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A Review of Methods for Removal of Contaminants in Used Lubricating Oil

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Authors' contributions

This work was carried out in collaboration among all authors. Author KOB designed the study, performed the statistical analysis, wrote the protocol and wrote the first draft of the manuscript. Authors OFJ and DKE managed the analyses of the study. Author BOE managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

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 have bio-accumulation effects on living organisms, reduces the inhabitants lifespan as a result of the diseases spread, poisoning and fouling of catalyst as well as corrode processing equipment. The contaminants removal in used lubricating oil is a major step to avoid pollution as discussed thoroughly by many researchers in literature. In addition, to curbing pollution, another advantage is converting 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 earmarked for further study.

Keywords: Used lubricatingoil; contaminants; removal methods; treatment.

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1. INTRODUCTION

Lubricating oils (LOs) are conventionally obtained from crude oil. Chemical composition of LOs consists on average about 80–90% base oil and about 10–20% chemical additives and other compounds [1]. Lubricating oils mainly helps in reducing friction, dust, corrosion, protection against wear and tear and provision of heat transfer medium in various equipment or machineries [2]. During operation time, LOs deteriorate, as well as their additives, and its physical and chemical properties become unsuitable for further use [3]. In the process of lube oil usage, temperature build up occurs which breaks down oil and weakens its properties which include pour point, flash point, specific gravity, viscosity etc. [4]. These renders the oil unsuitable for regular usage as results of contaminants in lube oil such as water, wear metals, carbon residue, ash content, gums, varnishes etc. Chemical changes in oil 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 [5].

Used lubricating oils (ULOs) are classified as hazardous wastes, and constitute a serious pollution problem not only for environment, but also for human health due to harmful contaminants presence, such as heavy metals, polychlorinated biphenyls (PCBs) and polycyclic aromatic hydrocarbons (PAHs) [6]. Poor management and careless disposal of used lube oil can affect the environment negatively [7,8]. Scientists have reported that in some geographical region e.g. West Africa, the dispersion of air pollutants could travel at a speed of 10-12 m/s [9]. The implication of this report is that air pollution from burning of waste lubricant is not localized to the pollution source but could travel with time to other locations. For example, it was recently reported that black soot covered a metropolitan city of Port Harcourt while remote sources were at the suburb settlement (about 22 km away from the city) [10].

On the other hand, ULOs can be considered as valuable resources, in the sense that it is possible to recover energy or profitable materials for further use [11]. The best 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 a variety of engine configurations and other applications. Its gross calorific value is greater than 42.9 MJ/kg [12]. The principal objective of any waste management plan is to ensure safe, efficient and economical collection, transportation, treatment and waste disposal and as well as satisfactory operation for current and foreseeable future scenarios [13]. The treatment of used lube oil is important due to: (1) It requires less energy and cost compared to conventional refining of crude oil ;(2) It helps in improving air quality, land and water pollution in the environment. The most preferred option by experts is the reuse of used lube oil generated by consumers [14]. In this paper, a thorough review of various removal and treatment methods for used lube oil would be considered starting from conventional to most current methods and their limitations, further developments of these fields were also touched. In addition, environmentally friendly and affordable solvent extraction and adsorbents would be developed as a means of removing contaminants in used lube oil.

2. CONVENTIONAL METHODS

The conventional methods of contaminants removal in used lube oil either requires a high cost technology such as vacuum distillation or use of toxic materials such as sulphuric acid. These methods also produce contaminating by-products which have highly sulphur levels, especially in Kurdistan region/Iraq [15].

2.1 Acid-clay

Assessment of different contaminants removal processes in used lube oils revealed that acid-clay process had the highest environmental risk and lowest cost. The method involve treatment of used oil with acid and clay [4,15,16]. They all used the clay as an adsorbent to remove the odour and dark colour. What makes acid-clay method unique from others are; with its simple method, affordable capital investment, low operating cost and does not need skilled operators [17-19].

However, this method has many disadvantages: It also produces large quantity of pollutants, is unable to treat modern multigrade oils and it's difficult to remove asphaltic impurities [20]. To reduce these hazardous contaminants from this method, the acid treatment stage of the process can be done under the atmospheric pressure to remove the acidic products, oxidized polar compounds, suspended particles and additives [21]. Princewill and Sunday [22] observed that high recovery rate of treated lube oil from used lube oil depend largely on the source of used lube oil, pre-treatment mechanisms, extent of contamination and the grade of acid used. He also showed that the volume of adsorbent (clay) used could affect the rate at which contaminants are removed and recovery percentage of the method.

In Abu-Elella [16] worked on used motor oil. He treated used motor oil with phosphoric acid, sulphuric acid, methanoic acid and acetic acid. He observed that methanoic acid, sulphuric acid and acetic acid have great changes on kinematic viscosity while phosphoric acid is not affected by used lube oil. He therefore concluded that treatment with acetic acid showed better results than formic acid-clay.

2.2 Solvent Extraction

This method has replaced acid-clay treatment as preferred method for improving oxidative stability and viscosity as well as temperature characteristics of 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 [23]. The solvent selectively dissolves the undesired aromatic components (the extract), leaving desired saturated components, especially alkanes, as a separate phase (the raffinate) [1].

Different solvents types have been used for solvent extraction such as 2-propanol, 1-butanol, methyl ethyl ketone (MEK), ethanol, toluene, acetone, propane etc. [24,25] used propane as solvent. He found out that propane was capable of dissolving paraffinic or waxy material and intermediately dissolved oxygenated material. Asphaltenes which contain heavy condensed aromatic compounds and particulate matter are insoluble in liquid propane. These properties make propane ideal for recycling the used engine oil, but there are many other issues that have to be considered. Propane is hazardous and flammable therefore this process is regarded as hazardous method.

Katiyar and Husain [26]; Sterpu [27] and Hassan [28] found out that methyl ethyl ketone has the highest performance due to its low oil percentage losses and high sludge removal while [29,30] found out extraction using butan-1-ol solvent produces the highest sludge removal rate. Rincon [1] and Oladimeji [31] 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 produces remarkable amounts of hazardous by-products [15,24,25].

Mineral ¨ol Raffinerie Dollbergen (MRD) solvent extraction process using N-methyl-2-pyrrolidone. The applied oil re-refining process is based on a patent held by AVISTA OIL [32]. The 'Enhanced Selective Refining' process uses solvent N-methyl-2-pyrrolidone (NMP), which is commonly used in petroleum refining industry. NMP is a powerful, aprotic solvent with low volatility, which shows selective affinity for unsaturated 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-refining. The NMP advantages over other solvents are non-toxic nature and high 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 aromatic hydrocarbons and PAH, NMP can be used for re-refining of waste oils with lower sludge, carbonaceous particles and polymer contents, such as waste insulating, hydraulic and other similar industrial oils [33]. The MRD solvent extraction process uses the liquid–liquid extraction principle.

The average base oil yield within the process is about 91% [34]. The base oils produced have high quality [5]. The process is characterized by optimized operating conditions which allow elimination of toxic polyaromatic compounds from the re-refined base oil and preservation of synthetic base oils like polyalphaolefin (PAO) or hydrocracked oils, which are increasingly present in used oils. However, this method need skilled personnel, proper disposal and management of it waste.

2.3 Vacuum Distillation

Extensive research work have been done on vacuum distillation on used oil by the following

[15,35-39]. In this method, used 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 of 240°C and pressure 20 mmHg. This results the production of a light fuel oil at a temperature of 140^0C (the light fuel oil can be used as fuel source for heating) and lubricating oil at 240°C. The lubricating oil vapour is condensed and sent for next stage [39]. The advantages of vacuum distillation process over atmospheric pressure distillation are: Columns can be operated at lower temperatures; more economical to separate high boiling point components under vacuum distillation; avoid degradation of properties of some species at high temperatures therefore thermally sensitive substances can be processed 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. The residue is in the form similar to that of tar, which can be used as a construction material, for example, road and bitumen production [40]. The disadvantage of this method is the high investment cost and/or the use of toxic materials such as sulphuric acid [41-42].

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 [37]. 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 [31].

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 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 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 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 [28].

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 [43].

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)] [8] were used for recycling the used engine oils. The process is carried out at 40^0C 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 [15,44].

Despite the above mentioned advantages, the expensive membranes may get damaged and fouled by large particles with time [15,44].

2.6 Catalytic Process

For example, Hylube process from Germany. This process allows production of mainly base oils. The Hylube process is a proprietary process developed by Universal Oil Products (UOP) for catalytic processing of used lube oils into re-refined lube base stocks for re-blending into saleable lube base oils [45]. 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 contaminants such as zinc, phosphorous, and calcium [46].

The Hylube unit operates with reactor section pressures of 60–80 bar and reactor temperatures in range 300–350°C [47]. The Hylube process achieves more than 85% of lube oil recovery from the lube boiling range hydrocarbon in feedstock [5]. Besides the advantages of these process, this method is very expensive. This method requires high level personnel due to high temperature and pressure operations.

3. COMBINED TECHNOLOGIES/ METHODS

These are advance methods that combines two or more generic methods in its process. Due to the complex nature of contaminants removal in used lube oils, using a single method may not give you the desired standard emission-controlled process. Therefore, some companies have developed specific processes for treatment and contaminants removal in used lube oils [5,43,48] these methods require sophisticated technologies, equipment and processes. Some of these complex processes are briefly discussed below.

3.1 Vaxon Process

This process contains chemical treatment, vacuum distillation and solvent refining units. The advantage of Vaxon process is the special vacuum distillation, where the cracking of oil is strongly decreased [46].

The chemical final stage does not, however allow the high-quality base oils production; although in Spain the Catalonia refinery produces base stocks accepted by an original equipment manufacturer (OEM). In connection with this fact, the lube distillate obtained from Vaxon process (Denmark) or North Refining (Netherlands) are precursors for Avista Oil base [5].

3.2 Chemical Engineering Partners (CEP) Process

This process combines thin film evaporation and hydroprocessing. The used oil is chemically 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 chemical treatment compound comprises sodium hydroxide, which is added in a sufficient amount to give a pH about 6.5 or higher [49].

Heavy materials (Metals, additive degradation products, etc.) are passed to a heavy asphalt flux stream. The distillate is hydropurified at high temperature (315°C) and pressure (90 bar) in a catalytic fixed bed reactor [50]. This process removes nitrogen, sulphur, chlorine and oxygenated organic components. In final stage of process, three hydrotreating (Hydrofinishing) reactors are used in series to reduce sulfur to less than 300 ppm and to increase the number of saturated compounds to over 95%, in order to meet the key specifications for API Group II base oil. The final step, in this process is vacuum distillation to separate the hydrotreated base oil into multiple viscosity cuts in fractionator. The yield of base oils is about 70% [5].

3.3 Ecohuile Process

The re-refining process was based on vacuum distillation and acid-clay treatment steps until the end of 2000 [51]. Clay adsorption was banned on 1 January 2001 and the plant was modified and upgraded to the Sotulub process [52]. Moreover, the addition of injection facilities of so-called Antipoll-additive (1–3 wt% of pure sodium hydroxide) has been provided and has allowed solving the following basic problems:

- Corrosion of dehydration column and cracking column top section due to organic acidity of used oil.
- Plugging of equipment and piping due to polymer formation in cracking section.
- High losses of base oil in oily clay due to high consumption of clay.

The Sotulub process [50] is based on treatment of used oil with 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 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 oil cuts. The process provides base oils with a yield of 82–92% [52].

3.4 Cyclon Process

This process combines the technology of vacuum distillation and hydrofinishing [41]. The process licence belongs to Kinetic Technology International (KTI) [53]. In this process, used oils taken from storage tanks are dewatered and the light hydrocarbons are removed by distillation. The heavier fraction is sent to high vacuum distillation, where the majority of base oil components are evaporated from the heavy residue. The oils in residues are extracted with propane in de-asphalting unit and sent to hydroprocessing unit where other oils are processed. Then they are treated with hydrogen and fractionated based on desired base oil features. The re-refined base oil products obtained are of high quality due to hydrogenation [34,54].

3.5 Studi Tecnologie Progetti S.p.A. (STP) Method

This is another advance method that combines vacuum distillation and hydrofinishing process [43]. It produces less harmful pollutants therefore its environmentally friendly [5]. This method involves dehydration, vacuum distillation, separation of lubricating fraction and hydrofinishing of base oil separation from residue.

3.6 Interline Process

Interline proposes a process based on propane de-asphalting at ambient temperature and under a pressure that facilitates separation in liquid phase. The lubricating oil yield declared for Interline process is 79% [55-57]. The extraction process removes the majority of additives. The process is interesting from economics point of view because it eliminates thin film distillation and need for hydrogenation. Both investment and maintenance costs are low.

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

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 product state desired and finally purification and stabilization stages. In certain plant configurations, a heavy boiling fraction is recycled back to fired process heater. Gasoil in liquid state is led to 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 [5].

4. MODERN TECHNOLOGIES FOR USED OIL RE-REFINING

Used lube oil normally tends to have a high concentration of potentially harmful pollutant 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 environment when dumped into ground or into water streams including sewers. This may result in ground water and soil contamination [58]. Therefore, development of environmentally safe, sustainable and cost-effective solution is required for recycling of used lubricant [59].

Nowadays due to different treatment and finishing methods, there are currently available many new technologies,[37] such as pyrolytic distillation method (PDM), pyrolysis process (PP), thin film evaporation (TFE), including combined TFE and clay finishing, TFE and solvent finishing, TFE and hydrofinishing, thermal de-asphalting (TDA), TDA and clay finishing, TDA and hydrofinishing etc. In addition, environmentally friendly and affordable solvent extraction and adsorbents are being developed as a means of removing contaminants in used lube oil. Some of current methods are briefly discussed below.

From research conducted by Arpal [60], a fuel named as diesel-like fuel (DLF) was produced by applying pyrolytic distillation method. Filtration of waste engine oil sample was done using a quantitative filter. Three additives known as Na_2CO_3, zeolite and CaO were blended with 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 [31].

Also, Pyrolysis process (PP) has been used as an alternative means of effective conversion of used lubricants to a refined one [8,61]. Lam [8], describe pyrolysis as a thermal process that heats and decomposes substance at high temperature (300-1000°C) in an inert environment without oxygen. 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 process includes Microwave Pyrolysis Process (MPP) and Conventional Pyrolysis Process (CPP). The MPP is a thermo-chemical process applied to waste to wealth process of electrical power input of 7.5kW at a flow rate of 5kg/h [31].

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 evaporator. This is done in order to reduce coking, which is caused by cracking of hydrocarbons due to impurities in used oil. Cracking starts to occur when temperature of feedstock oil rises above 300°C.

However, any coking which does occur will foul 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 extraction tower to facilitate separation. Solvent-to-oil ratios are kept high because this enhances rejection of asphalt from propane/oil phase. Counter-current extraction takes place in a tall extraction tower. Typical operating conditions can be found in the work by [62].

Recent studies showed that propane can be replaced by an alcohol–ketone mixture, which reduces coking and fouling problems during distillation [63-65]. The solvent chosen should meet the following requirements: Maximum solubility for the oils and minimum solubility for additives and carbonaceous matter; ability to be recovered by distillation. New plant units increasingly use N-methylpyrrolidone because it has the lowest toxicity and can be used at lower solvent/oil ratios, saving energy. Independent of contacting method used, the end result is two product streams. The raffinate stream is mainly extracted oil containing a limited amount of solvent, while the extract stream is a mixture of solvent and aromatic components. The streams are handled separately during solvent recovery and recovered solvent streams are recombined and recycled within the plant.

However, solvent recovery is an energy-intensive part of solvent extraction process. For several years, catalytic hydrotreatment stood out as modem and successful refining treatment from the point of view of yield and quality of finished products. Hydroprocessing is more often applied as a final step in rerefining process in order to correct problems such as poor colour, oxidation or thermal stability, demulsification and electrical insulating properties [5].

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 gaseous phase and then stripped to remove traces of dissolved gases or water. Typical reactor operating conditions for hydrofinishing can be found [62].

The following reactions can be operative: Hydrorefining reactions with 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 [51].

Hydrotreatment catalysts are made of an active phase constituted by molybdenum or tungsten sulfides as well as by cobalt or nickel on oxide carriers. Generally applied combinations are Co-Mo, Ni-Mo, and Ni-W for active phase and high surface area γ-alumina (transition alumina) carrier. The metal content, expressed as oxides can reach 12–15 wt.% for Mo and 3–5 wt.% for Co or Ni. Co-Mo catalysts are preferentially used for hydrodesulphurization and Ni- Mo for hydrogenation and hydrodenitrogenation. Ni-W catalysts are applied for low-sulphur feeds. The most-used carriers are alumina and alumina-silica, the latter being characterized by a higher cracking activity [51].

The currently applied catalysts in rerefining are modified in order to improve 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 technology adopted, the total cost might be lower than in solvent extraction process due to the high operating costs to make up for solvent losses. On other hand, solvent extraction and chemical treatment processes do not require catalyst regeneration. Moreover, it is not necessary to establish a hydrogen gas supply facility in these methods which in addition reduces a risk concerning operation safety [5].

In this paper, a thorough review on various removal and/or treatment methods for used lubricating oil were considered starting from conventional to the most current methods and their limitations; further developments of these fields were also touched. A gap was identify after indebt discussions of existing methods. As a result of the percentage yield, waste disposal, cost of processing, skilled personnel, environmental compliance etc. Therefore, it would be necessary to develop environmentally friendly and affordable solvent extraction and adsorbents method as a means of removing contaminants in used lubricating oil.

5. CONCLUSION

Currently applied technologies can be compared in terms of their operating and capital costs, quality of feedstock and products obtained. These advance combine technology processes 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 require complex and expensive equipment which are rarely found in developing countries. Under increasing environmental pressure of conventional treatment method such as acid-clay treatment, which was the first oil regeneration process used, it was substituted in majority of European countries with new technologies based on solvent extraction, pyrolysis, 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 and requires highly skilled personnel (compared to conventional method) is the major drawback. Also, the challenge of cost reduction resulting from vacuum distillation and hydroprocessing technique. The combined treatment methods have shown remarkable well 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. Therefore, there is the need to developed viable, efficient, environmentally friendly, affordable treatment and high yield technique such as solvent extraction coupled with adsorption process to remove contaminants in used lube oil.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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