# **Review Article**

# **3** A review of methods for removal of contaminants in used lubricating oil

4

1 2

# 5 Abstract

6 Management and disposing of used lubricating oil (ULO) poses deleterious effects to air, land and water pollution. These contaminants not only causes environmental problems, they also 7 8 have bio-accumulation effects on living organisms, reduces the inhabitants lifespan-of 9 inhabitants as a result of the diseases spread-of diseases, poisoning and fouling of catalyst as 10 well as corrode processing equipment. The contaminants removal Removal of contaminants in used lubricating oil is a major step to avoid pollution as discussed thoroughly by many 11 researchers in literature. In addition, to curbing pollution, another advantage is converting 12 13 waste to wealth. This review paper presents insight into various methods for removal of contaminants in used lubricating oil. The advantages and drawbacks of each method were 14 earmarked for further study. 15

16 Keyword: Used lubricating oil, Contaminants. Removal Methods, Treatment

### 17 Introduction

Lubricating oils (LOs) are conventionally obtained from crude oil. Chemical composition of 18 LOs consists on average of about 80–90% base oil and about 10–20% chemical additives and 19 20 other compounds (Rincón et al., 2005). Lubricating oils mainly helps in reducing friction, dust, corrosion, protection against wear and tear and provision of heat transfer medium in 21 22 various equipment or machineries. (Shri et al., 2014). During operation time, LOs deteriorate, as well as their additives, and its physical and chemical properties become unsuitable for 23 further use (Tsai, 2011). In the process of lube oil usage of lube oil, temperature build up 24 occurs which breaks down-the oil and weakens its properties which include pour point, flash 25 point, specific gravity, viscosity etc. (Udonne and Bakare, 2013). These renders the oil 26 27 unsuitable for regular usage as results of contaminants in-the lube oil such as water, wear 28 metals, carbon residue, ash content, gums, varnishes etc. Chemical changes in the-oil 29 occurred due to thermal degradation and oxidation. Europe represents 19% of total worldwide market volume of lubricants, consuming around 6.8 million tons in 2015 (Kupareva et al., 30 2013). 31

Used lubricating oils (ULOs) are classified as hazardous wastes, and constitute a serious 32 33 pollution problem not only for-the environment, but also for human health due to-the harmful contaminants presence of harmful contaminants, such as heavy metals, polychlorinated 34 35 biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) (Kanokkantapong et al., 2009). Poor management and careless disposal of used lube oil can affect the environment 36 negatively (Lam and Chase, 2012 and Lam et al., 2016). Scientists have reported that in some 37 38 geographical region e.g. West Africa, the dispersion of the air pollutants could travel at a speed of 10-12 m/s (Emetere, 2017). The implication of this report is that air pollution from 39 40 burning of waste lubricant is not localized to the pollution source the source of pollution but 41 could travel with time to other locations. For example, it was recently reported that black soot

**Formatted:** Font: Italic

Formatted: Font: Italic

42 covered a metropolitan city of Port Harcourt while remote sources were at the suburb
43 settlement (-about 22 km away from the city) (Temitayo *et al.*, 2018).

On the other hand, ULOs can be considered as valuable resources, in the sense that it is 44 45 possible to recover energy or profitable materials for further use (Guerin, 2008). The best 46 environmental options, for the management of used lube oils follow the 'waste hierarchy' by recycling, recovering and then disposing. Used lube oils can be used as an alternative fuel in 47 a variety of engine configurations and other applications. Its gross calorific value is greater 48 than 42.-9 MJ/kg (Ketlogetswe, C., 1998). The principal objective of any waste management 49 plan is to ensure safe, efficient and economical collection, transportation, treatment and waste 50 disposal of waste and as well as satisfactory operation for current and foreseeable future 51 52 scenarios (Stoll and Gupta, 1997). The treatment of used lube oil is important due to: (1) lit requires less energy and cost compared to conventional refining of crude oil; (2) Lit helps in 53 54 improving air quality, land and water pollution in the environment. The most preferred option by experts is the reuse of the-used lube oil generated by consumers (Jafari and Hassanpour, 55 2015). In this paper, a thorough review on various removal and treatment methods for used 56 57 lube oil would be considered starting from conventional to the most current methods and their 58 limitations i further developments of these fields were also touched. In addition, environmentally friendly and affordable solvent extraction and adsorbents would be 59 developed as a means of removing contaminants in used lube oil. 60

### 61 2. Conventional Methods

The conventional methods of <u>contaminants</u> removal <u>of contaminants</u> in used lube oil either
 requires a high cost technology such as vacuum distillation or the use of toxic materials such
 as sulphuric acid. These methods also produce contaminating by-products which have highly

sulphur levels, especially in the Kurdistan region/Iraq (Hamawand *et al.*, 2013).

# 66 2.1. Acid-clay

Assessment of different contaminants removal of contaminants processes in used lube oils
 revealed that acid-clay process had the highest environmental risk and lowest cost. The

69 method involve treatment of used oil with acid and clay (Udonne and Bakare, 2013;

70 Hamawand et al., 2013; Abu-Elella et al., 2015). They all used the clay as an adsorbent to

71 remove the odour and dark colour. What makes acid-clay method unique from others are; with

real its simple method, affordable capital investment, low operating cost and does not need skilled

73 operators (Giovanna *et al.*, 2012; Nwachukwu *et al.*, 2012 and Isah, A.G., 2013).

74 However, this method has many disadvantages: 14 also produces large quantity of pollutants, is unable to treat modern multigrade oils and it's difficult to remove asphaltic impurities 75 (Fox, M.F., 2007). To reduce these hazardous contaminants from this method,; the acid 76 77 treatment stage of the process can be done under the atmospheric pressure to remove the 78 acidic products, oxidized polar compounds, suspended particles and additives (Falah and Hussein, 2011). - Princewill and Sunday, (2010) observed that high recovery rate of treated 79 80 lube oil from used lube oil depend largely on the source of the used lube oil, pre-treatment mechanisms, extent of contamination and the grade of-the acid used. He also showed that the 81 volume of-the adsorbent (clay) used could affect the rate at which contaminants are removed 82 83 and the recovery percentage of recovery of the method.

Formatted: Font: Italic

Formatted: Font: Italic

Formatted: Justified
Formatted: Font: Italic
Formatted: Font: Italic
Formatted: Font: Italic
Formatted: Font: Italic

84 In Abu-Elella *et al.*, (2015) worked on used motor oil. He treated used motor oil with

85 phosphoric acid, sulphuric acid, methanoic acid and acetic acid. -He observed that methanoic

86 acid, sulphuric acid and acetic acid have great changes on the kinematic viscosity while

87 phosphoric acid is not affected by used lube oil. He therefore concluded that treatment with

88 acetic acid showed better results than formic acid-clay.

# 89 2.2 Solvent Extraction

This method has replaced acid-clay treatment as the preferred method for improving the
oxidative stability and viscosity as well as temperature characteristics of the base oils. Base
oils obtained from Solvent Extraction are of good quality and contains less amounts of
contaminants. In contrast to acid-clay treatment, it operates at higher pressures, requires
skilled operating system and qualified personnel- (AERCO 1995). The solvent selectively
dissolves the undesired aromatic components (the extract), leaving the desired saturated

components, especially alkanes, as a separate phase (the raffinate). (Rincon *et al.*, 2005).

97 Different solvents types of solvents have been used for solvent extraction such as 2-propanol,

98 1-butanol, methyl ethyl ketone (MEK), ethanol, toluene, acetone, propane etc. (Quang *et al.*,

99 1974) and (Rincon *et al.*, 2003) used propane as a solvent. He found out that the propane was

100 capable of dissolving paraffinic or waxy material and intermediately dissolved oxygenated

101 material. Asphaltenes which contain heavy condensed aromatic compounds and particulate

102 matter are insoluble in liquid propane. These properties make propane ideal for recycling the

103 used engine oil, but there are many other issues that have to be considered. Propane is

104 hazardous and flammable therefore this process is regarded as hazardous method.

Katiyar and Husain (2010); Sterpu *et al.*, (2012) and Hassan *et al.*,(2012) found out that
methyl ethyl ketone has the highest performance due to its low oil percentage losses and high
sludge removal while Hussein *et al.*, (2014) and -Aremu *et al.*, (2015) -found out extraction
using butan-1-ol solvent produces the highest sludge removal rate. (Rincon *et al.*, 2005) and
Oladimeji *et al.*, 2018) used a composite solvent of methyl ethyl ketone and 2-propanol the
oil resulting from this process is comparable to that produced by acid-clay method, its cost
was high.

Solvent extraction, in general, involves solvent losses and highly operating maintenance.
 Also, it occurs at pressures higher than 10 atm and requires high pressure sealing systems
 which makes solvent extraction plants expensive to construct, operate and the method also

115 produces remarkable amounts of hazardous by-products. (Quang *et al.*, 1974); (Rincon *et al.*,

116 2003) and Hamawand *et al.*, (2013).

Mineral "ol Raffinerie Dollbergen (MRD) solvent extraction process using N-methyl-2-117 pyrrolidone. The applied oil re-refining process is based on a patent held by AVISTA OIL. 118 119 (P"ohler et al., 2004) The 'Enhanced Selective Refining' process uses solvent N-methyl-2-120 pyrrolidone (NMP), which is commonly used in-the petroleum refining industry. NMP is a 121 powerful, aprotic solvent with low volatility, which shows selective affinity for unsaturated 122 hydrocarbons, aromatics, and sulphur compounds. Due to its relative non-reactivity and high selectivity, NMP finds wide applicability as an aromatic extraction solvent in lube oil re-123 124 refining. The NMP advantages of NMP over other solvents are the non-toxic nature and high 125 solvent power, absence of azeotropes formation with hydrocarbons, the ease of recovery from solutes and its high selectivity for aromatic hydrocarbons. Being a selective solvent for 126

Formatted: Font: Italic

Formatted: Font: Italic

Formatted: Font: Italic Formatted: Font: Italic

Formatted: Fo	ont: Italic
Formatted: Fo	ont: Italic

1	Formatted: Font: Italic
1	Formatted: Font: Italic
	<b>Comment [EC1]:</b> Veriy the bibliographic citation pattern
1	Formatted: Font: Italic
1	Comment [EC2]: I could understand
1	Formatted: Font: Italic

aromatic hydrocarbons and PAH, NMP can be used for<u>the</u> re-refining of waste oils with

128 lower sludge, carbonaceous particles and polymer contents, such as waste insulating,

129 hydraulic and other similar industrial oils.( Lukic, J et al., 2005). The MRD solvent

130 extraction process uses the liquid–liquid extraction principle.

131 The average base oil yield within the process is about 91 %.( Schiessler, <u>*P et al.*</u>, 2007). The

base oils produced have high quality (Kupareva *et al.*, 2013). The process is characterized by

133 optimized operating conditions which allow elimination of toxic polyaromatic compounds

134 from the re-refined base oil and preservation of the synthetic base oils like polyalphaolefin

135 (PAO) or hydrocracked oils, which are increasingly present in used oils. However, this

136 method need skilled personnel, proper disposal and management of it waste.

# 137 2.3 Vacuum distillation

Extensive research work have been done on vacuum distillation on used oil by the following 138 Martins, J.P. (1997); Shakirullah et al., 2006; Bridjanian and Sattarian (2006); Emam and 139 140 Shoaaib, (2012); Hamawand et al. (2013) and Kannan et al., (2014). In this method, used 141 lube oil collected is heated at a temperature of 120°C to remove the water added to the oil during combustion. Then the dehydrated oil is subjected to vacuum distilled at a temperature 142 of 240°C and pressure 20 mmHg. This results-to the production of a light fuel oil at a 143 temperature of 140°C (the light fuel oil can be used as fuel source for heating) and lubricating 144 145 oil at 240°C. The lubricating oil vapour is condensed and sent for next stage. (Kannan et al., 146 2014). The advantages of vacuum distillation process over atmospheric pressure distillation 147 are: Ceolumns can be operated at lower temperatures; Memore economical to separate high boiling point components under vacuum distillation; Aavoid degradation of properties of 148 149 some species at high temperatures therefore thermally sensitive substances can be processed 150 easily.

However, the remaining oil generated at this temperature (240°C) contains the dirt, degraded additives, metal wear parts and combustion products like carbon and is collected as residue.

153 The residue is in the form similar to that of tar, which can be used as a construction material, 154 for example, used and hittmen mediation (Civianne et al. 2002). The disadvertage of this

for example, road and bitumen production. (Giovanna *et al.*, 2003). The disadvantage of this method is the high investment cost and/or the use of toxic materials such as sulphuric acid.

156 (Havemann, 1978 and Puerto-Ferre, & Kajdas, 1994).

# 157 2.4 Hydrogenation

To avoid formation of harmful products and environmental issues based on above methods,
some modern processes have been used and the best one is hydrotreating. (Bridjanian and
Sattarin, 2006). This method follows vacuum distillation. In this process, the distillate from
vacuum distillation is hydrotreated at high pressure and temperature in the presence of
catalyst for the purpose of removing chlorine, sulphur, nitrogen and organic components. The
treated hydrocarbons resulted in products of improved odour, chemical properties and colour.
(Temitayo *et al.*, 2018).

Another important aspect of this method is that, this process has many advantages: Produces of high Viscosity Index lube oil with well oxidation resistance and a good stable colour and yet having low or no discards. At the same time, it consumes bad quality feed. In addition to

that, this method has advantage that all of its hydrocarbon products have good applications



Formatted: Font: Italic



Formatted: Font: Italic

Formatted: Font: Italic

Comment [EC3]: I could understand!

Formatted: Font: Italic

and product recovery is high with no (or very low) disposals. Other hydrocarbon products
are: In oil refinery the light—cuts can be used as fuel in the plant itself. Gas oil may be
consumed after being mixed with heating gas oil and the distillation residue can be blended
with bitumen and consumed as the paving asphalt, because it upgrades a lot its rheological
properties. Also, it can be used as a concentrated anti-corrosion liquid coating, for vehicles
frames. (Hassan A. D, 2014).

The disadvantage of this method is that the residue resulting from the process is of high boiling range of hydrocarbon product fractionated into neutral oil products with varying viscosities which can also be used to blend lube oil (Basel Convention, 2002).

### 178 2.5 Membrane Technology

Membrane technology is another method for removal of contaminants of used lubricating oils. In this process, three types of polymer hollow fibre membranes [polyethersulphone (PES), polyvinylidene fluoride (PVDF), and polyacrylonitrile (PAN)] (Lam *et al.*, 2016) were used for recycling the used engine oils. The process is carried out at  $40^{\circ}$ C and 0.1 MPa pressure. The process is a continuous operation as it removes metal and particles and dusts from used lube oil and improves the recovered oils liquidity and flash point. (Dang, C.S., (1997) and Hamawand *et al.*, (2013).

Despite the above mentioned advantages, the expensive membranes may get damaged and
fouled by large particles with time. (Dang, C.S., (1997) and Hamawand *et al.*, (2013).

## 188 2.6 Catalytic Process

189 For example, Hylube process from Germany. This process allows production of mainly base

- 190 oils. The Hylube process is a proprietary process developed by Universal Oil Products (UOF
- 191 for the catalytic processing of used lube oils into re-refined lube base stocks for re-blendin

into saleable lube base oils (Kalnes *et al.*, 2006). This is the first re-refining process in which
as received used oil is processed, without any pre-treatment, in a pressurized hydrogen
environment. A typical HyLube process feedstock consists of a blend of used lube oils
containing high concentrations of particulate matter such as iron and spent additive

196 contaminants such as zinc, phosphorous, and calcium (Chari-K.R, 2012).

The Hylube unit operates with reactor section pressures of 60–80 bar and reactor temperatures in the range 300–350°C (Kalnes–T.N and Schuppel–A, 2007).– The Hylube process achieves more than 85% of lube oil recovery from the lube boiling range hydrocarbon in the feedstock (Kupareva *et al.*, 2013). Besides the advantages of these process, this method is very expensive. This method requires high level personnel due to high temperature and pressure operations.

# 203 3. Combined Technologies/methods

These are advance methods that combines two or more generic methods in its process. Due to the complex nature of <u>contaminants</u> removal <del>of contaminants</del> in used lube oils, using a single method may not give you the desired standard <del>emission controlledemission-controlled</del> process. Therefore, some companies have developed specific processes for treatment and

208 <u>contaminants</u> removal of contaminants in used lube oils (Basel Convention, 2002; Brinkman

209 2010 and Kupareva <u>et al.</u> 2013), these methods require sophisticated technologies,

equipment and processes. Some of these complex processes are briefly discussed below;

16 16	
re	Eormattad: Eopt: Italic
$D_{g}$	- Formatted. Form, Italic
a te	
C S	
ο,	Eormatted: Font: Italic
'	
nd	
	Formatted: Font: Italic
se	
P)	
ıg	
h_	<b>Formatted:</b> Font: Italic
en	
ls	
/e	
<b>.</b>	
51	
30	
sc_ rh	Formatted: Font: Italic
gn	
t	
10 10	
ie	
<u>D:</u>	
nd	
n,	

#### 3.1 Vaxon process 211

212

Formatted: Font: Italic Formatted: Superscript Formatted: Font: Italic Formatted: Font: Italic Formatted: Font: Italic

Formatted: Superscript

-This process contains chemical treatment, vacuum distillation and solvent refining units. The advantage of the Vaxon process is the special vacuum distillation, where the cracking of oil is 213 214 strongly decreased. (Chari, K.R., 2012). 215 The chemical final stage does not, however allow the high-quality base oils production-of 216 high quality base oils; although in Spain the Catalonia refinery produces base stocks accepted by an original equipment manufacturer (OEM). In connection with this fact, the lube distillate 217

obtained from the Vaxon process (Denmark) or North Refining (Netherlands) are precursors 218

for the Avista Oil base. (Kupareva et al., 2013). 219

#### 220 3.2 CEP process

-This process combines thin film evaporation and hydroprocessing. The used oil is chemically 221 222 pre-treated to avoid precipitation of contaminants which can cause corrosion and fouling of

- the equipment. The pre-treating step is carried out at temperatures within 80–170°C. The 223 224 chemical treatment compound comprises sodium hydroxide, which is added in a sufficient 225 amount to give a pH about 6.5 or higher. (Magnabosco L.M. and Rondeau W.A., 1993).
- Heavy materials (such as residues, mMetals, additive degradation products, etc.) are passed to 226 227 a heavy asphalt flux stream. The distillate is hydropurified at high temperature (315°C) and 228 pressure (90 bar) in a catalytic fixed bed reactor. (Merchaoui et al., 1994). This process removes nitrogen, sulphur, chlorine and oxygenated organic components. In the final stage of 229 the process, three hydrotreating (Hydrofinishing) reactors are used in series to reduce sulfur 230 231 to less than 300 ppm and to increase the amountnumber of saturated compounds to over 95%, in order to meet the key specifications for API Group II base oil. The final step, in this 232
- process is vacuum distillation to separate the hydrotreated base oil into multiple viscosity cuts 233

in the fractionator. The yield of base oils is about 70%. (Kupareva et al., 2013). 234

#### **3.3 Ecohuile process** 235

The re-refining process was based on vacuum distillation and acid-clay treatment steps until 236 237 the end of 2000. (Audibert, F., 2006). Clay adsorption was banned on 1 January 2001 and the 238 plant was modified and upgraded to the Sotulub process. (Sotulub re-refining process. 2005). Moreover, the addition of injection facilities of so-called Antipoll-additive (1-3 wt% of pure 239 sodium hydroxide) has been provided and has allowed solving the following basic problems: 240

- · Ceorrosion of dehydration column and cracking column top section due to the organic 241 acidity of the used oil; 242
- 243 • Pplugging of equipment and piping due to polymer formation in the cracking section;
- Hhigh losses of base oil in the oily clay due to the high consumption of clay. 244
- The Sotulub process (Merchaoui, M, H et al., 1994) is based on treatment of the used oil with 245
- 246 an alkali additive called Antipoll and high vacuum distillation. The used oil is pre-heated to about 160°C and mixed with a small amount of Antipoll-additive, which decreases equipment 247
- 248 fouling. This process, allows a final product to be obtained with acceptable quality without
- any additional finishing stage. Oil obtained is additionally fractionated to obtain various base 249
- 250 oil cuts. The process provides base oils with a yield of 82–92-%. (Sotulub re-refining process.

2005). 251

### 252 3.4 Cyclon process

-This process combines the technology of vacuum distillation and hydrofinishing. 253 (Havemann, 1997). The process licence belongs to Kinetic Technology International (KTI). 254 255 (Kajdas, C, 2000). In this process, used oils taken from storage tanks are dewatered and the 256 light hydrocarbons are removed by distillation. The heavier fraction is sent to high vacuum 257 distillation, where the majority of base oil components are evaporated from the heavy residue. The oils in-the residues are extracted with propane in the-de-asphalting unit and sent 258 to the hydroprocessing unit where the other oils are processed. Then they are treated with 259 hydrogen and fractionated based on the-desired base oil features. The re-refined base oil 260 products obtained are of high quality due to the hydrogenation. (Schiessler, N, et al., 2007 261 262 and Tsalavoutas, S. et al., 2002)

# 263 **3.5 STP method**

This is another advance method that combines vacuum distillation and hydrofinishing process
(Basel Convention, 2002). It produces less harmful pollutants therefore its environmentally
friendly (Kupareva *et al.*, 2013). This method involves dehydration, vacuum distillation,
separation of the lubricating fraction and hydrofinishing of base oil separation from the

268 residue.

## 269 **3.6 Interline process**

-Interline proposes a process based on propane de-asphalting at ambient temperature and
under a pressure that facilitates separation in the liquid phase. The lubricating oil yield
declared for the Interline process is 79 %.(-Monier-V and Labouze-E, (2001); European IPPC
Bureau, Spain (2003)- and Aramburu–J.A, (2003). The extraction process removes the
majority of additives. The process is interesting from the economics point of view because it
eliminates thin film distillation and the need for hydrogenation. Both investment and
maintenance costs are low.

The drawbacks of the Interline process are that the feed should not contain polychlorinated
biphenyls (PCBs), and its chlorine content should be below 1000 ppm, since this process has
no final hydrofinishing step.

# 280 3.7 Propak thermal cracking process

The Propak process consists of screening and dewatering sections, followed by a thermal cracking section, a separation or distillation depending on the product state desired and finally purification and stabilization stages. In certain plant configurations, a heavy boiling fraction is recycled back to the fired process heater. Gasoil in the liquid state is led to the stabilization section from distillation.

This technology is characterized by a large operational and product flexibility. Process operating conditions (temperature, pressure, residence time) can be varied to produce a desired product such as heavy fuel oil, gasoil or base oil. (Kupareva *et al.*, 2013)

# 289 4.0 Current Technologies for Used Oil Re-refining

290 Used lube oil normally tends to have a high concentration of potentially harmful pollutant 291 materials and heavy metals which could be dangerous to both living and non-living things on

the earth. Used lube oil may cause damage to the environment when dumped into the ground

Formatted: Font: Italic Formatted: Font: Italic

### Formatted: Font: Italic

or into water streams including sewers. This may result in ground water and soil
contamination- (Hopmans, 1974). Therefore, development of environmentally safe,
sustainable and cost-effective solution is required for recycling of used lubricant. (Stehlik,
2009).

297 Nowadays due to different treatment and finishing methods, there are currently available many new technologies (-Bridjanian, H and Sattarin, M, 2006) such as pyrolytic distillation 298 method (PDM), pyrolysis process (PP), -thin film evaporation (TFE), including combined 299 300 TFE and clay finishing, TFE and solvent finishing, TFE and hydrofinishing, thermal deasphalting (TDA), TDA and clay finishing, -TDA and hydrofinishing etc. In addition, 301 302 environmentally friendly and affordable solvent extraction and adsorbents are being 303 developed as a means of removing contaminants in used lube oil. Some of the current methods are briefly discussed below.; 304

From the research conducted by Arpal *et al.*, (2010), a fuel named as diesel-like fuel (DLF) was produced by applying pyrolytic distillation method. Filtration of the waste engine oil sample was done using a quantitative filter. Three additives known as Na<sub>2</sub>CO<sub>3</sub>, zeolite and CaO were blended with the purified oil at different ratios and were exposed to thermal and pyrolytic treatment to convert them into a diesel-like fuel. Conclusively, effects of DLF on the oil properties shows a closer range to that of diesel fuel. (Temitayo *et al.*, 2018)

311 Also, Pyrolysis process (PP) has been used as an alternative means of effective conversion of used lubricants to a refined one (Lam et al., (2016); and Manasomboonphan and Junyapoon, 312 (2012). Lam et al., (2016), describe pyrolysis as a thermal process that heats and decomposes 313 substance at high temperature  $(300-1000^{\circ}C)$  in an inert environment without oxygen. 314 315 Pyrolysis process is not yet widespread but it has been receiving much attentions nowadays due to its potential to produce energy-dense products from materials. Examples of pyrolysis 316 process includes Microwave Pyrolysis Process (MPP) and Conventional Pyrolysis Process 317 318 (CPP). The MPP is a thermo-chemical process applied to waste to wealth process of electrical 319 power input of 7.5kW at a flow rate of 5kg/h. (Temitavo et al., 2018).

Thin film evaporation technology includes a rotating mechanism inside the evaporator vessel which creates high turbulence and thereby reduces the residence time of feed-stock oil in the evaporator. This is done in order to reduce coking, which is caused by cracking of the hydrocarbons due to impurities in the used oil. Cracking starts to occur when the temperature of the feedstock oil rises above 300°C.

However, any coking which does occur will foul the rotating mechanism and other
mechanisms such as tube-type heat exchangers are often found in thin film evaporators.
Solvent extraction processes are widely applied to remove asphaltic and resinous
components.

Liquid propane is by far the most frequently used solvent for de-asphalting residues to make lubricant bright stock, whereas liquid butane or pentane produces lower grade de-asphalted oils more suitable for feeding to fuel-upgrading units. The liquid propane is kept close to its critical point and, under these conditions, raising the temperature increases selectivity. A temperature gradient is set up in the extraction tower to facilitate separation. Solvent-to-oil ratios are kept high because this enhances rejection of asphalt from the propane/oil phase. Formatted: Font: Italic
Formatted: Font: Italic
Formatted: Font: Italic

Formatted: Font: Italic

Formatted: Font: Italic

Formatted: Superscript

Counter-current extraction takes place in a tall extraction tower. Typical operating conditions can be found in the work by (Mortier and Fox, 2010).

Recent studies showed that propane can be replaced by an alcohol-ketone mixture, which 337 338 reduces coking and fouling problems during distillation. (Kamal and Khan, 2009 & Durrani et al., 2010). The solvent chosen should meet the following requirements: Mmaximum 339 solubility for the oils and minimum solubility for additives and carbonaceous matter; ability 340 to be recovered by distillation. New plant units increasingly use N-methylpyrrolidone 341 because it has the lowest toxicity and can be used at lower solvent/oil ratios, saving energy. 342 Independent of the contacting method used, the end result is two product streams. The 343 344 raffinate stream is mainly extracted oil containing a limited amount of solvent, while the 345 extract stream is a mixture of solvent and aromatic components. The streams are handled separately during solvent recovery and the recovered solvent streams are recombined and 346 347 recycled within the plant.

However, solvent recovery is an energy-intensive part of the solvent extraction process. For
several years, catalytic hydrotreatment stood out as the modem and successful refining
treatment from the point of view of the yield and quality of the finished products.
Hydroprocessing is more often applied as a final step in the rerefining process in order to
correct problems such as poor colour, oxidation or thermal stability, demulsification and
electrical insulating properties... (Kupareva et al., 2013).

In hydrofinishing, used oil and hydrogen are pre-heated and then oil allowed to trickle
downwards through a reactor filled with catalyst particles where hydrogenation reactions take
place. The oil product is separated from the gaseous phase and then stripped to remove traces
of dissolved gases or water. Typical reactor operating conditions for hydrofinishing can be
found (Mortier and Fox, 2010).

The following reactions can be operative: <u>Hhydrorefining</u> reactions with the objective of removing heteroelements and to hydrogenate olefinic and aromatic compounds, and hydroconversion reactions aiming at modifying the structure of hydrocarbons by cracking and isomerization- (Audibert, 2006).

Hydrotreatment catalysts are made of an active phase constituted by molybdenum or tungsten 363 364 sulfides as well as by cobalt or nickel on oxide carriers. Generally applied combinations are Co-Mo, Ni-Mo, and Ni-W for the active phase and high surface area  $\gamma$ -alumina (transition 365 alumina) carrier. The metal content, expressed as oxides can reach 12-15 wt. % for Mo and 366 3-5 wt. % for Co or Ni. Co-Mo catalysts are preferentially used for hydrodesulphurization 367 and Ni- Mo for hydrogenation and hydrodenitrogenation. Ni-W catalysts are applied for low-368 sulphur feeds. The most-used carriers are alumina and alumina-silica, the latter being 369 characterized by a higher cracking activity- (Audibert, 2006).-370

The currently applied catalysts in rerefining are modified in order to improve the-product base oil quality and to decrease the coke formation, however, their composition is typically not disclosed in an open literature. The technologies applying hydroprocesses require relatively high investments compared with others. However, depending on the technology adopted, the total cost might be lower than in solvent extraction process due to the high operating costs to make up for the solvent losses. On the other hand, solvent extraction and chemical treatment processes do not require catalyst regeneration. Moreover, it is not

378 necessary to establish a hydrogen gas supply facility in these methods which in addition

reduces a risk concerning operation safety. (Kupareva *et al.*, 2013).

# 380 5.0 Conclusion

Currently applied technologies can be compared in terms of their operating and capital costs, 381 382 quality of feedstock and products obtained. These advance combine technology processes 383 and/or methods are mainly found in developed countries but not available in developing countries. These methods when applied generates reduced concentrations of pollutant but 384 require complex and expensive equipment which are rarely found in developing countries. 385 Under increasing environmental pressure of the conventional treatment method such as acid-386 387 clay treatment, which was the first oil regeneration process used, it was substituted in the 388 majority of European countries with new technologies based on solvent extraction, pyrolysis, 389 membrane etc. The modern technologies based on solvent extraction, pyrolysis, membrane etc. are environmentally controllable but their operating and capital costs are high, low yields 390 391 and requires highly skilled personnel (-compared to conventional method) is the major drawback. Also, the challenge of cost reduction resulting from the vacuum distillation and 392 hydroprocessing technique. The combined treatment methods have shown remarkable well 393 394 with high treatment efficiency, environmentally friendly. However, the problem of high cost and season skilled operating personnel remains a major gap in used lube oil treatment. 395 Therefore, there is the need to developed viable, efficient, environmentally friendly, 396 397 affordable treatment and high yield technique such as solvent extraction coupled with adsorption process to remove contaminants in used lube oil. 398

399

408

409

410

# 400 References

- Abu-Elella1, R.; Ossman, , M.E.; Farouq, R.; Abd-Elfatah, M.(2015): Used Motor
   Oil Treatment: Turning Waste Oil Into Valuable Products, *International Journal of Chemical and Biochemical Sciences*, 7, 7, pp:57-67
- AERCO Inc. P.S. (1995); Solvent extraction technology for used oil treatment. Final
   report for Recycling Technology Assistance, 30p.
- Aramburu J.A, (2003); Rerefining used oils by propane extraction: a proven
   technology. Ing Quim (Madrid, Spain) Special Issue: 55–61.
  - 4. Aremu MO, Araromi DO, Gbolahan O.O; (2015). Regeneration of Used Lubricating Engine Oil by Solvent Extraction Process. International Journal of Energy and Environmental Research; Vol.3, No. 1, pp.1-12.
- Arpal, O.; Yumrutas, R.; Demirbas, A. ;( 2010). Production of diesel-like fuel from
  waste engine oil by pyrolytic distillation. Appl Energy; 87: 122-127
- 413 6. Audibert, F., (2006); Waste Engine Oils. Elsevier Science, Netherlands.
- Basel Convention, (2002); Technical Guidelines on Used Oil Re-Refining of Other
   Re-Uses of Previously Used Oil, series/SBC No.02/05, ISBN: 92-1-158605-4
- 8. Bridjanian H and Sattarin M, (2006); Modern recovery methods in used oil
  rerefining. Petrol Coal 48:40–43.
- 9. Brinkman D.W. (2010); Kirk-Othmer Encyclopedia of Chemical Technology, John
  Wiley & Sons, Inc.
- 10. Chari K.R, (2012). Compendium of recycling and destruction technologies for waste
   oils,UNEP,Japan,Exform;http://www.unep.org/ietc/Portals/136/Publications/Waste%

- 422 20Management/IETC%20Waste\_Oils\_Compendium-Full%20Doc-
- 423 for%20web\_Nov.2012.pdf
- 424 11. Dang, C.S. (1997); Re-refining of used oils A review of commercial processes.
   425 *Tribotest* 3, 445-457.
- 12. Durrani H.A, Panhwar M.I and Kazi R.A, (2010); Rerefining of waste lubricating oil
  by solvent extraction. Mehran UnivRes J Eng Technol 30:237–246.
- 428 13. Eman A, Emam, Abeer M. Shoaib. Re-refining of Used Lube Oil, II- by Solvent/Clay
  429 and Acid/Clay-Percolation Processes. ARPN Journal of Science and Technology
  430 2012; Vol 2 [11] 1034- 1041.
- 431 14. Emetere, M.E.,(2017): Investigations on aerosols transport over micro and macro
  432 scale settings of West Africa., *Environmental Engineering Research*;Vol.22(1),
  433 pp.75-86
- 434 15.
- 435 16. Falah Bani Hani and Hussien Al-Wedyan. (2011). Regeneration of Base-Oil from
  436 Waste-Oil under Different Conditions and Variables. *African Journal of*437 *Biotechnology*, 1150-1153.
- 438 17. Fox, M.F. (2007); Sustainability and environmental aspects of lubricants. In
  439 *Handbook of Lubrication and Tribology*, George, E.D., Totten, E., Eds.; Taylor and
  440 Francis: New York, NY, USA.
- 441 18. Giovanna FD, Khlebinskaia O, Lodolo A, Miertus S, (2003) Compendium of used oil
   442 regeneration technologies. UNIDO, Trieste. Exform http://institute.unido.org
- 443 19. Giovanna, F. Dalla; Khlebinskaia, A. Lodolo; Miertus, S. (2012). Compendium of
  444 Used Oil Regeneration Technologies. Trieste: United Nations Industrial development
  445 Organization
- 446 20. Guerin Turlough, F. (2008): Environmental liability and life-cycle management of
  447 usedlubricating oils, *journal of Hazardous Materials*; 160, pp.256-264
- 448 21. Hamawand,I.; Talal, Yusaf and Sardasht Rafat. (2013). Recycling of Waste Engine
  449 Oils Using a New Washing Agent. Energies, pp. 1023-1049.
- 450 22. Hassan Ali Durrani, Muhammed Ibrahim Panhwar, Rafique Akthar Kazi. (2012)
  451 Determining an Efficient Solvent Extraction Parameters for Re-Refining of Waste
  452 Lubricating Oils. *Journal of Engineering & Technology*; Volume 30, NO. 2, 265-270
- 453 23. Hassan Ali Durrani, (2014). Re-Refining Recovery Methods of Used Lubricating Oil;
   454 International Journal of Engineering Sciences & Research Technology; Vol. 3(3); pp.
   455 1216-1220
- 456 24. Havemann, R. (1978); The KTI used oil re-refining process. In Proceedings of the
  457 3rd International Conference of Used Oil Recovery & Reuse, Houston, TX, USA,
  458 16–18.
- 459 25. Hopmans, J.J.(1974); The problem of Processing of Spent Oil in the Member States
  460 of EEC; Report for the European Economic Community(EEC); National Institute for
  461 Wastewater Treatment: Dordrecht, The Netherlands
- 462 26. Hussein M, Amer AA, Gaberah AS, (2014). Used Lubricating Oils Re-Refining by
  463 Solvent Extraction. American Journal of Environmental Engineering and Science;
  464 Vol. 1, No. 3, pp. 44-50.
- 465 27. Integrated pollution prevention and control. Best available techniques for the waste
   466 treatments industries, European IPPC Bureau, Spain (2003).

- 28. Isah, A. G. (2013). Regeneration of Used Engine Oil. Proceedings of the World 467 Congress on Engineering, Vol I. London: World Congress on Engineering 468
- 469 29. Jafari, A.J., Hassanpour, M. (2015), Analysis and comparison of used lubricants, regenerative technologies in the world. Resour. Conserv. 470 Recy., http://dx.doi.org/10.1016/j.resconrec.2015.07.026 471
- 30. Kajdas, C, (2000); Major pathways for used oil disposal. Part 1. Exform http:// 472 473 www.tribologia.org/ptt/kaj/kaj10.htm.
- 31. Kalnes T.N, Shonnard D.R and Schuppel A. (2006); LCA of a spent lube oil re-474 475 refining process. 16th European Symposium on Computer Aided Process Engineering and 9th International Symposium on Process Systems Engineering. 476 Elsevier BV, 713-718. 477
- 32. Kalnes TN and Schuppel A. (2007); Hylube process commercialization: recovering 478 value from used motor oil. AICHE Meeting, Houston, Texas. Exform 479 480 http://www.aiche-fpd.org/listing/HP7.pdf

481

504 505

- 33. Kamal A and Khan F. (2009); Effect of extraction and adsorption on rerefining of used lubricating oil. Oil Gas Sci Technol 64:191-197. 482
- 483 34. Kanokkantapong, V.; Kiatkittipong, W.; Panyapinyopol, B.; Wongsuchoto, P.; Pavasant, P.(2009) : Used lubricating oil management options based on life cycle thinking; 484 485 Resources, Conservation and Recycling, Vol. 53 (5), pp. 294-299
- 35. Kannan, S. C., Mohan Kumar, K. S., Sakeer Hussain, M., Deepa Priva, N. K. (2014). 486 Studies on reuse of refined used automotive lubricant oil, Research Journal of 487 488 Engineering Science, 3(6), 8-14.
- 36. Katiyar, V., Husain, S., (2010). Reclamation of used lubricating oils. Curr. World 489 490 Environ. 5 (1), 79-84
- 491 37. Ketlogetswe, C., (1998). Management of waste oil In Botswana and the possibilities of energy recovery from waste oil. Botswana J. Technol., pp.11-18 492
- 493 38. Kupareva, A.; M"aki-Arvela, P.; Murzin, D.Y. (2013); Technology for rerefining used lube oils applied in Europe; Society of Chemical Industry; J Chem Technol 494 Biotechnol, 88: 1780-1793 495
- 39. Lam, S.S.and Chase, H.A.(2012) : A review on waste to energy processes using 496 microwave pyrolysis, Energies, Vol.5, pp. 4209-4232 497
- 40. Lam, S.S; Liewa, K.K; Jusoh, A.A; Chong, C.T; Ani, F.N; Chase, H.A. (2016). 498 Progress in waste oil to sustainable energy, with emphasis on pyrolysis techniques; 499 Renewable and Sustainable Energy Reviews, Vol. 53; pp. 714-753. 500
- 41. Lukic J, Orlovic A, Spiteller M, Jovanovic J and Skala D, (2005); Rerefining of 501 waste mineral insulating oil by extraction with N-methyl-2- pyrrolidone. Sep 502 503 PurifTechnol 51:150-156.
  - 42. Magnabosco L.M; and Rondeau W.A, (1993); Improved process for the production of base stock oils from used oil. Patent EP 0574272A2.
- 506 43. Manasomboonphan, W., and Junyapoon, S.; (2012). Production of liquid fuels from 507 used lube oils.
- 44. Martins, J.P. (1997); The extraction-flocculation re-refining lubricating oil process 508 using ternary organic solvents. Ind. Eng. Chem. Res., 36, 3854-3858. 509
- 45. Merchaoui, M, H; Khalef, N; Jaafar, A; Ouazzane A, Boufahja MA and Meziou S, 510 (1994); Process and plant for regeneration of lubricating oils. Patent WO9421761 511 512 (A1).

- 46. Monier V and Labouze E, (2001); Critical review of existing studies and life cycle
  analysis on the regeneration and incineration of waste oils. European Commission.
  DG Environment A2 Sustainable Resources Consumption and Waste, France.
  Exform <u>http://ec.europa.eu/environment/waste/studies/oil/waste\_oil.pdf</u>
- 517 47. Mortier RM and Fox M.F. (2010); Chemistry and Technology of Lubricants.
  518 Springer, Dordrecht
- 48. Nwachukwu, M. A.; Alinnor, J. and Huan Feng. (2012). Review and assessment of
  mechanic village potentials for small scale used engine oil recycling business. *African Journal of Environmental Science and Technology* Vol. 6(12), pp. 464-475,
  464-475.
- 49. Oladimeji, Temitayo, E; Sonibare, Jacob A.;Omoleye, James, A; Adegbola, Abiola
  A.; Okagbue, Hilary I.(2018). Data on the treatment of used lubricating oil from two
  different sources using solvent extraction and adsorption. *Data in Brief*, 19, pp.
  2240–2252
- 527 50. Oladimeji, Temitayo, E.; Sonibare, Jacob, A.; Omoleye, James, A; Emetere, Moses,
  528 E.; Elehinafe, Francid, B; (2018); A Review on Treatment Methods of Used
  529 Lubricating Oil, *International Journal of Civil Engineering and Technology*, 9(12),
  530 pp.506-514.
- 531 51. P"ohler, J.,;M"odler, M.; Bruhnke, D,; Hindenberg, H. (2004); Method for
  532 reprocessing waste oils. US Patent 6712954 B1.
- 533 52. Puerto-Ferre, E.; Kajdas, C. (1994); Clean technology for recycling waste lubricating
  534 oils. In Proceedings of 9th International Colloquium, Ecological and Economic
  535 Aspects of Tribology, Esslingen, Germany, 14–16.
- 536 53. Princewill Nemibo Josiah, Sunday Sunday Ikiensikimama. (2010). The Effect of
  537 Desludging and Adsorption Ratios on the Recovery of Low Pour Fuel Oil (LPFO)
  538 from Spent Engine Oil. *Chemical Engineering Research Bulletin*, pp.25-28.

539

540

543

544

545

546

547

548

549

550 551

552

- 54. Quang, D.V.; Carriero, G.; Schieppati, R.; Comte, A.; Andrews, J.W.(1974) Propane purification of used lubricating oils. *Hydrocarb. Process*, 53, 129–131.
- 55. Rincon, J.; Canizares, P.; Garcia, M.T.; Gracia, I. (2003) Regeneration of used
  lubricant oil by propane extraction. *Ind. Eng. Chem. Res.* 42, 4867–4873.
  - 56. Rincon, J.; Canizares, P.; Garcia, M.T. (2005). Regeneration of used lubricating oil by polar solvent extraction. *Ind. Eng. Chem. Res.*, Vol. 44, pp.43-73
  - 57. Rincon, J.; Canizares, P.; Garcia, M.T. (2005); Waste oil recycling using mixtures of polar solvents. *Ind. Enf. Chem. Res*, 44, 7854-7859
    - 58. Schiessler, N.; Thorpe, E.; Jones, W.; Philips, L.; DOL-E.L (2007): Recovery of base oil fractions from used oil lubricants. LIFE and Waste Recycling: Innovative Waste Management Options in Europe III: 28
  - Shakirullah, M.; Ahmed, I.; Saeed, M.; Khan, M.A., Rehman, H.; Ishaq, M.; Shah, A.A. (2006); Environmentally friendly recovery and characterization of oil from used engine lubricants. J. Chin. Chem. Soc., 53, 335–342
- 60. Shri Kannan, C.; Mohan Kumar, K.S.; Sakeer Hussain, M.; Deepa Priya, N.;
  Saravann, K., (2014): Studies on Reuse of Re-Refined Used Automotive Lubricating
  Oil. Research Journal of Engineering Sciences; Vol.3(6), pp. 8-14
- 556 61. Sotulub re-refining process(2005).Exform http://www.investintunisia.tn/
   557 document/307.pdf

558	2. Stehlik, P. (2009); Contribution to advances in waste-to-energy technologies. J	
559	Clean Prod; 17; 919-931	

- 560 63. Sterpu AE, Dumitru AI, Popa MF.(2012) Regeneration of used engine lubricating oil
  561 by solvent extraction, J Ovidius University Annuals Chem.;23:(2):149-54.
- 563 64. Tsai, W.T. (2011): An analysis of used lubricant recycling, energy utilization and its
  564 environmental benefit in Taiwan; *Energy* Vol.36(7), pp. 4333-4339
- 565 65. Tsalavoutas, S., Kapoutsis, G.; Loukas, L. (2002). Survey of the Greek recycling
  566 sector. Greek sector study report, Athens, Exform http://www.biat.uni567 flensburg.de/biat/Projekte/Recy-Occupation/ Sector Study Greece.doc
- 568 66. Udonne, J. D., and Bakare, O. A.; (2013). Recycling of Used Lubricating Oil Using
  569 Three Samples of Acids and Clay as a Method of Treatment. *International Archive of*570 *Applied Sciences and Technology*, 4(2), pp.8–14.
- 67. Stoll, U. and Gupta, H.( January 1997) "Management strategies for oil and grease
  residues," Waste Management Resources, vol. 15, no. 1, pp. 23-32,
- 573

562