Used lubricating oil recovery process and treatment methods: A review

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Abstract. Used lubricating oil (ULO) is considered hazardous as it is able to cause pollution and affect the environment. The presence of degraded additives, contaminants, and by-products of degradation render ULO more toxic and harmful to health and environment than virgin base oils. Recovery of ULO generally comprises cleaning, drying, and adsorption in order to eliminate water, sludge, and impurities. As the ULO is one of the hazardous wastes generated in various industries, such as industrial and automotive, it should not be used or disposed of in ways that are harmful for the environment. Recovery of ULO carries out many advantages which includes lower environmental impact, higher energy saving and lower risks. The main objective of this paper was to thoroughly review various recovery process principles and treatment methods for ULO. Importance of ULO recycling and various techniques along with their limitations were also discussed. The significance of this study lies in reviewing the roles of adsorbent and adsorption reclamation processes of ULO and few promising adsorbents were earmarked for further study.

Keywords: used lubricating oil, recycling; treatment, pollutants, contaminants, removal methods, environmental pollution

1. Introduction
Lubricating oil (LO) is petroleum derived product and consists of a mixture of hydrocarbons which are a longer chain of isoparaffins, monomycroalkanes and several short branches on the ring of monoaromatics [1]. LOs are made of two components which are the base oil and the additives and are formed by distillation of petroleum with a temperature range from 300 °C to 400 °C [2]. LO is being classified into five different groups which are industrial oil, grease, automotive oils, metalworking fluids and process oils that are widely being used in industries globally [3]. According to Moura et al. [2], LOs are base oils that are mixed with additives to enhance their characteristics which then can be used to minimize wear that are caused by physical friction. During usage, the LO will undergo changes in terms of degradation, oxidation and contamination, which make the LO ineffective for further application and need to be replaced [4]. Literally, LO is used widely in all moving machinerries to reduce the friction between two moving parts.
that are rubbing against each other [5]. In other words, it helps to reduce the build-up of temperature and formation of corrosion within the moving parts that indirectly reduces the efficiency of the machine [1].

Basically, the LOs consist of two components, the base oil and the additives. The main additives include antioxidants, detergents, anti-wear materials, metal deactivators, corrosion inhibitors, rust inhibitors, friction modifiers, pressure-resistant products, anti-foaming agents, viscosity index enhancers, demulsifying or emulsifying agents, stickiness enhancers and so on. However, the performance of the LO will come to an end after a period of time and will reduce the efficiency as the additives become chemically altered and the oil becomes polluted with various harmful toxins and contaminants due to numerous physical and chemical interactions [6]. Degradation of LO also may be due to a mixture of different unwanted matters that contains filings, metal powder, other oils and additives as well. The oxidation of LO produces used lubricating oil (ULO) that has a darker colour, the acid value increases to produce precipitate, bad odour, oil sludge and water content [6,7]. The occurrence of oxidation process will make the LO acidic and these acidic products in the oil will cause corrosion to the internal engine parts, deposits and insoluble oxidation products that literally decrease the performance of the engine. The more the oxidation, the more compounds with higher oxidation states are produced and the acidity of the lubricant increases with the increasing levels of carboxylic acids [8]. Furthermore, at high temperature, the engine oil (EO) will still degrade even with the absence of oxygen. As the temperature increases, the viscosity of the oil increases which is also one of the causes of EO deteriorates [6]. Thus, it could be concluded that by improving thermal stability of EO will also enhance the oxidation stability and viscosity of LO, besides improving the volatility and evaporation rate of EO.

Contaminants containing phenolic compounds, aldehyde, metals, varnish, gums and asphaltic compounds are produced due to the internal combustion of the engine [9]. Emam and Shoaib [10] mentioned that the cause of impurities formation is due to oxidation or thermal degradation of the oil. The further usage of deteriorated ULO in the machineries or equipment’s, the contaminants in the oil deposit on the surface of the moving parts and cause malfunctions of the instrument [11].

The lubricant waste has a very high hazard as it contains massive amounts of ash content, carbon residue, asphaltenic materials, metals, water and other dirty materials [12,13]. Various environmental problems can arise if the waste is not disposed correctly [14,15]. Waste oil management is a growing concern to many nations, especially in the industrial and urban areas as this hazardous waste has been disposed of into the environment and caused a lot of complications.

Somehow, the idea of oil recovery is a great effort and it has been a four decades tradition since it was first presented in the 1930s. Initially, the ULO had undergone combustion for energy production then it was re-blended with engine oil (EO) after treatment. The evolution of recycling technology is crucial for re-refining the ULO to remove the physical, mechanical and chemical contaminations through the process such as distillation, hydrogenation, acidic refining, solvent refining, or combinations of the formers [10]. Lately, there are numerous studies conducted on the recovery of ULO using adsorbent through the process called adsorption [16]. Adsorption is a process whereby an adsorbate is used to adsorb the unwanted contaminants from the used oil to improve the properties [14,16–18].

Hence, the main objective of this paper is to thoroughly review various recovery process principles and treatment methods for ULO. This study was based on addressing problems associated with improper application and disposal of ULOs using recovery techniques and re-refining options for protecting the environment and conserving resources. The significance of this study lies in reviewing the roles of adsorbent and adsorption reclamation processes of ULO and few promising adsorbents were earmarked for further study.
2. Cause of LO degradation

According to Rammohan, 2016 [19], if the LO is not frequently changed, contaminants such as the dirt will accumulate in the engine and fail to lubricate the moving part in the engine. Dirty oil can cause serious damage to the moving parts in the engine whereby replacement of the engine might occur in the near upcoming period. Aljabiri [6] found that the vital mechanisms that cause the depletion of LO additives are due to thermal degradation, oxidation, neutralization, shearing, hydrolysis, water washing, particle scrubbing, filtration, contamination, rubbing contact, surface adsorption, rubbing contact, condensation settling and evaporation. There are three major factors that are identified contributing to the degradation of LO, which includes extreme heat, extreme cold and common contaminants [19,20]. However, the growth of contaminants and chemical variation in the oil itself will cause the LO to be degraded and unsuitable for further usage in the engine parts. The factors of degradation also include entrained air, heat, moisture, incompatible gases, process constituent, radiation, internal or external contamination, and inadvertent mixing of fluids.

Water present in the engine due to condensation, can lead to corrosion. Corrosion related damages in the engine would be more critical when water is present in hotter oil due to the severe chemical reaction. Presence of water in LO for a prolonged period of time, can emulsify the oil and a corrosive mixture is formed which will then form sludge that may block the oil passage or filters [6]. Acidic solutions can also be combined with the water within the engine and create an emulsion that can cause problems with oil filters and passageways [21]. The hydrocarbon reacts with oxygen in the initiation phase of the chemical reaction and forms highly reactive peroxide radicals. Finally, hydroperoxides detach and form oxygenated compounds such as aldehydes, ketones, alcohols, and water. Such compounds further react to form high molecular weight organic acids and polymeric materials. Further polymerisation and polycondensation of these products contribute to an insoluble product called sludge, which will precipitate as a thin film to form lacquers or varnishes on cold or hot metal surfaces [22].

High temperature with increased time of usage will degrade the desired properties of LO and thus need to be evacuated and replaced with a new one [23]. ULO contains metals such as lead, zinc, barium, arsenic, chromium and cadmium [24,25]. In terms of chlorinated hydrocarbons, the ULO consists of compounds such as dichlorodifluoromethane and trichlorotrifluoroethane. Moreover, other organic matter is also present in ULO such as benzene, xylene, toluene and naphthalene [25]. The ULO main constituent is the degraded additives, base oil, metallic debris, carbon soot, and oxidation product. A huge number of additives are used to convey the lubricants' performance and during use, these additives lose their characteristic. In addition, the LO picks up a fraction of various metals as the result of wearing of components. Other contaminants such as chlorinated solvents, water, unburned fuel, carbon and dust are also picked up during storage [26].

The air contaminants may contain moisture, dirt, and dust, and even the air itself is a contaminant that could cause the oil to foam. Such contaminants move into the engine via the air-cleaner and may consist of minor silicate elements [6]. Meanwhile, the engine contaminants are the metallic particles due to the wear of the engine, carbonaceous particles from the incomplete combustion, metallic oxides from the metal corrosion, water from the cooling system leakage and the product of combustion, and the fuel additive or its by-product which might reach the engine crankcase [20]. In addition to that, since the LO act as an anti-corrosive, cooling and cleaning agent, it picks up a variety of impurities and additional components which include the metal particles such as steel, iron, copper, zinc, lead and other compounds of sulphur, water, barium, carbon, and ash [27]. These contaminants directly interfere with the lubricant’s viscosity, lowering its efficiency and performance.
3. Effects of ULO contaminants to human health and environment

The contaminants in ULO render a wide range of problems to human healthiness and environment [28,29]. The presence of degraded contaminants, additives and by-products such as water and sediments will transform the ULO into a toxic one compared to virgin base oil. The improper disposal of ULO can lead to adverse environmental and health problems. For landfills with permeable soils, water-soluble materials in lubricants used can easily penetrate waste and end up contaminating groundwater by runoff leaching and percolation. The waterways and the coastal waters can be affected if the ULO is being disposed of in storm water or waterways. Excessive ULO will cause a threat to plant and animal life, which may lead to economic losses in fishing and recreation industries [24]. Musemić and Bašić, [30] mentioned that ULO has a variety of serious, short and semi-permanent impacts on the environment if disposed of in an uncontrolled manner. The ULO inhibits the penetration of chemical elements such as oxygen into water, therefore degrading the condition of life of several species. ULO contains various poisonous substances, together with polycyclic aromatic hydrocarbons, that are famed to cause cancer if they are inhaled or ingested [31]. Waste oil that has been disposed of in the water will form a thin oil layer that blocks sunlight on the water surface that inhibits the oxygen from dissolving in water [14,31]. This will lead to extinction or death of the aquatic plants and animals in water. Besides, when exposed to air, at high temperature, the ULO can be vaporized to air polluting the fresh air as well. As mentioned, ULO consists of numerous contaminants such as hydrocarbons, heavy metals, halogen compounds, gums, varnish and polychlorinated biphenyls that can harm human beings, aquatic life and environment as well [32]. Not only that, presence of dangerous elements such as polycyclic hydrocarbons (PAHs) and aliphatic hydrocarbons in ULO had a higher flexibility to cause carcinoma [33]. ULO are also very toxic to animals, particularly embryos and new-born animals. Table 1 summarizes the effects of ULO contaminants and impact of oil emissions to human health and environment.
Table 1. Effects of ULO contaminants and impact of oil emissions to human health and environment.

| Oil contaminants | Effects | References |
|------------------|---------|------------|
| Water            | Causes rusting of iron or steel | [6] |
|                   | Results in the development of water sludge | |
|                   | Leads to foaming problems. | |
| Solid particles   | Causes excessive wear, bearing surface score, and potential failure to absorb metal fatigue. | [6] |
|                   | Sludge deposits are clogging narrow oil passages and clearances. | [6] |
| Sludge and lacquers | Causes the valve to stick and resist the continuous operation of the oil pump. | |
| Liquid contaminants | Unburned fuel from engines dilutes LO and may lessen its viscosity. | [6] |
|                   | Impact of oil emissions | |
| Base oils derived from crude oil | Eco-toxicity, incomplete biodegradability, and highly probable carcinogenicity | [29] |
|                   | Causing significant health issues such as changes in the lungs, liver, kidneys, adrenal glands, and heart. | [28] |
| Oil mist          | It results in a negative influence on the respiratory and nervous systems. | |
|                   | It is also absorbed by skin and causes skin cancer | [29] |

4. Importance of ULO recycling

As previously mentioned ULO is classified as hazardous and constitutes extensive damage to the environment, creating risks of contaminating water, air, and soil with the presence of harmful substances. Consequently, the management of ULO is predominantly vital due to the large amounts generated worldwide, the potential for straight reuse, regenerating, reprocessing, and environmental damaging effects if not handled, treated or disposed of properly. As the number of vehicles increases gradually, the volume of ULO produced is also growing. The disposal and management of the wastage is indeed a rising fear, especially in industrial and urban areas. Many nations are now addressing the predicament of environmental pollution posed by LO wastage [12].

ULO recovery not only guards the environment but likewise preserves a treasured non-renewable resource [25]. ULO is likewise a valuable source of carbon and is considered as one of the important energy sources in the world. ULO may be re-refined into base oil, reprocessed as oil or used as feedstock to supply petroleum-based merchandise or different commercially valuable products through totally different ways. Additionally, the valuable non-renewable resources would be conserved besides solely protecting the environment from ULO [34]. From the point of energy conservation, utilization of ULO expeditiously saves precious, renewable resources. During these days, many companies are investing billions of greenbacks in recovering, exploring and processing ULO into quality LO. Moreover, ULO generated by the customers will be recycled back to helpful merchandise like pure fill oil again and once more, tremendous time and money can be saved as well [34,35]. Presence of refining equipment can reduce cost for hydro treatment, storage of product and pollution can be decreased. On top of that, instead
of improper disposal of ULO, it is recycled, re- fined or reused to prevent contamination of soil, groundwater and surface water.

In a nutshell, if ULO is well managed, the scenario of depending on fossil fuels from other countries would be decreased [35]. Proper assortment and regeneration of ULO will be useful to the nation thereby reducing environmental pollution, conserving the mineral resources and saving the foreign exchange for importing the virgin oil [14].

5. **ULO recovery and reuse techniques**

After being consumed in a period, the petroleum-based LO has become unsuitable for the designed purpose. Due to toxicity through use, the ULO is unable to handle anthropogenic pollutants and become undesirable [36]. However, this wastage can also be considered as a valuable resource in the sense that it can reclaim energy or any profitable material for further use. Thus, recycling or recovery of LO wastage may be an appropriate low-cost alternative and indeed a need of the hour due to its economic, environmental, legal reasons, and public health importance [7]. The disposal management and recovery of ULO have become a crucial issue due to the increasing necessity of environmental protection and strict environmental legislation [37]. One of the disadvantages of recycling ULO is the mixing of different types of waste oil. Besides, there is no clear and specific way of collecting or gathering ULO for recycling. In reality, recovered ULO can be used as an industrial fuel burner, hydraulic oil, integrated into other goods or re-engineered into new LOs [38].

Recovery of ULO can slow down the consumption of natural resources. Recycling aims to a large degree to combat global climate change. By reducing the energy expended on industrial production, recycling helps in minimizing greenhouse gas emissions. By minimizing the energy used, recycling also minimizes the usage of fuel, which in turn decreases the amount of harmful contaminants in the atmosphere [39]. Recovery of ULO also contributes to economic benefits. The recycling process creates employment opportunities for a large number of people involved in the various stages of the process [40].

The reuse of ULO can be approached by three basic methods, namely incineration, reprocessing and re-refining. In incineration, the ULO is treated with the presence of heat. The used oil is thermally destroyed. Diphare et al. [26] claimed that, during these contemporary days, incineration is rarely used due to the economic benefits of recycling. Moreover, burning of ULO releases toxic contaminants to the environment. Thus, such a type of recycling is unsafe and impractical. In reprocessing technology, the ULO is heated and processed for energy recovery. It involves water and particulates removal from the oil before burning the fuel to generate heat. Unfortunately, this type of recycling does not provide the best result to recycle the contaminant high ULO. This is because reprocessing methods of ULO may produce by-products that are even more toxic as the secondary pollutant. Furthermore, ULO can be also recycled through a re-refined process. In this process the ULO is re-refined back into lube oil that can be recycled and reused. In this method, the contaminants such as water and dissolved low boiling organic is eliminated. Different treatments are used to recover the lube oil. Re-refining is different from incineration and reprocessing, whereby in this process, the ULO is treated to remove the impurities and increase the properties of the oil, in order to be used as a virgin LO. Re-refining technology of ULO into LO requires less energy than incineration and reprocessing [26]. Thus, it is being preferred as it consumes less energy, produces no damage to the environment and less waste generation. However, the reprocessing of the recovered material is not always free from pollution. Some reprocessing technologies produce residues that are difficult to treat. The acid-clay method for the re-refining of ULO is one of the examples where residual sludge has polluted the soil. Moreover, conventional acid-clay treatments usually require the use of highly expensive adsorbents such as kaolin, alumina, silica gels etc. According to Shakirullah et al. [41], existing acid-clay treatment techniques for recovery require endless chemicals and give low yield. Table 2 summarizes the comparison of re-refining methods.
| Sl. no. | Method                                      | Type of sample | Outcome                                                                 | References |
|--------|--------------------------------------------|----------------|------------------------------------------------------------------------|------------|
| 1.     | Acetic acid-clay treatment                  | Used car EO    | Water and sediment content: 0.013 ml, Carbon residue: 1.65 wt%, Total acid number (TAN): 1.8 mg KOH/g oil, Metal content- Cu:3.56 ppm; Mg:65 ppm; Pb:11.2 ppm; Zn:780 ppm | [42]       |
| 2.     | Solvent extraction/clay re-refining process | ULO            | Sulfur: 0.81 wt%, Water content: Nil; Specific gravity@ 40 °C: 0.6991, Metal content- Fe: Nil; Cu: Nil; Zn: 117.8 ppm | [9]        |
| 3.     | Acid treatment/clay – percolation re-refining | ULO            | Sulfur:0.42wt%, Water content: Nil; Specific gravity@ 40°C: 0.8202, Metal content- Fe: Nil; Cu: 1.048 ppm; Zn: | [9]        |
| 4.     | Extraction by composite solvent followed by acid treatment | Used EO        | Flash point: 150 °C, Specific gravity:0.88, Viscosity @ room temperature: 94 cp, Pour point:-15°C, Ash content (%): 0.09, Iron: 13 ppm | [27]       |
| 5.     | Ionizing radiation process                  | Used automotive LO | Mg, Al, Ti, Cr, Mn, Fe, Ni, Cu, Se, Mo, Nb, Cd, Sn, Ba, Bi and Pb : <10 μg.ml⁻¹ | [43]       |
| 6.     | Solvent extraction process                  | Used motor EO  | The sludge formation increases with increasing solvent 1-butanol produce the best extraction performance in sludge formation | [44]       |
| 7.     | Microwave pyrolysis reactor                 | Car EO         | Formation of single ring alkyl aromatics such as benzene, toluene, ethyl-benzene and xylene | [45]       |
| 8.     | Vacuum distillation recycling by continuous extraction with dense propane | ULO            | Metal content- Pb: 0.1 ppm; Fe: 0.06 ppm; Zn: 0.01 ppm; Ca: 0.01 ppm | [46]       |
| 9.     | Distillation-clay treatment                 | LO             | Water content: 0.66 v/v Specific gravity: 0.86 Viscosity index: 85.80 | [1]        |
6. Adsorbent and adsorption process in the treatment of ULO

The adsorption process is primarily used for purification or separation purposes. Adsorbent is a micro porous structure that is manufactured to have adsorption properties. The high porous medium offers a large amount of micropores; this results in a large capacity for adsorption. The porous solid material has very small pores in thickness, where adsorbed molecules can find their way to the micropore surface [48].

There are many factors that literally affect the efficiency of the adsorption process such as contact time, temperature and adsorbent dosage [16–18,49]. Few studies have been conducted using activated carbon (AC) in the treatment of the ULO by replacing clay to adsorbents. ULO was recovered by using various types of adsorbents such as date palm kernels powder, bentonite, eggshell powder and so on. According to Riyanto et al. [50], the AC is a good material for treatment of ULO, especially to reduce the metal concentrations. The rising amount of AC increases the adsorption potential of the metal in the LO waste. The waste oil treatment using clay or adsorbent can be improved by addition of a catalyst. Bhaskar [51] presented the supported iron oxide catalyst to be used in the conventional clay treatment in producing fuel oil from ULO. The adsorbent has its advantages due to its flexibility and simplicity of design, ease of operation, and insensitivity to toxic pollutants. However, some adsorbents require activation to change the raw material to a more porous material so it can operate at its finest. The main parameters upon selecting the adsorbent are the choice of adsorbent and particle size [52–55].

7. Recommendation of adsorbents for the ULO treatment

Fuller’s earth (FE) is a type of sedimentary clay having high magnesium oxide content which is commonly used in bleaching, refining edible oils and clarifying petroleum. FE is preferred due to its high adsorption capacity and low purchasing cost. It decolorizes the oil without variation in chemical properties of oil [56]. Previous studies have reported that it has the ability in removing heavy metals in wastewater and to extract impurities of ULO [56–58]. Recovery of ULO might be done by using different acid solvents paired with FE through acid-clay treatment. River sand (RS) has the ability to remove metal contaminants from aqueous solution was inspired by its wide application in wastewater treatment. It has the ability to remove contaminants and mercaptans from liquid as well. Many characteristics of the RS can

|   |   |   |
|---|---|---|
| 10 | Acid treatment | LO |
|   |   | Flash point: 168 °C |
|   |   | Water content: 0.60 v/v |
|   |   | Specific gravity: 0.86 |
|   |   | Viscosity index: 84.40 |
|   |   | [1] |
| 11 | Activated Charcoal-clay treatment | LO |
|   |   | Flash point: 170 °C |
|   |   | Water content: 0.47 v/v |
|   |   | Specific gravity: 0.86 |
|   |   | Viscosity index: 86.80 |
|   |   | Flash point: 178 °C |
|   |   | [1] |
| 12 | Acid treatment | Motor oil |
|   |   | Formic acid specific gravity: 0.915 |
|   |   | Flash point: 230 °C |
|   |   | Sulphuric acid specific gravity: 0.920 |
|   |   | Flash point: 210 °C |
|   |   | Phosphoric acid specific gravity: 0.930 |
|   |   | Flash point: 200 °C |
|   |   | Acetic acid |
|   |   | Specific gravity: 0.915 |
|   |   | Flash point: 218 °C |
|   |   | [47] |
make it a very good and cheap adsorbent [59]. However, RS needs to be properly treated in removing contaminants from the ULO.

Bleaching earth are actually bentonite clays or known as montmorillonite that are used as absorbent. Spent bleaching earth (SBE) is known as the by-product that is produced in the palm oil refinery [16,60]. SBE contains lower specific surface and higher pore volume compared to fresh bleaching earth. According to Loh et al. [61], the empty pores are being filled by the metals after oil refining. In the palm oil industry bleaching earth is commonly used to adsorb the unwanted colour pigment and undesirable residues. SBE commonly disposes of the landfill upon use which leads to the increased amount of landfill waste. Disposal of SBE in the dumpsite can cause environmental issues as the leaching of the residual fat by rain may pollute groundwater and fire hazards. SBE could be regenerated via chemical purification, solvent extraction or combination of these methods [62–63]. After regeneration SBE could be used for the adsorption treatment of ULO [64–66]. However, for the recovery of ULO using adsorbents and optimization of the adsorption process parameters is still lacking in much of the published literature.

8. Conclusion
ULO is a highly hazardous waste that requires very responsible waste management. It can cause detrimental effects not just to the environment but also to humans if it is not properly disposed of, handled or treated. In this literature review, few examples of recycling ULO have been discussed. Besides, the roles of adsorbent in recovering the ULO also have been discussed thoroughly. In order to prove that adsorption can recover the oil, previous studies have been listed and few promising adsorbents were earmarked for further study. However, it is recommended to investigate the optimization of adsorption process parameters which could maximize the efficiency of ULO recovery.

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