurized fluids are as follows:

Accelerated solvent Extraction (ASE)

ASE is a relatively new extraction technique in which organic solvents are used at certain temperatures and pressures. ASE provides faster extraction and needs less solvent volume in compare with classic extraction methods such as Soxhlet extraction. Generally, the pressure and temperature of the extraction, which are the practical parameters of the ASE, affect its efficiency. In addition, the sample matrix also has an effect on the extraction efficiency(Hawthorne, Grabanski, Martin, & Miller, 2000).

Subcritical water extraction (SCWE)

This process is based on the polarization change in water, which occurs during the temperature raise from ambient to subcritical temperatures (374 ° C). To maintain water in liquid state, the system must operate with normal pressures of 50 to 200 bar. Increasing the temperature to higher than 200 ° C reduces the water dielectric constant. Similarly, the temperature has a similar effect on two other parameters, surface tension and viscosity(Yang, Belghazi, Lagadec, Miller, & Hawthorne, 1998) . Considering the benefits of hot water properties, the solubility of some PAHs increases up to a maximum of five times.

Supercritical fluid extraction (SFE)

Supercritical fluid extraction (SFE) is listed as an alternative approach to conventional extraction techniques. A supercritical fluid (SCF) is identified as any combination at temperature and pressure above the critical values (above the critical point), in this area, the fluid properties are between the liquid and gas state. For instance, the fluid density is nearer to liquid phase density, and the viscosity and permeability of the fluid is more similar to the gas phase. Supercritical fluids have zero surface tension and easily enter the solid matrix(Goodarznia & Esmaeilzadeh, 2006; Street & Guigard, 2009). In a supercritical state, the fluids are highly sensitive to the slightest changes in temperature and pressure, thus it is probable that in a setting of pressure and temperature, the liquid absorbs the pollutant and in another setting this fluid disposes the same pollutant(Tomasko, Macnaughton, Foster, & Eckert, 1995). So, pressure and temperature are the two major factors in this process. Supercritical fluid extraction depends on the density of the fluid, which, in turn, can be manipulated by controlling the pressure and temperature of the system. In supercritical fluid extraction method, the extracted contaminants are first dissolved in supercritical solvent, and then these contaminants are separated from the supercritical solvent by a simple change in pressure and temperature conditions using a separation process. Carbon dioxide is the most common SCF, due to its low temperature (31.1 ° C, 73.8 times), low cost and non-toxicity(Sahena et al., 2009)

Supercritical fluid extraction (SCFE), on the other hand, uses fluid which is heated and pressurized above the critical point, otherwise known as supercritical fluid (SCF)(Gan, Lau, & Ng, 2009). Due to the high temperatures and pressures, SCF exhibits gaseous–liquid properties such as liquid-like density, high diffusivity, low viscosity and no surface tension(Kiran, Debenedetti, & Peters, 2012). The efficiency of SCFE depends greatly on the solubility and mass transfer of the contaminant into the SCF (Morselli et al., 1999). The most commonly used fluid for SCFE is carbon dioxide due to its non-polar nature and liquid solubility characteristics. It is also inexpensive, non-toxic, and vastly available and owns a moderate critical pressure and temperature. Other SCFs considered include propane and butane. While the latter SCFs exhibit excellent oil and grease removal from soil and sludge, these fluids are highly flammable that make their use potentially hazardous to human health(Al-Marzouqi, Zekri, Jobe, & Dowaidar, 2007; Low & Duffy, 1995) investigated the use of CO2 for the extraction of crude oil from contaminated soil under pressures ranging from 80– 120 bars for temperatures between 40–60 °C, and 200–300 bars for temperatures between 100–140 °C.

Oxidation in extraction process

In any case, the extraction stage as a single treatment technology is not the definitive solution for PAH contaminated soils. However, the degradation of polycyclic aromatic hydrocarbons makes it extremely important. Bioremediation of infected sites may be an efficient and economically attractive option as a single technology. However, the kinetics of the process is very slow due to the limited range of soil contaminant molecules in most cases. Similarly, soil bio improvisation is considered (Laitinen, Michaux, & Aaltonen, 1994; Marcus, 2019; Sánchez-Camargo, Parada-Alonso, Ibáñez, & Cifuentes, 2019). Advantages and Disadvantages of chemical oxidation are listed below. (Table 1).

Table 1. Advantages and Disadvantages of chemical oxidation

Advantages	**Disadvantages**
Short cleaning time	This technology is not economic compared to environmental processes
conditions can be changed	Need recycling organic solvents
Chemical oxidation is relatively insensitive to external changes	reduction of chemical reactions at low temperature and pressure
Chemical oxidation can be combined with the extraction process	due to the amount of chemicals required, its not be economical

Apparatus and procedure

Apparatus and procedure of superheated steam extraction:

The experimental apparatus for superheated steam extraction (SHSE) is illustrated in Figure. 1. The extraction vessel with a volume of 1960 ml was made of 316 stainless steel and designed for a maximum pressure and temperature of 20 MPa and 350 °C, respectively. A 1 kW heater was used to maintain the temperature at the desired value during extraction. The vessel was completely surrounded by an insulation jacket, except for the top surface. The inner top (T1) and bottom (T2) temperatures of the extraction vessel were measured by two K-type thermocouples, and the interior pressure was determined using a universal pressure transmitter with a range of 0–25 MPa (DG1300-PJ-1-2-25, Guangzhou Senex Instrument Ltd., China).

A sample boat made of 316 L SS wire netting with a pore size of 20 μm was fixed in the middle of the extraction vessel. The net was designed in a way that subcritical water and liquid products could pass through but the solid products are intercepted. A preheater with a maximum power of 3 kW was used to preheat the water from room temperature to the target temperature, which was automatically adjusted with a temperature controller. Water was delivered by a high-pressure pump (LC-3060, Chin-Fine Technology, China) with a flow range of 0.01–49.99

ml.min^{-1}. All the tubes were made of 316 SS, and the inner diameters of the inlet and outlet were 1.5 mm and 4 mm, respectively. The system pressure was controlled by a back-pressure valve.

First, an OBDC sample of 50.00±1.00 g (min) was loaded in the sample boat. Valve 2 was opened and a diaphragm vacuum pump (C410, Wiggens) with a vacuum of −0.07 MPa removed the non-condensable gases (air). Then, all valves were closed, and the heater and pre-heater were turned on to warm up the extraction vessel to the target temperature (T). Next step was opening of valve 1 to pump water at a pre-set flow rate (F). After a certain amount of water (V0 in Table 2) was delivered, the desired pressure would be achieved. The steam was condensed to liquid water and gathered at the bottom of the extraction vessel. The liquid products were seeped out and collected every 5 minutes. The effects of different pressures (0.6–5.5MPa), temperatures (175–225 °C), and water flow rates (2–8 ml min-1) on the removal efficiency were studied in a semi-continuous laboratory setup.

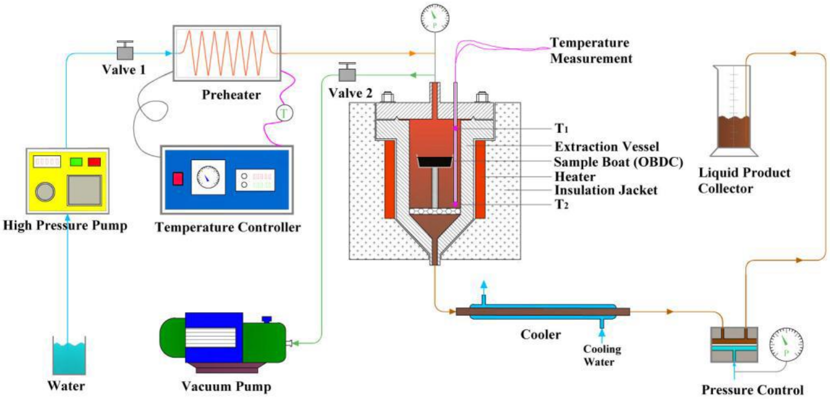

Figure 1. Schematic of the experimental apparatus superheated steam extraction

Supercritical water oxidation of polychlorinated biphenyls using hydrogen peroxide:

Although production of polychlorinated biphenyls (PCBs) is prohibited due to high toxicity, a large amount of them had been produced and are still in use. Despite various methods such as molten salt (Harry, 1985), incineration (Schlegel, 1988), chemical transformation (Lauch, 1989) and plasma treatment (Hollis, 1983) for destruction of PCBs, no significantly satisfactory results have been obtained. Recently, supercritical water oxidation (SCWO) has been studied as an advantageous oxidation process for treatment of hazardous wastes. PCBs can be easily mixed with supercritical water (SCW) due to the static dielectric coefficient of SCW which is close to the dielectric coefficients of non-polar organic solvents such as n-hexane, and there is no possibility of interfacial mass transfer that limits the reaction rate. Operating at supercritical conditions leads to a homogeneous reaction mixture in which organics, water, and oxygen can make a single phase. In these studies, oxygen played a dominant role as oxidant. The SCWO processes have been generally performed in higher temperature ranges (873-923 K). A reduction in the reaction temperature due to changing the oxidant may result in a reduction in energy consumption as well as more favorable choice of material for reactors in the SCWO system.

In order to achieve the destruction of PCBs in SCWO at lower temperatures than 873-923 K in the presence of oxygen (Thomason & MODELL, 1984), hydrogen peroxide is proposed as an alternative oxidant in the SCWO system (Hatakeda, Ikushima, Ito, Saito, & Sato, 1995). In the supercritical state, both PCBs and H_2O_2 have complete miscibility with water, and a homogenous fluid phase containing PCBs, H_2O_2 and water can act as the optimum medium for decomposition.

Total petroleum hydrocarbon (TPH) recovery using subcritical water extraction technology:

In this study, total hydrocarbon oil (TPH) present in soil is considered as the target pollutant(Islam, Jung, Jung, & Park, 2017). The concentration of oil in the contaminated soil was 4.08% of its weight (40832 mg / kg). The pH of the contaminated soil was 8 and the average moisture content was 26%. Experimental condition was as follows: «extraction time 90 minutes at 260 ° C in absolute pressure of 8 MPa and water-to-soil ratio of 3: 1». After extraction, the oil removal efficiency was about 86% of the remaining soil and recycled almost 39% of the oil. The equipment can operate up to 15 MPa and 300 ° C conditions.

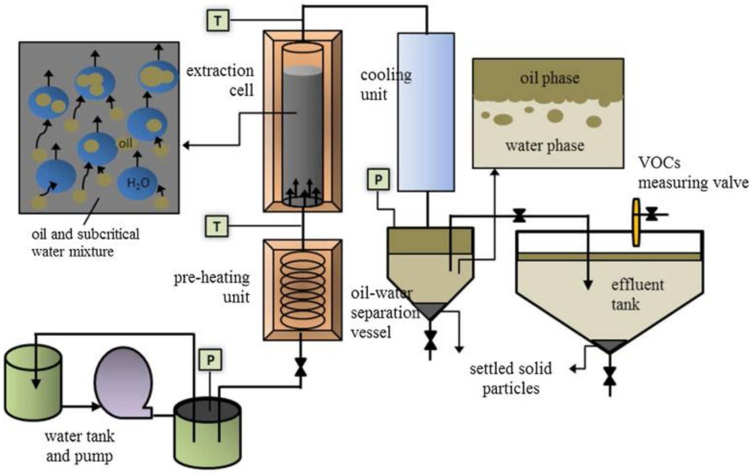

Figure 2. An overview of the SWE device and the dissolving mechanism and subcritical oil separation

A specific amount of water (4.5 liters, water to soil ratio being 3: 1) was transferred to the extraction chamber. The most suitable temperature for removing oil from soil was 260 ° C in this study. The reactor pressure was kept above the vapor pressure (at the respective temperature) being 8 MPa and the water was in liquid state and its extraction time was 90 minutes. The water was quickly pumped at a rate of 15 ml/min until the desired temperature reaches the extraction cell (60 minutes). Afterwards, the flow rate was increased to 40 ml/min and kept constant during the experiment (90 minutes). After full water extraction, the remaining TPH in the soil was measured by the remaining soil extraction with dichloromethane. The oil and waste water phases achieved from the oil separation tank and the waste water reservoir were separated by a separation funnel and then the oil separated from the oil layer was measured and analyzed as recycled oil. The concentration of TPH in the oil layer and the aqueous phase were analyzed.

Figure 3. Oil samples and covered oil samples.

Restoration of polycyclic aromatic hydrocarbons from soil using supercritical water extraction: The extraction study (SWE) aims to remove polycyclic aromatic hydrocarbons (PAHs) including naphthalene, phenanthrene, anthracene and pyrene from contaminated soil(Khanjari et al., 2016). Prior to using distilled water, is cleaned for 30 minutes with nitrogen to remove its oxygen.

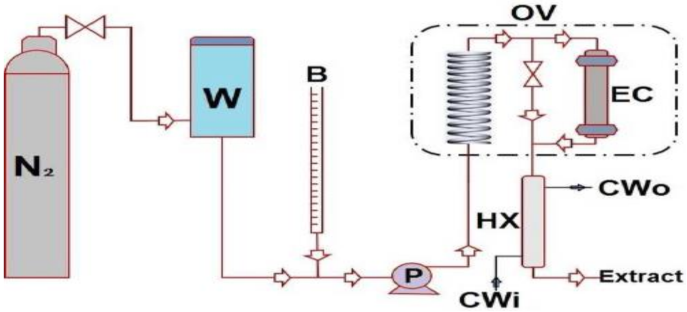

Figure 4. The supercritical water extraction system diagram: OV: furnace; HX: heat exchanger; EC extraction chamber; W: water storage; CWi: cooling water; CWo cooling the water; B: Burette

The pump operates at constant pressure. The steel extraction chamber is filled with 5 g of contaminated soil and water. The extraction chamber is located in the oven. The pump is switched on and the pressure is raised up to 20 bar. Then, the pump is switched off and oven temperature is increased to the desired value, the pump is switched on again and the flow is adjusted at the required rate. The water passes through the top-to-bottom in extraction chamber. When the temperature of the extraction chamber reaches the desired point, the extraction time begins. At the end of the extraction time, the oven and the pump are turned off and the pressure is adjusted to atmospheric pressure. For UV analysis, pure soil is collected to verify the efficiency of SWE.

Result and discussion

The results indicated that the temperature was the most important parameter, followed by water flow rate and pressure. The OBDC characteristics significantly affected the removal efficiency as well. Both the superheated steam and subcritical water removed not only all extractable organics but also some other compounds. Under the optimal conditions of 2.3MPa, 225 °C and 6 ml min^{-1}, 78.56% and 83.09% of total organic carbon were removed. After treatment, the level of hazardous components in OBDC was significantly decreased as their oil content has dropped to almost zero and the OBDC could be categorized as a non-hazardous waste. Further analysis confirmed that this method possessed the advantages of solvent extraction (excellent solubility) and thermal desorption (high temperature). The results of X-ray diffraction (XRD) and scanning electron microscopy (SEM) indicated that the microstructure and crystalline structure of inorganics in OBDC were not obviously changed during the treatment. As a result, this method can be considered as a new potential separation technique for organics removal from OBDC.

Several researchers have attempted to interpret the oxidation mechanisms in which oxygen in supercritical water is involved using the knowledge of gas-phase combustion processes for hydrocarbons (Rofer & Streit, 1989). The initiation reaction for the gas-phase oxidation of hydrocarbons involving oxygen is known to be the hydrogen abstraction from hydrocarbons by oxygen gas to produce hydrogen-deficient and hydro-peroxide radicals (Hatakeda, Ikushima, Sato, Aizawa, & Saito, 1999; Westbrook & Dryer, 1984). However, when hydrogen peroxide is used as the oxidant, a different oxidation mechanism is expected, especially in the chain initiation step where hydrogen peroxide thermally decomposes into the hydroxyl radical ($O\overset{o}{H}$). The initiation by hydroxyl radical is expected to be much faster than oxygen in typical wet air oxidation conditions. Hydrogen peroxide is used as an oxidant for wet oxidation (Gulyas, Von Bismarck, & Hemmerling, 1995; Pignatello & Chapa, 1994) and as an oxygen resource with SCW (Thammanayakatip, Oshima, & Koda, 1998). However, there is the question whether hydrogen peroxide can remain as a stronger oxidant than oxygen even in supercritical water conditions.

Recent works has reported the decomposition of hydrogen peroxide under SCW (Croiset, Rice, & Hanush, 1997). On the other hand, the use of hydrogen peroxide for oxidation of acetic acid and dichlorophenol using batch reactor system under SCW condition is reported as well (Lee, Gloyna, & Li, 1990). The purpose of using H_2O_2 as an oxidant in this work is to produce $O\overset{o}{H}$ radical instead of $H\overset{o}{O}_2$ radical in the reactor. $O\overset{o}{H}$ is extremely reactive in comparison to $H\overset{o}{O}_2$ radical which is generated in SCWO with oxygen (Gopalan & Savage, 1994).

In this experiment H_2O_2 was introduced immediately before the flow reactor, and is expected to be converted into the unstable $O\overset{o}{H}$ before its decomposition into oxygen. Although these two radicals are be present in the mixture, $O\overset{o}{H}$ is likely to be more important in SCWO using H_2O_2 because of its relative abundance and capability in complete decomposition of PCBs at a lower temperature of 673 K. In spite of hydrogen peroxide's higher price in comparison to chap oxygen, it has noticeable advantages some of which are as follows:

(1) Energy consumed for decomposing toxic compounds can be greatly reduced, leading to a reduction in cost.

(2) Any metals are readily corroded at higher temperatures than 773 K in supercritical water, particularly in the presence of Cl^-, and special materials such as ceramics should be used for the SCWO reactor, resulting in great increase in cost.

The purpose of this work is to experimentally evaluate the complete conversion of PCBs to carbon dioxide, water, and hydrochloric acid (Figure. 5) in SCWO conditions in a batch reactor using hydrogen peroxide and to establish the conditions which best fitted to the flow-reactor system.

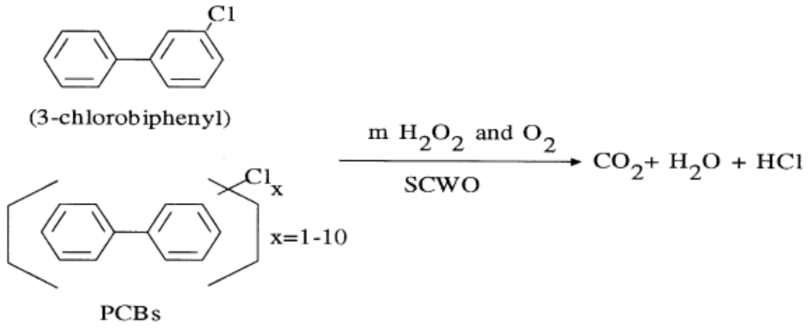

Figure 5. Reaction of PCBs by SCWO

The SWE method presented in this study is known as a desirable candidate for the treatment of solid fatty wastes. After 90 minutes of extraction at 260 ° C in a water-to-soil ratio of 3: 1, the removal efficiency was about 86% and oil recovery was about 39%. After 90 minutes extraction at 260 ° C in a water-to-soil ratio of 3:1, the Recovered oil had a higher level of density and specific gravity than commercial crude and had similar levels of elements and amounts of heat. After the extraction, there was a significant decrease in TPH content in soil residues. In summary, this research has found notable results in the field of oil removal and recovery, and indicated the feasibility of using the SWE technique, which can be an effective and plausible way to restrain hazardous, highly moist, contaminated soils.

Although supercritical CO_2 could only remove up to 4% of crude oil in soil at pressures of 80 bars in the temperature range studied, its efficiency was seen to increase with pressure, reaching up to a maximum of 72.4% at 300 bars and 100 °C. Other documented works on the use of supercritical CO_2 for the remediation of petroleum hydrocarbons include the study by Morselli et al. (1999) (Morselli et al., 1999). This study reported that with an addition of 5 v/v % of acetone to supercritical CO_2 at 80 °C and pressure of 227 atm, the removal efficiency of crude oil from soil increased from 60% to 75% without the acetone addition. The results suggest that the acetone exerts a swelling action on the soil which helps to pry open the interlayer's of the soil structure. Likewise, Geranmayeh et al. (2012) also used carbon dioxide for the extraction of oil from contaminated soil from the Pazanan II production unit site in Gachsaran, Iran. In this study, the optimum operating parameters such as pressure, temperature, flow rate and duration were obtained to optimize the removal of the contaminant (Geranmayeh, Mowla, Rajaei, Esmaeilzadeh, & Kaljahi, 2012).

PCB removal efficiency strongly depends on the water temperature. The effect of the two parameters of water and extraction time in purification efficiency was investigated. The water temperature varied from 100 to 180 ° C and the extraction time was from 5 to 20 minutes. It was found that more than 99.9% of PCBs decomposition can be achieved by supercritical water under any conditions when a certain amount of H_2O_2 is added by the stoichiometric demand. Under the supercritical water conditions, hydrogen peroxide was found to be significantly efficient. The conversion of 3-PCB and KC-300 exceeded 99% by using hydrogen peroxide at the temperature of 673 K(Islam et al., 2017).

Reza Khanpour et al investigated the supercritical extraction process for removal of contaminants from the drilling mud, in this regards effect of different parameters including extraction temperature (313–338 K) and pressure (100–200 bar), flow rate of CO2 (0.05–0.36 cm3/s) and static time (20–130 min) on the removal of contaminations from drilling mud was examined. The highest removal efficiency were obtained at temperature and pressure of 333 K and 180 bar, respectively, flow rate of lower than 0.1 cm3/s and the static time of 110 min(Khanpour et al., 2014). The supercritical water oxidation (SCWO) of OBDC was investigated by Zhong Chen et al. in a batch reactor under the conditions of various conditions. The total organic carbon (TOC) removal efficiency was reached up to 89.2% within 10 min at 500 °C(Chen et al., 2017). Also the investigated the superheated steam decontamination of OBDC at near-saturation pressure. The effects of different pressures (0.6–5.5 MPa), temperatures (175–225 °C), and water flow rates (2–8 ml min−1) on the removal efficiency were studied. Temperature was the most effective parameter. At optimal condition of 2.3 MPa, 225 °C, and 6 ml min−1, 83.09% of TOC was removed (Chen et al., 2018). Supercritical fluid extraction of OBDC results indicate that SC CO2 supercritical extraction was reduced the OBDC contamination to a level that allow offshore disposal(Eldridge, 1996). C.G. Street et al, using supercritical carbon dioxide to remove the OBDC. Extraction efficiencies as high as 98% have been observed. And the hydrocarbons weren't changed in extraction process and could be recovered and reused(Street & Guigard, 2009). Supercritical carbon dioxide extraction of South Pars gas field OBDC has been investigated by Goodarznia et al at a range of temperatures (55 to 79.5 °C), and over a pressure range of 160 to 220 bars. Pressure of 200 bars and temperature of 60 °C was obtained optimum condition(Goodarznia & Esmaeilzadeh, 2006). Esmaeilzadeh was inverstigated the solubility of OBDC in supercritical carbon dioxide at 200 bar pressure, over a temperature range of 55–79.5 °C. The SAFT model with respect to the PR and SRK EOS models is good approximation for the solubility of OBDC by SC CO2 (Esmaeilzadeh, Goodarznia, & Daneshi, 2008). In the table 2, a summary of the methods used Processes using pressurized fluids to extract pollutants is presented.

Table 2. Summary of the methods used Processes using pressurized fluids for extraction of pollutants

Method	Experiments	Result	Ref
Pressurized hot water extraction (PHWE method) from phthalic anhydride and benzoic acid from petrochemical waste using supercritical liquid extruder and a central composite design for optimization	pressure (220-260 bar), temperature (160-140 ° C), dynamic time (5 to 45 minutes) and flow velocity (0.2-1 mL / min)	At 140 ° C, the pressure, extraction time and solvent flow rate were 118 bar, 27 min, and 0.2 ml / min respectively for 100% yield of PA and these values are 118 bar, 29 min and 0.9 ml / min for 98% extraction of BA. Moreover, the maximum selection of PHWE at 100 ° C was obtained as 220 bar, 5 min, and 0.2 ml / min.	(Kamali & Ghaziaskar, 2010)
Hot water / steam Extraction under pressure from polychlorinated dibenzofurans and naphthalene from industrial soil	Water at 10 or 50 atmospheres (excluding 50 atmospheres and 200 °C) is in the gaseous phase and in 250 atmospheres at all temperatures, except at 400 ° C.	The optimal extraction conditions for these compounds were found to be between 300 and 350 ° C. The highest extraction efficiency was achieved in gas phase at 50 atmospheres. With Steam at 300 ° C and 50 horsepower extraction	(van Bavel, Rappe, Hartonen, & Riekkola, 1999)
Restoration of PAH contaminated soils by extraction using subcritical water	Water temperature from 100 to 300 ° C, extraction time from 15 to 60 minutes and flow rate in the range of 0.5 to 2.0 ml / min	The results showed that extraction efficiency depends mainly on the temperature and extraction time. There is also a significant dependence on flow rates. More than 95% of extracted phenanthrene, fluoranthene, and pyrene from contaminated soil is done at 300 ° C for 30 minutes and 250 ° C for 60 minutes with constant pressure of 100 bar.	(Islam, Jo, & Park, 2012)
Extraction of selected categories of polar, relatively polar and nonpolar magmatic wastes from hydrocarbon wastes using subcritical water	Little extraction of high-molecular-weight alcohols was achieved only with super-heated steam (250 and 300 ° C at 5 atmospheres).	the results indicate the recovery of all target analytes by extraction of subcritical waters (generally 90 to 120 percent).	(Yang, Hawthorne, & Miller, 1997)
Ability to recover oil from contaminated soil at oil leakage site using subcritical water extraction technology	extraction time 90 minutes at 260 ° C in absolute pressure of 8 MPa and water-to-soil ratio of 3: 1	the oil removal efficiency was about 86% of the remaining soil and recycled almost 39% of the oil	(Islam et al., 2017)
Correction of oil-contaminated soils using Dynamic SCWE and Static-dynamic SCWE methods	Subcritical water extraction (SCWE) is a green technology that uses heated water (100 to 374 ° C) at a pressure above 22.1 MPa to maintain it in liquid form.	The study indicates that the static-dynamic mode has a significant effect on extraction efficiency. The time and volume required for a dynamic static mode is much lower than those required for dynamic mode.	(Islam, Jo, & Park, 2014)

Table 2. (continued)

Method	**Experiments**	**Result**	**Ref**
Pilot Scale subcritical water use to correct Soil Contaminated with Poly-aromatic Hydrocarbons and Pesticides	Operation at various temperatures from 100 to 300 ° C, at a maximum pressure of 105 bar and in a water flow of 0.1 to 1 liter per minute	The extraction of PAH (2200 ppm total PAHs including naphthalene to benzoperylene) from contaminated soil with 275 ° C water, with high and low molecular weights achieve a detectable level (less than 0.5 ppm) in less than 35 minutes.	(Lagadec, Miller, Lilke, & Hawthorne, 2000) .
Hot water extraction at wet oxidation site: The kinetics of removing PAHs from the soil	A small-scale semi-continuous extraction with and without oxidation solution as the removal factor in the PAHs contaminated soil using subcritical water	in combined extraction and oxidation experiments, the remaining PAHs in the soil after the experiments were almost undetectable. In the combination of extraction and oxidation, after the first 30 minutes of experimentation, no PAH is detected in the liquid phase.	(Dadkhah & Akgerman, 2006)
Restoration of polycyclic aromatic hydrocarbons from soil using supercritical water extraction	To remove PAHs including naphthalene, phenanthrene, anthracene and pyrene from contaminated soil	the removal efficiency strongly depends on the water temperature. The water temperature varied from 100 to 180 ° C and the extraction time was from 5 to 20 minutes.	(Khanjari et al., 2016).
Single-stage correction of soil contaminated by sludge using supercritical water extraction along with oxidation	When the temperature and pressure in the oxidation vessel reaches the desired supercritical conditions, 400 ° C and 23.4 MPa, the solvent flow rate reduces. Oxygen velocity varied at three levels of 0.09, 0.5 and 1 cm3 / min.	that supercritical water oxidation was able to eliminate 99.5% of hydrocarbon contaminants present in the soil. The supercritical water oxidation of PAH-infected soil is a suitable alternative to conventional processes as a full one stage treatment.	(Kocher, Azzam, & Lee, 1995).
Destruction of PAHs from the soil using hot pressurized water extraction along with supercritical water oxidation	Oven 2: (385 ° C or 425 ° C) (pump 2: v = 1.0 or 2.0 ml / min) pump 1 (v = 1.0 ml / min) oven 1: 200 or 300 ° C extraction time: 20 or 40 minutes	The extraction efficiency increases with temperature and time; the best results were obtained at 300 ° C with 40 minute extraction time. In the oxidation stage, the conversion of PAHs increases with the reaction time and oxidant concentration.	(Dadkhah & Akgerman, 2002)
Extraction of polychlorinated biphenyls from soil under sub and supercritical general conditions	The water is placed in contact with the sample for 30 minutes at a flow rate of about 1 mL / min with the super and subcritical pressure and temperature.	The best result from the removal of PCBs in most cases (considering the different conditions of subcritical and supercritical temperatures and the proportional pressure) was more than 85%.	(Hartonen, Inkala, Kangas, & Riekkola, 1997).
A laboratory evaluation of superheated steam extraction process for decontamination of oil-based drill cuttings	Different pressures (0.6–5.5MPa), temperatures (175–225 °C), and water flowrates (2–8 ml min-1)	the temperature was the most important parameter, followed by water flowrate and pressure. Both the superheated steam and subcritical water removed not only all extractable organics but also some other compounds.	(Chen et al., 2017)
Supercritical water oxidation of polychlorinated biphenyls using hydrogen peroxide	Batch-reactor system: The reaction pressure and the feed concentration of PCBs were fixed at 30 MPa and 9000 mg/L, respectively.	Under the supercritical water conditions, hydrogen peroxide was found to be significantly efficient. The conversion of 3-PCB and KC-300 exceeded 99% by using hydrogen peroxide at the temperature of 673 K.	(Hatakeda et al., 1999; Westbrook & Dryer, 1984).

All results show that Compared to the conventional extraction processes using solvents, in this process; we can finally remove the pollutant from the solvent by changing the pressure and temperature. Also, its extraction time is lower than the conventional method, less solvent is consumed, less waste is produced and less toxic residues. The solvent used in this method is easily recovered and can be used in subsequent extractions. In this method, less energy is consumed and the composition and structure of the soil are retained (Alonso, Cantero, Garcıa, & Cocero, 2002). Compared to the biological processes, this process is faster and more efficient (Alonso et al., 2002; Marr & Gamse, 2000). But compared to the biological processes, this process requires the excavation, which increases the costs. This process can only remove pollutants from soil, and unlike the biological process, does not have the ability to degrade and convert pollutants into materials.

In developing this process, water was also used as an extraction fluid. However, the use of supercritical water is limited because of the high temperature (more than 374 ° C) and pressure (more than 218 atmospheres) and equipment corrosion. Therefore, the use of subcritical water extraction (SWE), known as pressurized hot water extraction (PHWE), is easier. In the PHWE method, while the water temperature rises from 100 ° C to 274 ° C, Hydrogen bonds between water molecules weaken and dielectric constant of water is reduced. This reduces the polarity of water molecules(Manahan, 2006). Subcritical water, therefore, is more likely to absorb organic compounds than water in environmental conditions. Research has also shown that the SWE method has a better performance in extracting PAHs. The combination of the SWE process with the oxidation process by adding oxidizing agents like air, oxygen, hydrogen peroxide, etc. improves the extraction process in this method(Manahan, 2006; Saldana, Nagpal, & Guigard, 2005).

Conclusion

The supercritical fluid process is always used to remove pollutants such as PHCs, PAHs, PCBs, dioxins, phenols, chlorophenols, insecticides, metals and radioactive substances. The results have shown that the supercritical fluid method using CO_2 has the ability to effectively eliminate organic and inorganic compounds in different contaminated soils (Anitescu & Tavlarides, 2006). Moreover, the cost of this method is acceptable compared to other available methods. On the other hand, the use of this method using water is more studied, as it is eco-friendly and has lower cost and better safety. Because of the high power of solubility, by using the PHWE method, various compounds can be extracted from different matrices at an acceptable temperature below the critical water temperature. In order to enhance the decomposition percentage, supercritical extraction is accompanied by oxidation process using H_2O_2. It shows that combination of supercritical extraction with an advanced oxidation process can significantly enhance the efficiency of the remediation process. The great advantage of this hybrid process is being eco-friendly due to using water as the solvent in the extraction process. For supercritical extraction process for removal of contaminants from the drilling mud, extraction parameters such as temperature and pressure, flow rate of CO2 and static time was examined. The highest removal efficiency were obtained at temperature and pressure of 333 K and 180 bar, respectively, flow rate of lower than 0.1 cm3/s and the static time of 110 min. Extraction efficiencies as high as 98% have been observed. And the hydrocarbons weren’t changed in extraction process and could be recovered and reused. The supercritical water oxidation (SCWO) of OBDC was investigated by some reserachers in a batch and continuous reactor under the conditions of various conditions.

The total organic carbon (TOC) removal efficiency was reached up to 89.2% within 10 min at 500 °C. Therefore, sub and supercritical water oxidation can be used for the conversion of OBDC to into CO2 or harmless compounds.

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