

# A Review on Biodegradable Polymeric Materials Derived from Vegetable Oils for Diverse Applications

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**Abstract:** Non-biodegradable polymers are causing severe damage to the environment. To counter this, need of biodegradable polymers is gaining a rapid growth in numbers as well in their applications and quantities used. In addition, due to the oil crises worldwide, focuses from synthesizing polymers from petroleum has shifted to the synthesis of polymers from renewable resources such as vegetable oils. The advantage of vegetable oils is they are easily available, have multiple functional groups attached like the double bonds, hydroxyl groups and other functional groups, which provides ease of reactions and modification like epoxidation etc. The use of vegetable oils in the polymer renders biodegradability to the polymer. Different researchers have used different vegetables oils like castor oil, linseed oil, soybean oil, sunflower oil and canola oils for synthesis of polymers and utilized them in diverse fields like biomedical, coatings, adhesives, IPN's and as structural materials. This review discusses the synthesis, characterization, degradation and applications of polymers obtained from natural oils such as castor oil, soybean oil, linseed oil and some other oils.

**Keywords:** Renewable sources; Triglyceride oils; Modified oils; Biodegradable polymers

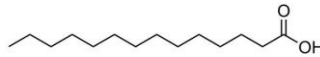
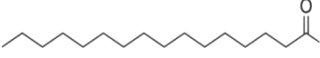
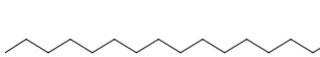
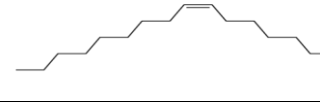
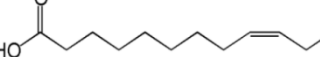
## 1. Introduction

In the history of humankind, people have always named various times as different eras according to the material used like the Stone Age, the Bronze Age and the Iron Age. Therefore in 1995, Griskey<sup>1</sup> named the present time "Polymer Age". All the applications such as packaging films, paints, adhesives, coatings, inks, agrochemicals, pharmaceuticals, construction material, and electronic application, etc., which have made polymers ideal for many industries, are leading to waste-disposal problem. The waste disposal problem is a threatening topic of sustainable development and green environment. Mostly all the synthetic polymers are derived from monomer originated from petroleum, and these polymers are non-biodegradable in nature. Due to its resistance to biodegradation, these polymers are causing severe damage to the environment<sup>2</sup>.

With the increasing oil price as well as the oil crises worldwide, a change in the approach is required. Instead of synthesizing polymer from petroleum derivatives, a better approach will be to go for renewable raw material such as vegetable oil, which may also be biodegradable in nature<sup>3</sup>.

Guner et al. in 2006<sup>4</sup> referred vegetable oils as triglycerides; they usually consist of one glycerol moiety and three fatty acid moiety. 94-96% of the weight of vegetable oil is because of the presence of a fatty acid moiety. Myristic acid (C-14), Palmitic acid (C-16), Stearic acid (C-18), Oleic acid (C-18), Linoleic acid (C-18), Linolenic acid (C-18),  $\alpha$ -Eleostearic acid (C-18), Ricinoleic acid (C-18), Vernolic acid (C-18), Licanic acid (C-18) are some of the most common fatty acids. Table 1 shows the structures of these fatty acids.

**Table 1:** Structure of different fatty acids

S No.	Name of the acid	Structure
1	Myristic acid	
2	Palmitic acid	
3	Stearic acid	
4	Oleic acid	
5	Linoleic acid	

6	α-Eleostearic acid	
7	Ricinoleic acid	
8	Vernolic acid	
9	Licanic acid	

Due to the difference in the structure of the fatty acid, there is the difference in their physical properties like viscosity, density, melting point, refractive index, specific gravity etc. Table 2 summarizes the physical properties of the common fatty acids.

**Table 2:** Physical properties of different fatty acids

S. No.	Name	Viscosity (cP, 110 °C)	Density (g/cm <sup>3</sup> , 80°C)	Melting point (°C)	Refractive index (n <sub>D</sub> <sup>20</sup> )
1	Myristic acid	2.78	0.8439	54.4	1.4273
2	Palmitic acid	3.47	0.8414	62.9	1.4209
3	Stearic acid	4.24	0.8390	69.6	1.4337
4	Oleic acid	3.41	0.850	16.3	1.4449

Different properties of vegetable oils depend on the fatty acids as well as the position of double bonds in them.<sup>4</sup> Table 3 shows the major constituents of the commonly used oils.

**Table 3:** Major constituent of various oils

S. No	Name	Major constituents
1	Castor Oil	Ricinoleic acid
2	Soybean oil	Linoleic acid and Oleic acid
3	Linseed oil	Linolenic Acid and Oleic acid
4	Palm oil	Oleic acid and Palmitic acid
5	Sunflower oil	Linoleic acid and Oleic acid
6	Rapeseed oil	Oleic acid and Linoleic acid

Saponification and iodine values indicate the different number and position of double bonds. These values are different for the different fatty acids and indicate the nature of the oils. In this review, we will detail, the versatile applications of vegetable oils based biodegradable polymers in diverse areas like biomedical, composites, coating, adhesives, films, packaging, sheet moulding compound, aviation etc.

## 2. Castor Oil

Castor oil's unique structure, having fatty acids with many hydroxyl groups, makes it a very useful for industrial purpose<sup>5</sup>. Castor oil mainly comprises of ricinoleic acid approximately 89% as a major fatty acid constituent. The presence of aliphatic long fatty acid chain, having different functional groups makes castor oil suitable for various types

of reactions and modifications. For instance, Mazo et al. in 2012<sup>6</sup> suggested that due to the presence of carboxyl group several esterification reactions are possible. This is also supported by other researchers as well<sup>7,8</sup>. Different researchers have suggested that the presence of double bonds can facilitate different modifications like epoxidation<sup>9,10,11</sup>. The castor oil based polymers are degradable and this property can be a boon in diverse applications of drug delivery and biomedical implants. Castor oil based polyesters degrade almost 80% in 25 days and may find use as drug implants for local drug delivery systems. On studying the drug release behaviour of the polyesters by doping it with 5-fluorouracil, it has shown a biphasic release with 100% release within 42 hours. Whereas on doping it with isoniazid 100% release took over 12 days<sup>12</sup>. Yeganeh and Talemi<sup>13</sup> used polyurethane networks prepared from castor oil as biomedical implants and in tissue engineering. Polymeric films when interacted with fibroblast cells have shown non-toxic behaviour and good cytocompatibility. Dong et al.<sup>14</sup> have used nano-hydroxyapatite/polyurethane composite based on castor oil as a substitute to human menisci of the knee joint and articular cartilage.

Polymeric adhesives based on oils have low cost and are biodegradable in nature as compared to the ones prepared from petrochemical sources; such adhesives can provide an alternative material for biomedical as well as for other applications. Surgical adhesives find use for haemostasis, sealing air leakages and tissue adhesion. Ferreira et al. in 2007<sup>15</sup> have developed bioadhesives based on vegetable oil. They synthesized castor oil based urethanes with free isocyanate group. These adhesives had the capacity to react with amino group present in biological molecules and were biocompatible and biodegradable in nature. Somani et al.<sup>16</sup> developed polyurethanes derived from castor oil as adhesive for wood-to-wood bonding. Adhesives made from the polyol and aromatic isocyanate adducts, gave superior bonding strength for wood-to-wood specimens. As the NCO/OH ratio increased, the lap shear strength increased and was even superior to some commercially available wood adhesives.

Surface coating is another field where the polymers based on castor oil have found many applications. Thakur and Karak<sup>17</sup> prepared varieties of polyurethanes, by utilizing the

hydroxyl groups of castor oil. They developed surface coating materials from castor oil based hyper-branched polyurethane (CHBPU) as well as from monoglyceride based hyper-branched polyurethane (MHBPU). Both the HBPU have shown good thermal and chemical resistant properties. They observed that mechanical properties of MHBPU were better than CHBPU due to the higher degree of crystallinity and compact structure. In 2011, Mulazim et al.<sup>18</sup> prepared castor oil based photo-curable highly hydrophobic coatings. They observed that the flame retardancy of the resulting polymer increases with the addition of fluorine alkoxysilane compounds. Trevino and Trumbo<sup>19</sup> synthesized acetoacetate esters, by reacting castor oil with t-butyl acetoacetate. They further used the resulting products for formulating various thermosetting coating compositions. After curing with a cross linkers such as multifunctional amines, at different temperature they observed the properties of the films cured at different temperature. They concluded that the films cured at elevated temperature were better than the ones cured at ambient temperature.

Many researchers synthesized interpenetrating polymer networks using polymers based on vegetable oils to improve the certain properties of the polymers. Xie et al. in 1994<sup>20</sup> synthesized simultaneous interpenetrating polymer network based on (castor oil-polyethylene glycol) polyurethane and poly(2-vinyl pyridine) (VP). They coupled castor oil and polyethylene glycol (PEG) with 2,4-toluene diisocyanate and vinyl pyridine by radical polymerization. They studied the properties of the resulting IPN's. They found that the properties viz. crystallinity and water absorption capacity of the IPN's was dependent on molecular weight of PEG used in synthesis as well as the NCO/OH molar ratio. The IPN's also showed good mechanical properties like high tensile strength and ultimate elongation. They further found that making complexes can result in improvement in the conductivity of the IPN's. Similarly Bai et al. in 1997<sup>21</sup> prepared castor oil polyurethanes and utilized it for interpenetrating polymer networks by varying polyurethane/polystyrene ratio. They observed that the mechanical properties of the synthesized polymers showed significant improvement beyond critical styrene concentration. In 2012, Chen et al.<sup>22</sup> worked on castor oil based interpenetrating polyurethane/epoxy resins. They found that the damping, thermal and mechanical properties of composites improved by the addition of carbon nanotubes. The modified composites can find applications as structural materials.

Plasticizer based on phthalates generally enhances the various properties such as flexibility & workability of a polymeric material. On the other hand, it is injurious to human health. These plasticizers may be replaced by eco-friendly plasticizer obtained from bio-resource. However, such plasticizer cannot completely replace these as they are comparatively inferior in performance. To overcome this, Jia et.al<sup>23</sup> synthesized two types of plasticizer i.e. epoxidized castor oil (ECO) and castor oil phosphate ester (COPE) and blended it with PVC. On studying the parameters such as SEM & torque evaluation a good interfacial adhesion and plasticization were reported between ECO, COPE and PVC. Decomposition temperature (Td) & transition temperature (Tg) values showed remarkable increase with the addition of

10gm of ECO & COPE in PVC blends. LOI (Limiting oxygen index) tests showed remarkable increase in the value from 24.2% to 27.8% with the addition of 10gm COPE. Thereby it improves the flame retardant performance of PVC blend.

### 3. Soybean Oil

Soybean oil is one of the most studied oil because of its eco-friendly nature and its cost effectiveness. Due to natural origin and biocompatibility of hydrolysed and epoxidized soybean oil based polymers, it finds use in a drug delivery system and pharmaceutical applications. Abdekhodaie et.al<sup>24</sup> reported regarding self-assembly, thermal transition, interaction with various drugs and surface activity of such soybean oil based polymer. Hydrolyzed polymers of soybean oil (HPSO) exhibited lower thermal transition temperature ( $T_{tr}$ ) than epoxidized soybean oil (HPESO) because  $T_{tr}$  value of the polymer in aqueous solution increases with increasing concentration. The critical micelle concentration was  $0.05\text{mg mL}^{-1}$  for HPSO and  $0.08\text{mg mL}^{-1}$  for HPESO. They further concluded that these polymers reduce the interfacial energy and contact angle on ibuprofen tablets and observed good molecular interaction between HPESO and doxorubicin (anti cancer drug).

In the food contact applications, modified soybean oil can replace the toxic plasticizers such as phthalates. Bueno-Ferrer et al. in 2010<sup>25</sup> used epoxidized soybean oil (ESBO) as a stabilizer for poly (vinyl chloride). On varying the concentration of ESBO from 30 to 50%, resulted in an increase in thermal degradation temperature. This increased its suitability for use in the field of food processing at elevated temperature without any risk of toxic degradation. Multi-arm star poly(DL-lactide) was synthesized using epoxidized linseed oil (ESO) as an initiator through ring opening polymerization. Molecular weight, polymerization rate as well as monomer conversion rate increased in the presence of ESO. Degradation study shows that star shaped epoxidized linseed oil poly lactide (ESOPLA) was more stable than PDLA. Comparison of the SEM images indicates that ESOPLA films were hydrolytically more stable than PLA films<sup>26</sup>.

Polyols are one of the predominant reactants in polyurethane synthesis. Soy-based polyols have been used as potentially low-cost materials in plastic and polymer industries for decades. Lu and Larock in 2008<sup>27</sup> developed methoxylated soybean oil polyols (MSOLs) with hydroxyl functionality ranging from 2.4 to 4.0 and utilized them for preparing waterborne anionic polyurethane dispersions. The properties of the resulting films namely the thermal, physical and mechanical properties were almost at par with the polyurethanes obtained from polyols based on petroleum. In 2011, Lu et al.<sup>28</sup> used the seeded emulsion polymerization technique to prepare surfactant-free core shell hybrid latexes, using waterborne soybean oil based polyurethane as shell and poly [styrene (S) & butyl acrylate (BA)] as the core. TEM images confirmed the formation of a core-shell structure in the hybrid latexes. The glass transition temperature of the films increased with increasing content of poly (ST-BA) and multifunctional vinyl cross linker. The resulting films have shown improved thermal and

mechanical properties as compared to pure polyurethane. Silva et al. 2012<sup>29</sup> blended Karawila seed oil with the synthesized soybean oil based alkyd resin. On examining the various parameters they concluded that the results were highly influenced with 30% incorporation of Karawila oil to the synthesized alkyd resins which are suitable for film applications.

Oprea and Doroftei<sup>30</sup> compared the biodegradation rate of polyurethane acrylate and acrylated epoxidized soybean oil blend elastomer in the presence of a fungus *Chaetomium globosum*. They found that the acrylated epoxidized soybean oil degraded more in comparison to polyurethane acrylate.

Sacristan et al. in 2010<sup>31</sup> prepared soybean oil based copolymers by cationic polymerization using boron trifluoride etherate as initiator and examined the flame retardant property. They reported LOI (limiting oxygen index) values from 23.7 to 25.6, the results indicate that material will not burn at normal condition. Generally, a polymer with LOI values below 21 burns continuously (ASTM D2863-00). Such kind of polymers may find applications in the area of aviation. In 2005, Lu et al.<sup>32</sup> synthesized thermosetting resins from soybean oil and utilized them as sheet moulding compound (SMC). For synthesis, they used acrylated epoxidized soybean oil (AESO) as starting material, and obtained rigid polymer by copolymerizing it with styrene. The synthesized resin worked well with formulating agents as well, the final properties of the final product was at par with the commercial resin. The findings indicate that this has a potential for commercial exploitation.

#### 4. Linseed Oil

Linseed oil is a drying oil finds extensive use in surface coatings. According to Conte et al.<sup>33</sup> it is a triglyceride oil composed of 6% stearic acid, 18% oleic acid, 15% linoleic acid, and 53% linolenic acid and 6% palmitic acid. It has approximately six C=C bonds per triglyceride. The high number of double bonds per triglyceride makes linseed oil particularly susceptible to cationic polymerization. Epoxidised linseed oil when blended with PVC (LOE/PVC) in different ratios, the blend with higher component of epoxidized linseed oil (i.e. 85/15) has shown good degradability, it degraded almost 68 weight % in 6 months. Riaz et al. in 2010<sup>34</sup> have suggested the potential applications for such sustainable resource based blend films include packaging material and biodegradable plastic sheets, which find applications in products such as bio-bags.

Waterborne polyurethane dispersions synthesized from linseed oil for wood furniture finishing applications. Chang and Lu<sup>35</sup> studied the curing of the synthesized polyurethane dispersion waterborne wood coating through different curing processes. They tried UV curing alone with photo-initiator or air-drying alone by mixing it with metal dryers as well as a combination of both UV/air and air/UV dual cured system. They found durability, light fastness and adhesive properties of films prepared using the UV/air dual curing process were better than only UV process. In 2012, Mahendran et al.<sup>36</sup> used acrylated epoxidized linseed oil for wood surface coating application by modifying it with photo-initiators.

Sharmin et al.<sup>37</sup> used poly (ester urethane) metallohybrid (PEUMH) with copper (II) acetate as inorganic precursors. The metallohybrids showed good physico-mechanical and chemical resistance performance. These metallohybrids can be exploited as anti-bacterial self-sterilizing protective coating due to oligodynamic effect of metal with safe usage up to 200°C. Suryanarayana et al. in 2008<sup>38</sup> synthesized microcapsule of urea formaldehyde resin with linseed oil and other constituent and exploited them for self-healing coatings with satisfactory anti corrosive property.

Carter et al.<sup>39</sup> used epoxidised linseed oil (ELO) as an alternative to poly (vinyl chloride) tiles in the field of flooring industries as the PVC tiles have environmental issues. The set of flooring tests shows that material developed from ELO and cross-linker is fit for providing a flooring surface, albeit at different ratings depending on processing parameters. On studying vegetable oil based fluorocarbon water repellent agent Zhao et al.<sup>40</sup> concluded that linseed oil based water repellent agent showed highest water repellence and durability. Hespel et al. in 2012<sup>41</sup> designed an atom transfer radical polymerization (ATRP) lipoinitiator and utilized this initiator for tert-butyl acrylate polymerization. Amphiphilic copolymers were obtained after the subsequent acidolysis of PtBA block into poly acrylic acid. The ability of a lipid-b-PAA copolymers to auto-associate in water imparts the copolymer the ability to form well defined micelles in acidic media whereas the micelles dissociated when the pH was raised above 7.

Lazko et al. in 2011<sup>42</sup> developed flax short fibers biocomposite based insulating material. They added linseed oil to the initial formulation of the biocomposite to improve its hydrophobicity. Not only the water absorption reduced to 10 times as compared to the samples in which no linseed oil was added, the mechanical and thermal properties were found to improve as well after 20 days of drying at 50°C. Samper et al. in 2015<sup>43</sup> prepared epoxidized linseed oil (ELO) matrix and slate fabrics based eco-friendly composite laminates by resin transfer molding technique. ELO-slate composite laminates showed better properties in comparison to other fiber based on carbon, aramids etc. ELO-slate composite showed overall mechanical properties at par with conventional glass & basalt fiber reinforced composite laminate. Ahmad et al.<sup>44</sup> obtained linseed oil based polyols by epoxidation and then further treated them with toluene diisocyanate (TDI) in various ratios to develop a series of polyurethanes. They reported that physico-mechanical and anti corrosive properties enhanced by incorporating 10% of TDI in Linseed oil based polyurethane.

#### 5. Other oils

Vegetable oil based biodegradable polymeric materials have been recently improved with hyper-branched architecture. Sunflower oil based hyper-branched polyurethane (HBPU) was synthesized with and without pentaerythritol as a multifunctional unit. <sup>1</sup>HNMR and FTIR were used for characterization. Physico-mechanical properties and biodegradability of the synthesized material were better than linear polyurethane (LPU). The bio-based synthesized HBPU with such properties has good potential as a thin film material for the advanced multifaceted application<sup>45</sup>. In 2012, Kong et al.<sup>46</sup> synthesized Canola oil based poly (ether



ester) polyols and they further utilized it to produce polyurethane (PU). Characterization studies reveal that the prepared PUs had high glass transition temperature, good hydrolytic stability and alkali resistance and formed highly cross-linked structures.

Lu et al.<sup>47</sup> developed rapeseed oil based waterborne polyurethane to get a biodegradable material with better performance. Furthermore, they blended synthesized polyurethane (PU) with plasticized starch (PS) so that the shortcomings of starch material such as inferior mechanical properties, water sensitivity etc. can be overcome. On comparing the values of the PS/PU blends with the pure PS films. They obtained higher values of the elongation at break (from 185 to 480%), the toughness of the material (from 1.8 to 7.1 MPa) and the tensile strength (from 2.8 to 4.1 MPa) by varying the polyurethane content.

Waskitoaji et al. in 2012<sup>48</sup> softened Poly (vinyl chloride) by a palm oil based plasticizer, which can be a substitute to commonly used DEHP phthalate plasticizers. Tanaka et al.<sup>49</sup> reported that the flexibility of palm oil based polyurethane foams increased on increasing the content of palm oil based polyols.

Oil palm fibre (OPF) consists of lignocellulosic fibre. OPF is thermally more stable compared to hemp and flax fibres. Its utilization in biocomposites can preserve the environment. The work of Shinoj et al. in 2010<sup>50</sup> indicates that by increasing the temperature, specific heat capacity of OPF increased. The opacity of linear low-density polyethylene (LLDPE) increased to 99% from 63.58% with the addition of 10% oil palm fibre. In 2012, Alam and Alandis<sup>51</sup> developed Polyetheramide (PEtA) based on olive oil. They further reacted it with toluene 2, 4-diisocyanate to obtain poly ether amide urethane (PEtAU). Studying the corrosion behaviours of the synthesized polymer they found that PEtAU coatings show good physico-mechanical and corrosion resistance properties. Jatropha oil is one of the most prominent oil for polymer synthesis as it can be chemically modified due to the presence of high degree of unsaturation. It consists of 78.9% unsaturated fatty acids with 43.1% and 34.4% oleic and linoleic acids<sup>52</sup>. Boruah et.al in 2012<sup>53</sup> prepared alkyd resin based on Jatropha Curcas oil. On evaluating its physico-chemical properties it has been concluded for surface coating purposes. In 2014, Gogoi et.al<sup>54</sup> further exploited Jatropha oil by reinforcing it with expanded graphite as a biocomposite by varying its loading percentage. Improved results were reported on studying its thermal, mechanical and flame retardancy properties. Gogoi et.al in 2015<sup>55</sup> incorporated NiO nanoparticle in Jatropha oil based alkyd resin. Enhanced tensile strength was reported with 3 wt% of NiO on comparing it with the pristine polymer. Limiting oxygen index value showed improved flame retardant property even with incorporation of 1 wt% NiO nanoparticle. Gaikwad et al.2015<sup>56</sup> exploited Karanja oil (*Pongamia glabra*) & cotton seed oil (*Gossypium arboreum*) to synthesize a series of polyester polyols. They further developed eco-friendly polyurethane coatings using dipentene (limonene) a green solvent and has shown very good thermal stability & coating properties like gloss, adhesion, pencil hardness, dryness to touch, impact and chemical resistance. Several scientist have

used different oil such as Nahar seed oil,<sup>57</sup> Yellow oleander,<sup>58</sup> Karanja,<sup>59</sup> *Ricinodendron heudelotti*<sup>60</sup> to synthesize alkyd resin. On characterization, the cured epoxy-blended resins were found to have properties suitable for coating application.

## 6. Future Direction & Conclusion

Vegetable oils are present in large quantities in nature; can be easily obtained and very cost effective. These have special functional groups, which impart them distinctive properties and make them biodegradable, renewable and eco-friendly materials. Vegetable oils are utilized in the past several years in paints and coatings however, current study and researches are being directed towards introducing some unique properties to enhance their performance and make them eco-friendly at reasonable prices. Initially vegetable oil based polymers were just the partial replacement of synthetic part in the content. Now, the trend should be to raise the amount of bio-based material, taking the overall properties in concern. One can mix the various vegetable oil to synthesize new product with enhanced properties in the upcoming days. Those agents that are renewable and eco-friendly must substitute the agents used for curing, drying, diluting and modifying. Genetically modified oil seeds can be produced in order to obtain vegetable oil with desired composition of fatty acids so that the properties of vegetable oil based polymer can be improved for various application. Modified vegetable oil can be utilized for achieving the goals of green chemistry along with its application in various fields with lesser drawbacks. Multi-step reactions are required for polymer synthesis based on vegetable oil. Its mainly involves reaction at high temperature for longer duration with large quantities of hazardous solvents. Therefore, strategies can be worked out for minimizing the shortcomings by using less or solvent free synthesis during formulation, processing and application of synthesized polymer. Longer synthesis times at elevated temperature reduce the quality as well as the quantity of the yield, which can be replaced by shorter reaction time at lower temperature for instance as in microwave synthesis. An approach towards enzymatic synthesis will also be a good alternative. The easiest method to enhance the feature of vegetable oil is to mix it with the commercially available polymer. This not only improves its performance but also makes it cost efficient.

The vegetable oils provide a large variety of options for the preparation of biodegradable polymer. The incorporation of vegetable oils into the polymer chain allows tailoring the properties of polymer product for their widespread application. The petroleum-based polymers are non-renewable and harmful to the environment. Vegetable oils based polymers provide an alternate material which possesses not only the biodegradability and biocompatibility but also are obtained from renewable resources. The contributions of the different scientist indicate that the vegetable oil based polymers can be a useful material in different applications. The wide variety of oils with different properties available offers a numerous possibilities, which need to be explored to develop environment friendly materials with desired properties to suit the applications.

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