Review Article

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# Coatings resins synthesized from oil-based precursors- A review

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## Abstract:

With the increasing focus in the past few years on finding renewable, sustainable, and environmentally friendly alternatives to traditional fossil fuel and other non-renewable raw materials, the coating industry has adapted to the use of oils from various naturally occurring oilseeds. These oilseeds are not only beneficial for the environment but also ameliorate and incorporate many properties in the traditional polymeric resins. In this review, four widely used polymeric resins, namely polyester amides, epoxies, alkyds, and polyurethanes are discussed. The use of oils in each of these resins and the various benefits are discussed.

**Keywords:** Sustainability, vegetable oils, coatings, polyurethane, epoxy, polyester amide, alkyd

## Introduction:

Oils are generally a product of the reaction between 1 mole of glycerol and 3 moles of fatty acids, better known as triglycerides. Oils can be broadly classified as petroleum-based and vegetable and animal-based oils, depending on their source of origin. Petroleum-based products, as we all know, have been one of the major factors behind environmental degradation and pollution and have, therefore, fallen out of favor. Also, the fact that their depletion is imminent, with some saying as early as 2050, does not help. The need to look forward to bio-based raw materials is a choice of interest for present-day research and academic scholars. [1-3] Thus, there is a shift in emphasis on resins and polymers obtained from renewable resources such as lignin, starch, protein, shellac, rosin, cellulose, polyhydroxyalkanoates, alginate, furanone, chitosan, vegetable oils [VO], and wool fibers. The applications of the products made from these are endless and include fields such as biodiesel, plasticizers, lubricants, biodegradable packaging materials, adhesives, printing inks, coatings, and paints. Vegetable Oils possess many great properties which benefit environmental sustainability like non-toxic, inexhaustible, locally abundant, non-volatile, and biodegradable resources. They yield resins and polymers capable of battling it out with fossil fuel-derived petroleum-based products and establish applications in the progress & benefits of paints and coatings, besides their other industrial applications. [4,5]

Vegetable oils have been used in the form of paints for as long as humans have known to be existing on this planet, which is evident by the numerous cave paintings that have been discovered. As development took place, more and more new resources were discovered, and by the time the Industrial Revolution came around, the use of petroleum products skyrocketed.[5] However, as time passed, humanity has realised its mistakes and started shifting towards 'greener' materials and resources. From an industrial and environmental perspective, the coatings and resins industry has shifted to the use of VOs because of their great abundance, ample possibilities of variations, functional attributes, cost-effectiveness, and greener monomer supply, which includes good biodegradability.[6-8] To get rid of petroleum dependence, researchers have explored various routes to make a variety of polymeric resins, which include polyurethanes, epoxies, polyesters, alkyds, phenols, acrylics, silicates, amides, and many more. [7] Vegetable oils, like most, are also triglycerides formed by ester linkages between three fatty acids and glycerol, as displayed in the figure below. (fig. 1) [9]

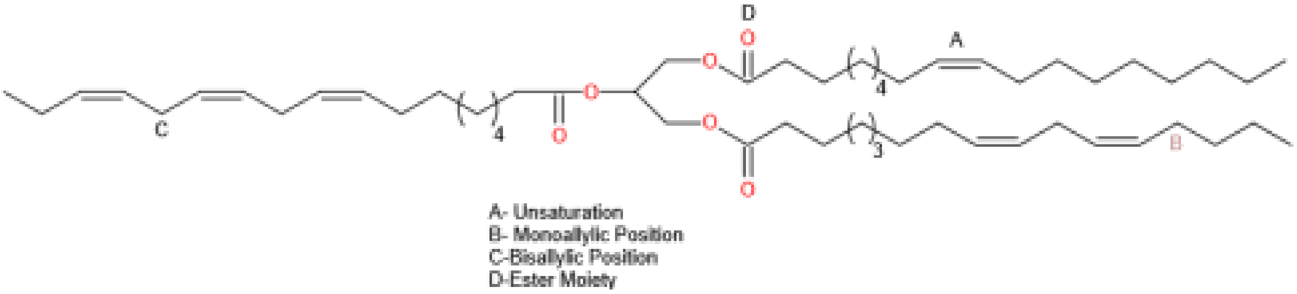

**Fig. 1: General structure of oils**

The fatty acids used typically contain 8-24 carbon atoms with 0-5 carbon-carbon double bonds. Most of them are cis configured and non-conjugated. Many articles give the number of carbon-carbon double bonds and the reactivity of each type of oil. [9,10]

Abundantly available in nature, the triglycerides of these fatty acids contain many suitable functionalities like hydroxyls, double bonds, esters, epoxies, and other functional groups in their backbone that can partake in several chemical reactions. [5] A natural phenomenon that exemplifies this is drying, wherein the VOs involved are termed as 'drying oils.' Drying is a natural feature of the VO and is dependent on various factors such as their unsaturation, their conjugation, geometrical arrangements of the substituents (cis or trans), etc. [11,12] While drying and semi-drying oils are generally preferred in surface coatings, non-drying oils too can be used for the same by incorporating proper functional groups in their backbone like hydroxyls, acrylics, vinyls, etc. by chemically reacting them and transforming them into film formers. [11] [12] The maintenance of natural flexibility and fluidity characteristics of VOs enable them to be used as reactive diluents and, in some cases, as solvent-less free-flowing coating materials, felicitating the development of solvent-less coatings. [5] Vegetable oils can be obtained from the seeds of various plants which include neem [13], palm [14], Nahar [15], rapeseed [16], castor [17], jatropha [18], soybean [19], etc.[20] The various ways in which these VOs can give epoxies, alkyds, polyols, etc., are given below. (fig. 2)

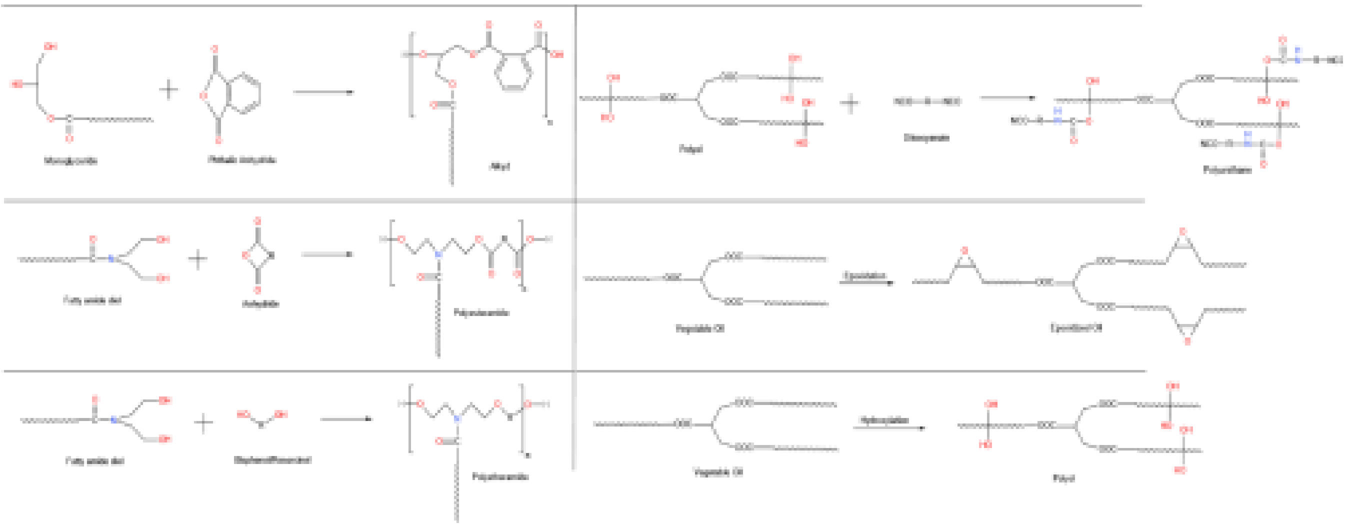

Fig. 2: Resins prepared from oils

## Polyester amides:

As the name suggests, polyesteramides or PEAs are made up of both ester and amide groups in a repeating manner in the polymer backbone. The esterification reaction between amide diol of vegetable oil and an acid/anhydride like phthalic anhydride results in the formation of amide-modified alkyds, better known as PEAs. [5,15,21-26] PEAs can be synthesized from various vegetable oils like cottonseed oil [26], Nahar seed oil [15], linseed oil [27], Pongamia glabra oil [28], jatropha oil [29], etc. in a two-stage process. The first stage involves the reaction of vegetable oil to be used with sodium methoxide to obtain methyl esters of the oil after solvent extraction. These methyl esters of the oil are then converted to amide diols by reaction with a diethanolamine in the presence of sodium methoxide and high temperature (depends on the oil). [15,26,28] Monomers with two hydroxyl groups and pendant vegetable oil (fatty) chains are amide diols. [5]

The second step utilizes the diethanol amide obtained from the 1st step to synthesize the polyester amide resin. The diethanol amide undergoes an esterification reaction with an anhydride like phthalic or maleic anhydride. The hydroxyl groups of the amide diols react with the anhydride in the presence of lead oxide (PbO) and at a high temperature to finally give the polyester amide resin. The reaction progress can be determined by acid value determination. [15,26,28] The amide, ester moieties, and pendant (fatty) chains present provide synergistic properties of any component, like improved chemical resistance, flexibility, thermal stability, and resistance to alkaline media, to the PEA resin. The fatty chains can additionally also serve as functional sites for further modification. The thermal and mechanical stability of PEA is influenced by the double bonds, amide groups, and hydrogen bonding. [5,25,30] The PEA obtained from different vegetable oils can be modified by various monomers like acrylic/vinyl monomers (styrene, vinyl acetate) [31], copolymers such as poly (styrene-co-maleic anhydride) [21,32], hydroxyethyl fatty acid [33], melamine [34,35], pyromellitimide ring [36]

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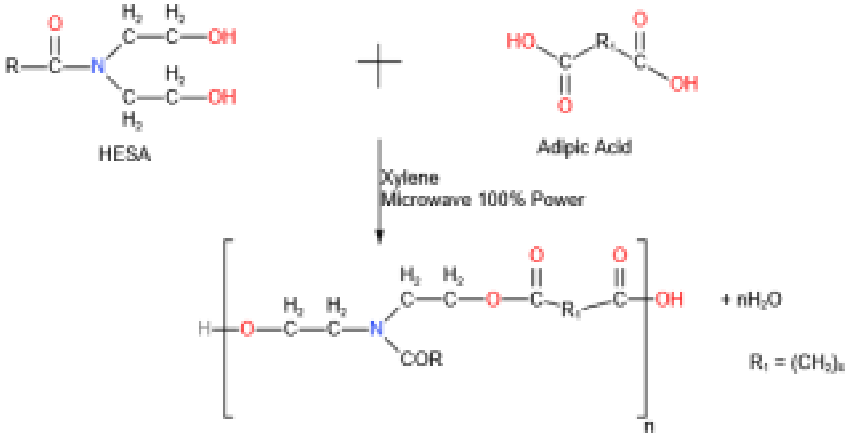
Each of these hassome unique properties that they impart to the resins. For example, vinylation improves the anti-corrosive, curing, and physio-mechanical properties. [31] Poly (styrene-co-maleic anhydride) increases the alkali resistance, impact resistance, thermal stability, and scratch hardness. [21] When pyridine is added into the polymer backbone, it enables the use of the formed resin safely up to a temperature of 140°C. [32] The use of hydroxyethyl fatty acid amide, depending on its content, gave better chemical and mechanical properties, whereas the incorporation of melamine gives good performance in alkali and the good amount of linkages impart good physio-mechanical properties. In addition, the Thermo-Gravimetric Analysis (TGA) shows that the resin can be used to a temperature of 200°C. [35] Pyromellitimic anhydride (PAA), a novel dibasic acid, can be used to replace phthalic anhydride (PA) and incorporate the pyromellitimide ring in the PEA. Enhanced physicomechanical properties such as scratch hardness, adhesion, gloss, and resistance to mechanical damage along with better film performance in terms of chemical resistance to acid, alkali and water and improved corrosion resistance are shown by the modified resin [36]

Polyester amides also witness the incorporation of metal/metalloid hydroxides (Al, B) and acetates (Zn, Cd, Cu, Co, Mn), leading to enhancement in their properties. [37] The minor incorporation of divalent Cadmium and Zinc using their acetates in linseed oil not only enhanced the physio-mechanical properties but also improved the anti-corrosive properties and curing temperature. [37,38] When copper salts of polyester amide resins are formed, it was found that they are self-polishing in nature during leaching. Cu-PEA resin also had better curing as compared to PEA and is used as a resin for anti-fouling coatings. The mechanical strength is found to be very low and chlorinated rubber resin has to be incorporated to improve them. [39] Cobalt is incorporated in a different way using sodium ethoxide as an initiator. Polyester amide is prepared from sunflower oil in a two-step process. First, it is reacted with diethanolamine forming hydroxyethyl sunflower amide (HESA), which is converted to PEA using glycerol and adipic acid. After preparing the PEA resin, it is dimerised using boric acid for incorporating cobalt. The di polyester amide (DPEA) is then reacted with cobalt chloride to form cobalt DPEA or CDPEA. The CDPEA synthesized is thermally stable and can be used for making more potent and safer antifungal drugs. 40

Synthesis of HESA



Synthesis of Polyesteramide

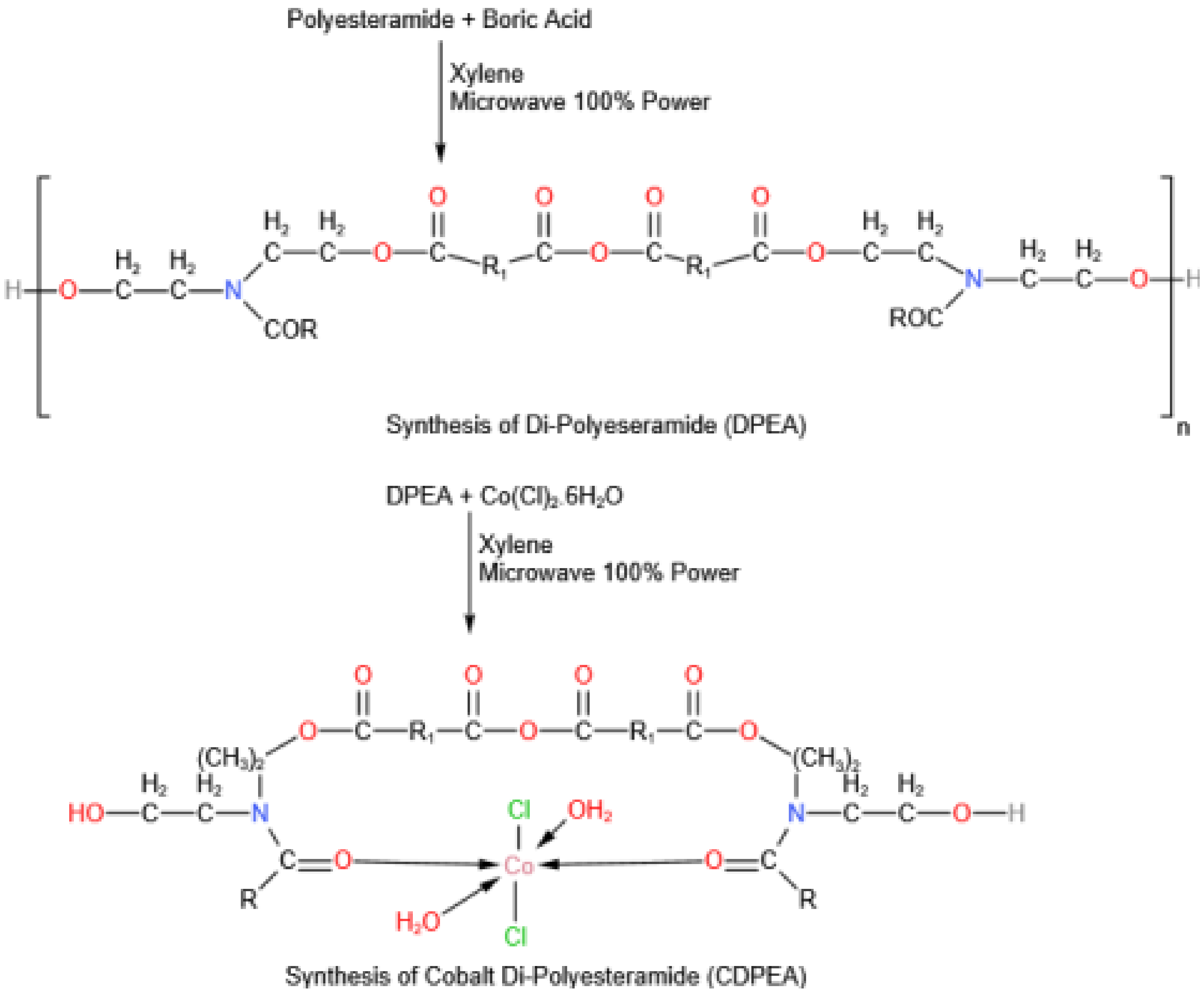


**Fig. 3: Synthesis of Polyesteramide and Cobalt Di-Polyesteramide**

Boron and Aluminium are unique as compared to the transition metals mentioned earlier since their incorporation is similar yet different. Jatropha curcas seed oil, a renewable and abundantly available raw material, sees the incorporation of boron in the PEA resin synthesized from it. Boron hydroxide or $B(OH)_3$ is used to incorporate boron in the polymer backbone and it reduces the baking temperature and curing time along with improving the physio-mechanical properties and corrosion/chemical resistance performance. [41] This boron-PEA can be modified further by reacting with tolylene-2,4- diisocyanate (TDI) to give boron polyester amide urethane (BPEAU). The incorporation of TDI causes gelation in the BPEAU and is a major benefitting factor since it improves the antimicrobial properties of the BPEAU coatings. The oil used for making PEA can also be varied. [42,43]

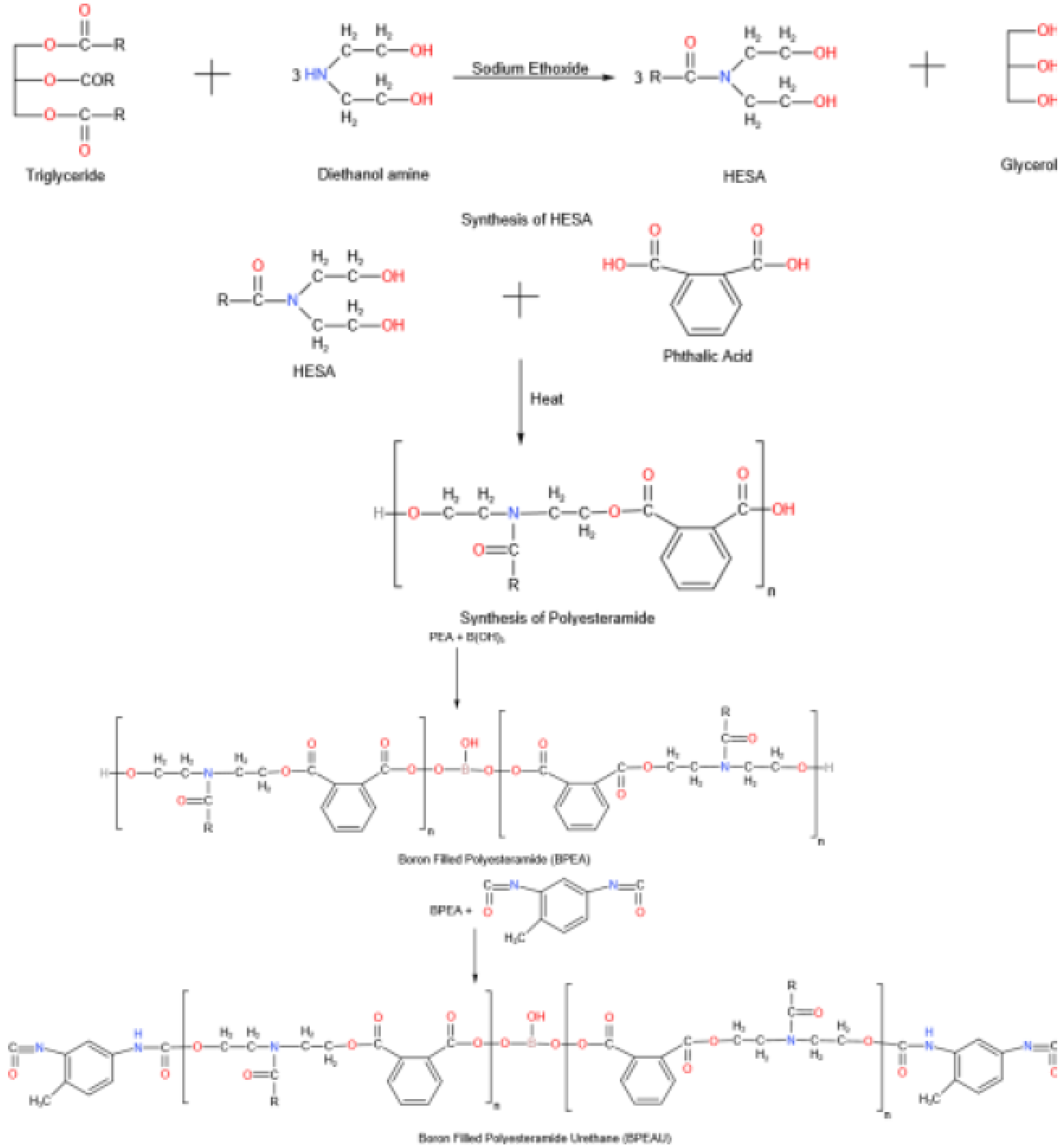


**Fig. 4: Synthesis of Polyesteramide and Boron filled Polyesteramide Urethane.**

Aluminum is by far the most used metal to fill the polyester amide resins. It is incorporated similar to boron by using $Al(OH)_3$. Aluminium hydroxide is reacted with the polyester amide resin, which is formed in the same way as mentioned earlier from oils. Many oils like linseed oil, Pongamia glabra oil, cottonseed oil, jatropha oil, Nahar oil, and many more have been used for this purpose. Even urethane has been incorporated in the Aluminium PEA to form aluminium polyester amide urethane (APEAU). The changes in the properties are significant, and mainly, the anti-corrosive and antimicrobial properties are improved along with others like physio-mechanical, curing temperature, etc. [23,26-30,44,45]

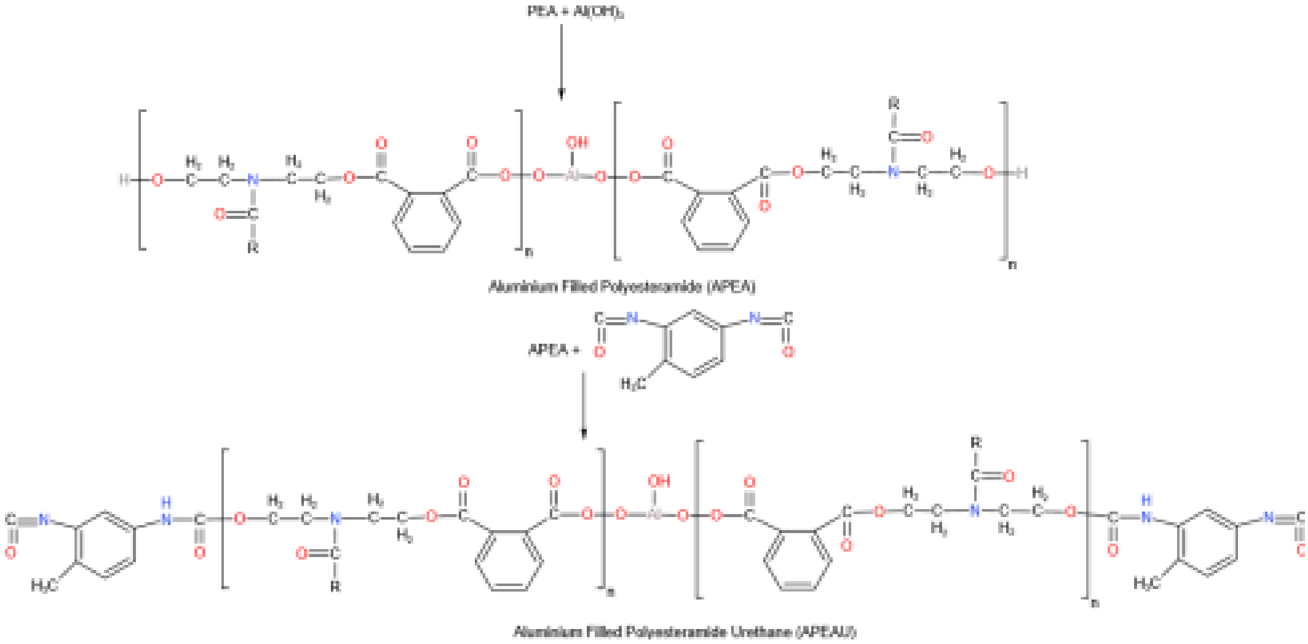


**Fig. 5: Synthesis of Aluminium filled Polyesteramide Urethane**

Generally, the solvent xylene is used in the synthesis of polyester amide from various oils. However, a method has been developed by Ahmad et al., which eliminates the use of solvents and also lowers the temperature of the process, reducing the overall toxicity of the process. The resin yielded also has better physicomechanical and chemical resistance properties as well as better thermal stability as compared to the commercial peers. [46] These bio-based polyesteramides can qualify as advanced surface coating materials in some cases. Hyper-branched polyester amide or HBPEA, synthesized from vegetable oil, is one such material. Its usage in polymeric surface coating application is fostered by the desirable impact strength, abrasion resistance, gloss, scratch hardness, adhesion strength, and mechanical properties. The rheological studies showed differential flow behaviour and the DSC analysis showed increased thermal stability as well. [47]

## Epoxy Resins:

Epoxy resins have a plethora of applications which include paints and other types of coatings, polymers, composites, etc. 41% of the global liquid epoxy resins produced are used for coatings, 31% are for adhesives, while the remaining 28% have a variety of other applications. [48] Bisphenol-A is one of the main ingredients used in the preparation of epoxy resins. However, the aforementioned compound has a lot of adverse effects on human health. [49] In applications such as thin can coatings, there is a high possibility of leaching and can prove to be really dangerous. [50] Bisphenol-A has even triggered the

World Health Organisation (WHO) to advise its discontinuation in food-related products. [51] Thus, the use of vegetable oils to synthesize epoxy resins has seen a substantial increase in the past few years.

As discussed earlier, bio-based oils or plant oils are triglycerides that can be distinguished as drying, non-drying, and semi drying, depending on the degree of unsaturation. The degree of unsaturation is typically expressed in terms of iodine value.

Drying oils have higher unsaturation and an iodine value of above 150, whereas the iodine value of semi-drying oils lies between 100-150. The iodine value of non-drying oils is under 100, which tends to make them greasy and rot. [9] [52] In the case of epoxy resins, the striking feature is the presence of an oxirane ring that differentiates them from the others. Some vegetable oils naturally contain these structures; however, there is the option to incorporate the oxirane ring at the unsaturation present in the vegetable oil using an all-to-familiar process called epoxidation. The process of epoxidation can

follow various routes such as chemo-enzymatic epoxidation, epoxidation by peracids, utilizing dioxirane, phase transfer catalysis, among others. [53,54] Many suitable curing agents like amines, acids, amides, anhydrides, etc., can be used as curing agents to obtain a cured polymer with preferable characteristics. [5,55]

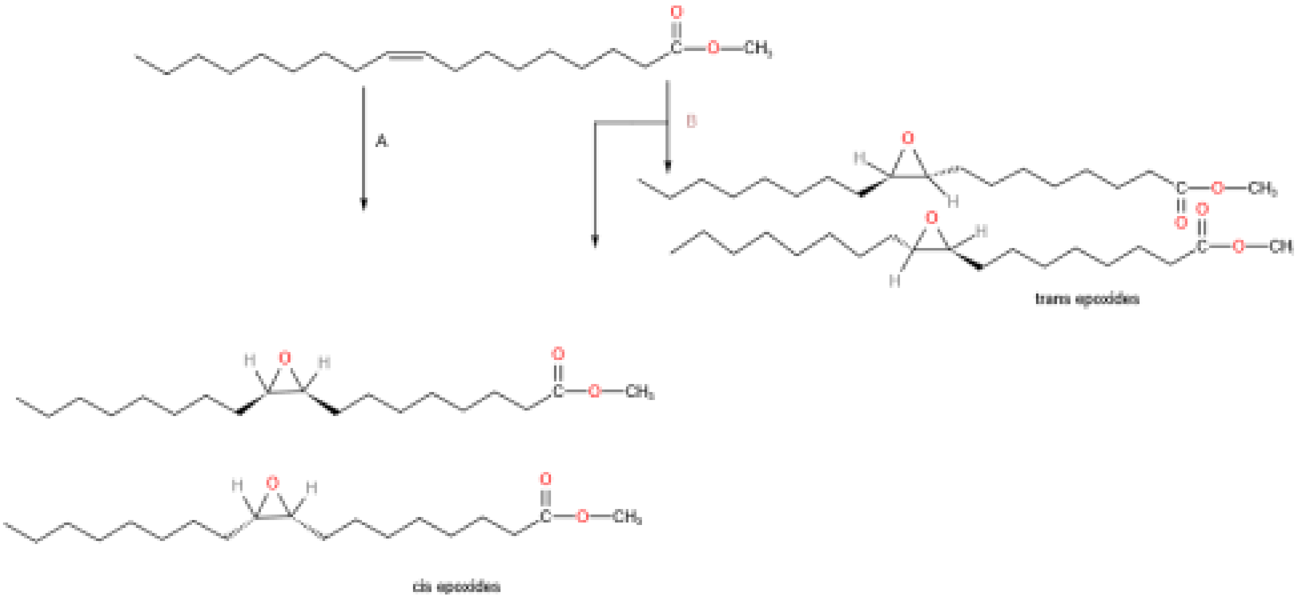


**Fig. 6: Cis and trans epoxide synthesis**

Some of the oils that are used in epoxy resins are canola oil, linseed oil, soybean oil, etc. [48] The epoxidized form of soybean oil or epoxidized soybean oil (ESO) is one of the most extensively used oils as a replacement for petroleum-based products and has an estimated market size of 300 million dollars by 2020. [56,57] Epoxy soybean oil cured with benzylsulfonium hexafluoroantimonate derivative cationic catalyst find its way in advanced applications like shape memory polymers, while the ones cured with boron trifluoride diethyl etherate catalyst diffuses the ESO into a paper to create paper composites with excellent water resistance. [58,59] As far as coatings are concerned, oil-based epoxy resins are used mainly in curing. The most commonly epoxidized oil is linseed oil due to the abundance of unsaturation in the linolenic acid chains. (fig. 7) [56]

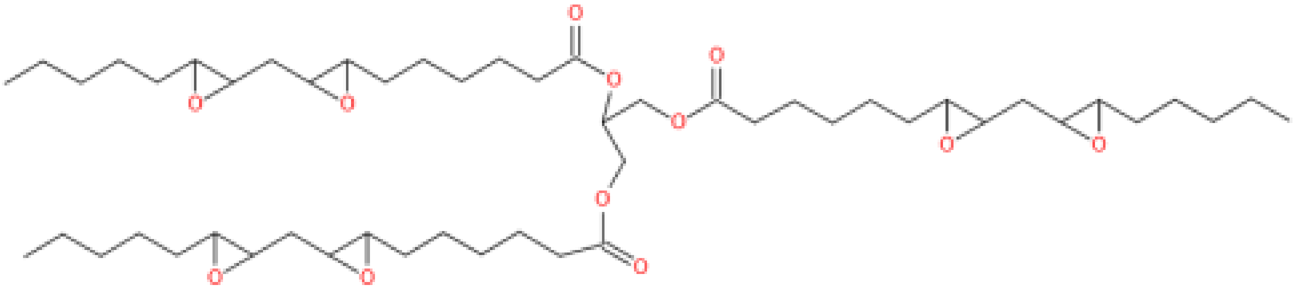


**Fig. 7: Epoxidized linseed oil**

It was discovered that the combination of epoxidized linseed oil (ELO) with curing agents/hardeners like anhydrides [60] or dicarboxylic acids [61] yielded highly cross-linked networks making them extremely favourable for industrial applications and creating thermally and mechanically stable materials. ELO, like ESO, is also used in making composite materials that yield crucial properties such as tensile and flexural strengths. [62-64] ELO, when cured with anhydrides, gives rise to a polymeric network with very interesting properties. These polymers are used in conjunction with natural fibres in composites and may be of great environmental interest. [55]

Epoxynorbornene linseed oils (ENLO) are another type of linseed oil that are used to make UV-curable hybrid coatings. The tetraethylorthosilane (TEOS) oligomers used for their situ synthesis and their hybrid structures influenced the hybrid film characteristics like fracture toughness, Young's modulus, tensile strength, etc. Along with that, ENLO being a novel type of epoxide from renewable sources replaces other synthetic epoxy resins and provides potential applications on adhesives, inks, and coatings. [65] Epoxidized Linseed oil (ELO) is also used in making anti-corrosive coatings. As reported by Ahmad et al., linseed oil is epoxidized in situ and then hydroxylated to give hydroxylated ELO (HELO).

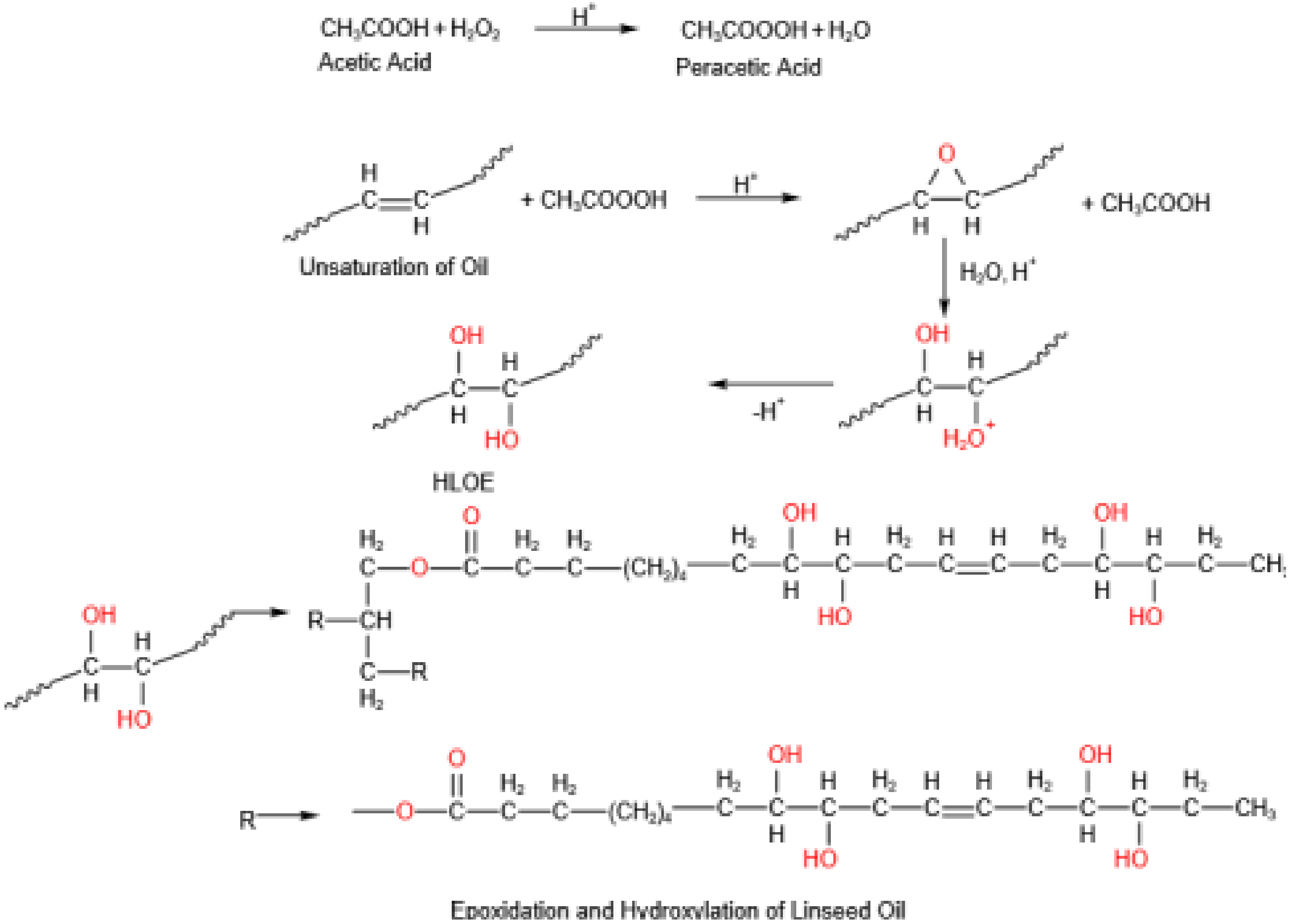


**Fig. 8: Epoxidation and Hydrolysis of Linseed Oil**

This HELO is further reacted with butylated melamine-formaldehyde (BMF) for curing in different weight percentages (approximately 10–30 wt.% on the weight of epoxy resin) for developing coating materials.

The cured films obtained offer an amalgamation of superior characteristics of aliphatic epoxy and melamine resin-like greater values of flexibility, gloss, chemical and thermal resistance, adhesion. These systems are safe to use till temperatures of 200 °C. [35] Linseed oil epoxies many a time do not have the desired properties needed for specific application and reaction is not the only way. Another way of achieving these desired properties is blending. To compensate for the lack of toughness and strength, linseed oil epoxies are blended with poly (methyl methacrylate) and polystyrene. The linseed oil epoxy polystyrene blends were found to have glass transition temperatures in the range of 60-77.6°C, and those of linseed oil epoxy- poly (methyl methacrylate) blends were found to be in the range of 75-82.4 °C. Only a small amount of PS and PMMA (16.6 % wt) in ELO turns it into a rigid mass. [66]

Another oil that is used in epoxy resins is canola oil. Although the progress is limited as compared to linseed & soybean oil, the work over the previous couple of years has been explored regarding epoxidation of canola oil using various catalysts. [67,68] Somidi et al. conducted a study in 2014 with a target to discover inextinguishable bio-based sources for lubricants and deduced that sulfonated $SnO_2$ is the most selective catalyst to carry out the epoxidation of canola oil (ECO).

[68] Epoxidized Canola Oil based thermosets along with phthalic anhydride as curing agents have been created. The molar ratio of ECO:PA affected the thermos-mechanical properties more, whereas the rate of reaction of curing was dependent on the temperature and the molar ratio of PA. [69] A grapeseed oil-based polyamine is explored as a curing agent for bio based epoxy resins. Animated Grapeseed Oil (AGSO) which is effectively synthesized by using UV-initiated thiol-ene coupling, is utilized in curing epoxidized linseed oil (ELO) and was collated with two other curing agents. One was the commercially available Priamine and the other was also derived from grapeseed oil, an animated fatty acid (AFA) curing agent. DSC analysis of the AGSO and ELO polymer revealed the glass transition temperature to be around -38°C. [70,71] Another similar compound used to obtain good epoxy resins is cardanol. Cardanol is a non-edible by-product that is obtained from cashew nut shell liquid and constitutes mainly a mixture of alkylphenols with differing degrees of unsaturation. [72,73] Epoxidized cardanol-based resins are used along with isosorbides as a replacement for Bisphenol-A. Epoxidized cardanol resins, when cured, give polymers with $T_g$ below 50°C and cross-linking densities roughly five times lesser than DGEBA resins. [72,74]

While not many epoxy resins aren’t synthesized from oils, there are many bio-based products from which epoxy resins are synthesized. Some of the examples for these include saccharides [75], isosorbides [76], furans [77], terpenes [78], rosin [79], natural rubber [80], etc. Thus, vegetable oils have proven to play a pivotal role in epoxy resin synthesis.

## Alkyds:

A polyol, a multifunctional acid, and an unsaturated fatty acid consisting of oil-modified polyesters, which are formed by polycondensation, are called alkyds.[5] These alkyds are classified as short oil (30-42%), medium oil (43-54%), long oil alkyds (55-68%), and very long oil alkyds (≥68%) based on the weight percentage fraction of vegetable oil in the resin. [81] Alkyds can be synthesized in two ways:

a) Fatty Acid Process

b) Monoglyceride Process

The structure below represents a conventional alkyd resin. Generally, about 60-70% of an alkyd resin constitutes biodegradable material (glycerol, fatty acids), and the remaining is made up of non-biodegradable material (phthalic anhydride). [5,81] As far as coatings are concerned, alkyds are one of the most prominently used resins in coatings. [82-84] Constant efforts have been made to make alkyd paints environmentally safe by using environmentally friendly diluents, catalysts, driers, pigments, etc., and have been successful. [85]

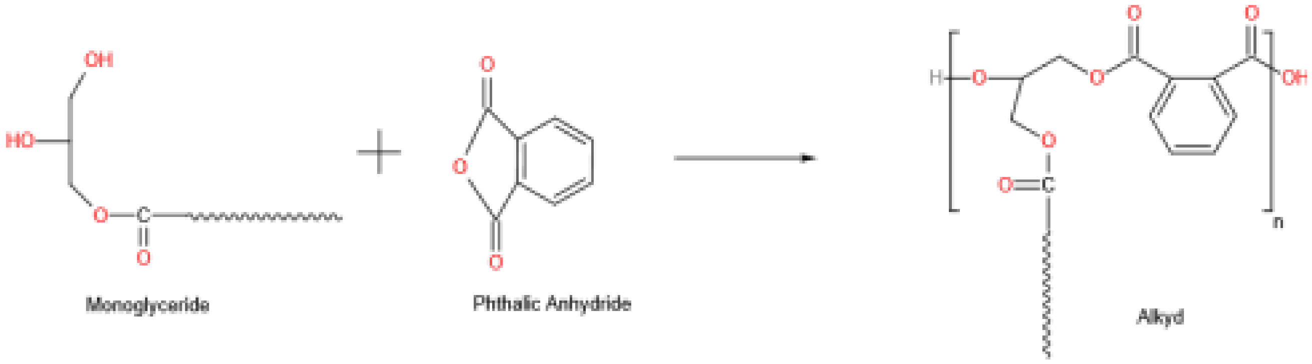


Fig. 9: Synthesis of Alkyd Resin

There have been many instances and recent developments in the use of oils to synthesize alkyd-based paints and coatings. Alkyd resins based on inexhaustible Jatropha Curcas oil and consisting of mixtures of phthalic and maleic anhydride in various ratios have been made by Boruah et al. The resin synthesized possessed ameliorated hardness, gloss, adhesion, and chemical resistance, which makes it ideal for binder composites, surface coating, etc. It also has high thermostability and an initial decomposition temperature of around 330°C. [82]

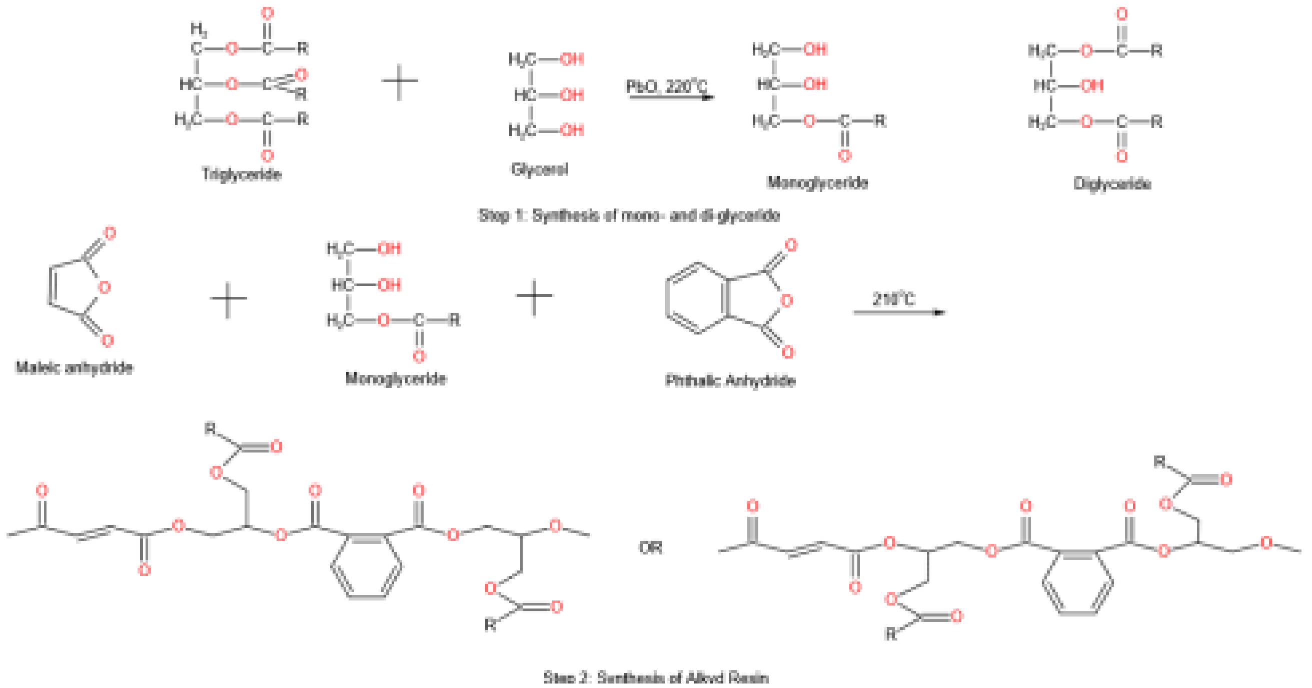


**Fig. 10: Monoglyceride process for the synthesis of alkyd resin**

For better flame retardancy, chlorinated rubber seed oil alkyd resins were used. They also showed better drying characteristics as compared to their unchlorinated counterparts. [86] To obtain advanced features in terms of drying time, flexibility, adhesion, chemical resistance, scratch resistance, and impact resistance Acrylated Albizia benth [ABO] medium oil alkyds are used. [87] Acrylated alkyds have also been used as binders in paints. Alkyd-acrylic hybrid binders have been synthesized using free radical polymerization of acrylic monomers in the showing of an unsaturated alkyd resin using the emulsion polymerization technique. Different ratios of alkyd and acrylic were used and it was found that the hybrid latexes display quicker drying compared to alkyd emulsion and the blend researched, suggesting that the drying mechanism of hybrids mirrors the drying process of acrylic latex in a greater proportion. It is concluded that alkyd-acrylic blends can achieve synergistic properties since they also provide homogeneous and smooth film surfaces along with penetration ability. [88]

Linseed oil alkyds are the most used alkyd resins in paints and coatings by a long shot. Linseed oil-based alkyds synthesized by D. ˙Is¸ eri-C¸ aglar ˘ et al. utilizes the usual monoglyceride process using trimethylolpropane and phthalic anhydride with calcium carbonate as a catalyst. They tested various properties of the paint formulations prepared, such as MEK rubbing, pencil hardness, pendulum hardness, gloss, adhesion along with contact angle. It was observed that by adding more quantity of huntite and Aerosol R972 in the paint composition, the thermal stability of paint improved. SEM studies conducted on the paint materials portrayed that silica particles were analogously dispersed throughout the organic matrix. [89] Linseed oil-based alkyds have also been used for designing high-performance coatings. Pathan and Ahmad have reported the synthesis of bio-hybrid transparent organic-inorganic coatings by attuning the properties of both linseed oil and 3-

isocynatopropyl triethoxy silane (IPTES) via sol-gel technique. Alkyd matrix was reinforced with mechanical, thermal, hydrophobic, transparency along with anti-corrosive properties because of the amalgamation of the soft and hard networks in a single material. This result was a combination of both a highly cross-linked network and the organic-inorganic bio hybrid nature and has the potential to be applied in transparent tapes for labeling & packaging, solar cells, and corrosion protective coatings. [90]

In the previous decade, a lot of focus has shifted towards developing novel coating systems for alkyds which comprise waterborne coatings, organic-inorganic hybrid coatings, and nanocomposite coatings. One such example is the waterborne alkyds synthesized by Aigbodion et al., which are based on rubber seed oil. Some of the key properties include environmental friendliness and low VOC. [91] The alkyd acrylate combination works in these new systems as well, which is proven by the work of Saravari et al. These resins were prepared by reaction between monoglycerides synthesized from palm oil and carboxy-functional acrylic copolymer succeeded by neutralization of carboxyl groups with diethanolamine. Homogeneous resin is formed when the copolymer quantity is 25-30% and the entirety of the water reducible acrylic-alkyd resins made were viscous yellowish liquids. Films of the same, which were obtained by baking at 190°C, showed good alkali resistance along with excellent water and acid resistance. The results also suggest that the reaction approach used is environmentally friendly. [92]

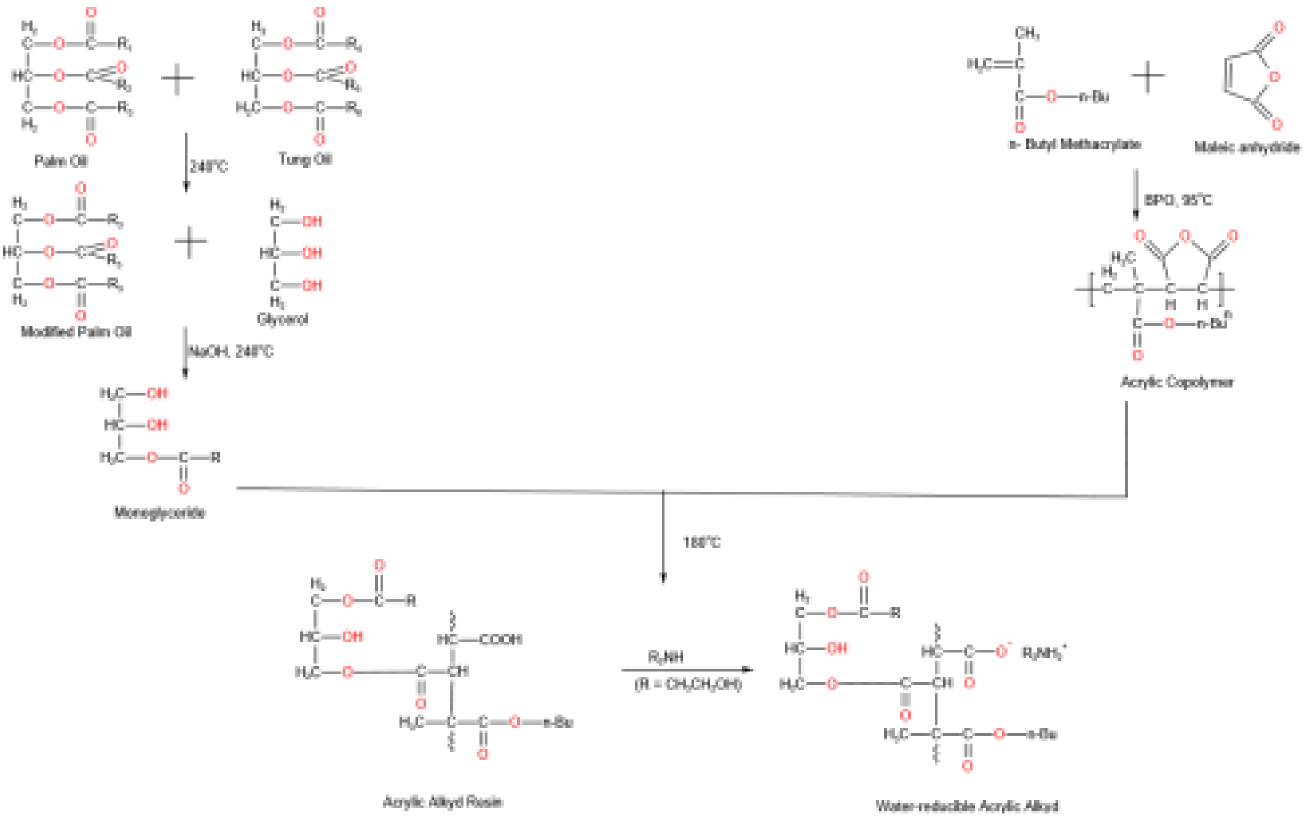


**Fig. 11: Synthesis of water reducible acrylic-alkyd resin**

Like acrylics, alkyds are also used in combination with other compounds/resins, such as urethanes. Athawale and Nimbalkar used acrylamide tertiary butane sulfonic acid as a monomer to develop emulsifiable air-drying alkyd resins that gave better thermal, chemical, and coating properties than those synthesized using the traditional dimethylol propionic acid as a monomer . [93] Hasebuddin et al. prepared a set of high solid alkyd polymers (80%) from soya oil fatty acid (SOFA) & dehydrated castor oil fatty acid (DCOFA) combinations with a differing percentage of dipentaerythritol (DPE, a hexafunctional polyol). High polydispersity, good gloss and gloss retention, and enhanced corrosion resistance with

decreasing DPE content are some noteworthy properties observed in these systems. [94] Waterborne alkyds see many other modifications too. Continuing with the VOC-free objectives of the coating industry, butylated melamine-formaldehyde (BMF)-modified soy alkyd (SA-BMF) were synthesized via a green route. Antibacterial activity was induced in the soy alkyd because of BMF, specifically against *S. aureus,* a gram-positive bacterium. The contact angle, Potentiodynamic Polarization (PDP), and Electrochemical Impedance Spectroscopy (EIS) studies proved that this coating has good corrosion protection performance against alkaline, acidic, and tap water media. Thus, it can be inferred that these coatings have potential applications in food packaging and surface coating industries. [95]

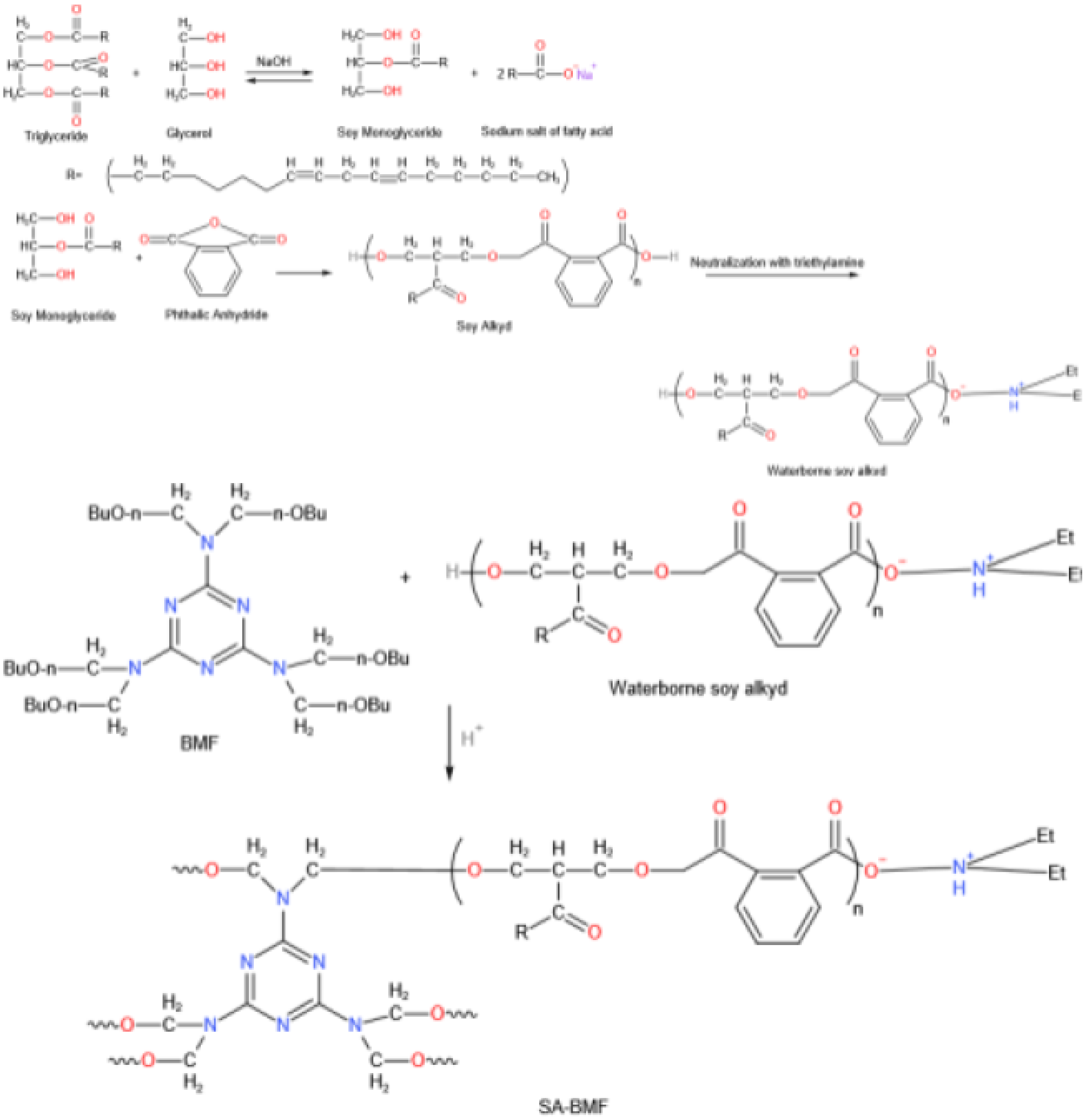


**Fig. 12: Synthesis of waterborne soy alkyd modified with butylated melamine formaldehyde**

The same type of modification can be done with waterborne castor oil alkyds. Waterborne castor alkyd cured using butylated melamine-formaldehyde (BMF) (WCA-BMF) were characterized utilizing different spectroscopic techniques. Because of being cured with s-triazine rings, these alkyds obtained better corrosion resistance, thermal stability, scratch hardness, and impact resistance behaviour. Studies on contact angle and electrochemical corrosion showed that the WCA

BMF coating show improvised anti-corrosive properties compared to previously obtained waterborne systems. [96] Castor oil is also used to prepare other modified resins like urethane alkyds. Castor oil is interesterified with jatropha oil followed by a reaction with toluene diisocyanate, yielding urethane alkyds. The alkyd synthesized had a lesser molecular weight and viscosity, marginally lower hardness, and substantially longer drying time than the traditional and commercial urethane alkyds. Apart from this, however, the properties of the film formed were broadly similar, inclusive of high flexibility coupled with remarkable adhesion and great impact resistance. In addition, they also displayed brilliant resistance to water and acid. [97]

Alkyds have also seen their fair share of nanocomposites use as well. The preparation of nano ferrite alkyd compositions was made to enhance the coating properties of plain alkyds, and they were able to do it as well. Soya oil alkyds containing nano-polyaniline were used for the same. [24] Khanna and Dhoke have conducted similar work on the effects of $Fe_2O_3$/ZnO nanoparticles in waterborne alkyd coatings. [98,99] The incorporation of $Fe_2O_3$ nanoparticles in waterborne alkyd coating composites displayed improved corrosion resistance, better mechanical properties including abrasion, scratch hardness, and enhanced UV blocking properties. The increase in nano- $Fe_2O_3$ concentration resulted in the improvement of properties as well. [98] On the other hand, the nano-ZnO composites had their properties tested with Scanning Electron Microscopy (SEM) and Atomic Force Microscopy (AFM). These compositions showed amelioration in corrosion resistance, UV resistance, and mechanical features like scratch and abrasion resistance of these coatings, indicating the constructive impact of the incorporation of nano-ZnO particles in the coatings. [99] These ZnO nanoparticles are also included in modified alkyd resins. Alkyd-based silicone-modified waterborne coatings prepared to utilize hexamethylmethoxymelamine (HMMM) as a cross-linking agent and para-toluene sulphonic acid (p-TSA) as catalyst also see the incorporation of nano-ZnO. The addition of nanoparticles resulted in enhanced heat resistance and mechanical characteristics such as scratch and abrasion resistance. Potential applications include automotive industries, heaters, smokestacks, furnaces, stoves, and incinerators, and further applications where extreme temperatures are involved. [100]

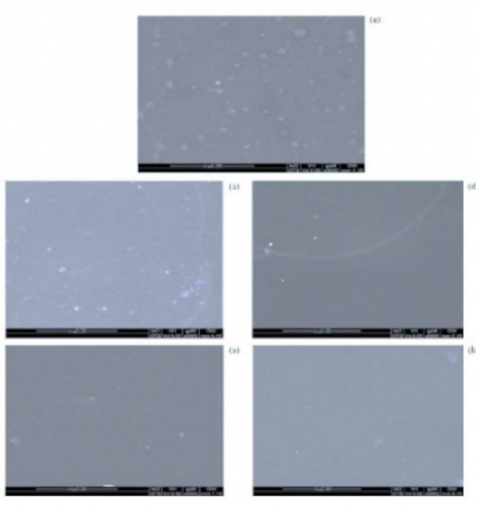


Fig. 13: SEM images of nano-ZnO and nano-$Fe_2O_3$ coatings

## Polyurethanes:

Polyurethanes (PU) are polymers synthesized by polyaddition between di- or poly-isocyanates like toluene diisocyanate, diphenylmethane diisocyanate, etc., and di- or poly-hydric alcohols or from compounds having two active hydrogen atoms. As for the use of vegetable oils in polyurethanes, polyols and amide diols of vegetable oils are utilised as commencing ingredients for PU synthesis in addition to aliphatic and aromatic isocyanates. [101,102] Yadav et al. synthesized poly (urethane fatty amide) from diol linseed fatty amide and toluene-2,4(6)-diisocyanate in varying ratios with the use of a one-shot technique at normal temperature

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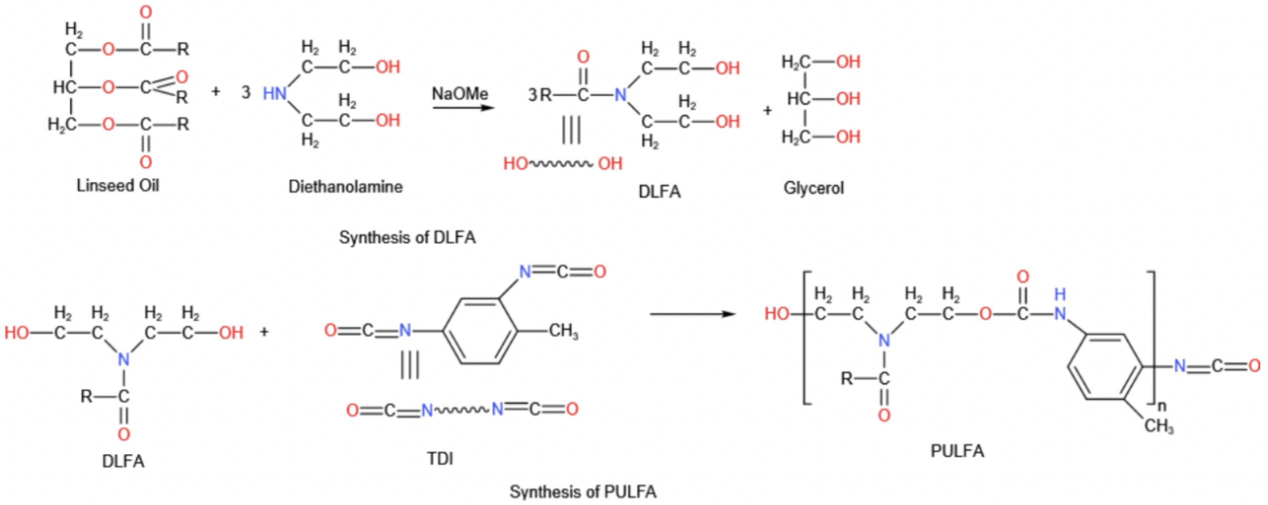


**Fig. 14: Synthesis of Poly(urethane linseed oil fatty amide)**

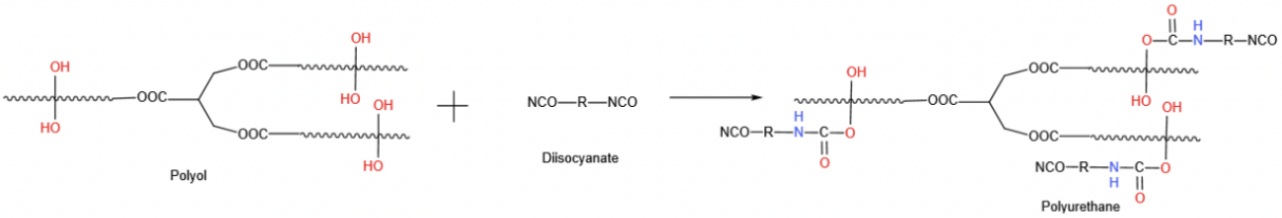


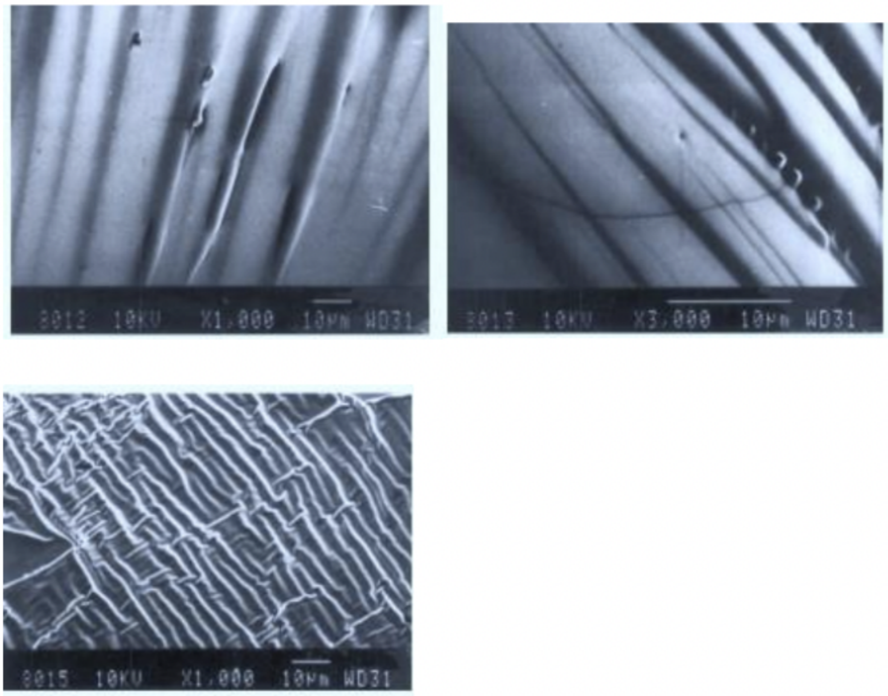
**Fig. 15: Synthesis of polyurethane using polyol and diisocyanate**

The main highlights were the very low requirement of energy as compared to its alkyd, urethane, and uralkyd counterparts and the use of the coatings as a corrosion-protective eco-friendly coating substance up to 230°C. [101] If prepared by using Pongomia Glabra oil, the polyurethane fatty amide provides good physiomechanical properties, excellent chemical/corrosion resistance, and moderate antibacterial properties. When compared with the polyester amide prepared using the same oil, the resin showed superior alkali resistance. [102] PU coatings are very versatile when it comes to synthesis i.e., they can be synthesized from a multitude of compounds with hydroxyl end groups. These include amide diols [101,102], alkyds [93], polyesteramides [43], polyether amides [102,103] and also their modified counterparts acrylated [104] and metal/metalloid incorporated polymers. [43] Urethane modification is used very widely for improving the various properties of polymers and resins. For example, urethane modification in polyether amides results in great improvement in anti-corrosive properties, glass transition temperature, and coating properties in general. [102,103]

Polyurethanes based on vegetable oils majorly contain urethane moieties along with functional groups such as amides, esters, vinyls, acrylics, etc. These groups often help in improving features like impact resistance, adhesion, flexibility, and scratch hardness. [102-104] The conventional structure of a polyurethane derived from a VO diol is given below. The proportion of degradable or bioderived content in such polyurethanes depends on the diol/polyol & isocyanate.

Another interesting type of polyurethane synthesized from vegetable oils is PU interpenetrating polymer network [IPN]. Interpenetrating polymer network or IPN is a type of polymeric network in which one polymer undergoes polymerization in the immediate presence of another. The common monomers used include VO-based polyols, acrylic monomers, and isocyanates. [105,106] Ashraf et al. synthesized semi-IPNs with polypyrrole and coconut oil-based poly(ester amide urethane) using the immersion method, which proved to be a simple path for developing conducting polymers with films properties like decent mechanical strength, flexibility, and proper conductivity. The SEM images of these conducting films are in the pictures below (fig. 16).[105]

Fig. 16: SEM images of conducting films



Stoving paints can also be prepared with the help of polyurethanes. Dutta et al. prepared two types of stoving paint systems using polyurethanes. The first system was prepared from partially butylated melamine-formaldehyde resin modified Mesua ferrea L. seed oil (MFLSO) poly(urethane ester) (PUE) in the ratio 70:30 while the second system consisted of bisphenol A-based epoxy resin with MFLSO-based PUE modification in the ratio 50:50. While both the paint systems were extremely well suited to be used as industrial paints, testing of the various characteristics revealed that the epoxy-based ones proved to be better than the melamine-formaldehyde ones.[107] The main differentiator that polyurethane coatings possess over their counterparts with other resins is their ambient curing temperature. The curing temperature can also be lowered by treating the coating material with isocyanate and this is generally done when the curing/drying temperature goes beyond 100°C.

There is also the added advantage of activity imbalance between the two isocyanate groups in TDI-based polyurethanes.[5] [43,108] In general, the curing of polyurethane coatings comprises three steps: solvent evaporation, chemical reaction of the free isocyanates and auto-oxidative cross-linking.

One of the biggest disadvantages of vegetable oil-based polyurethanes is their low glass transition temperatures ($T_g$). In an attempt to test the extent of this difference as compared to any other polymer, Janvi et al. conducted several experiments to certify the degradation of VO-based polyurethanes obtained from several different oils like castor oil, soybean oil, canola oil, arcol oil, etc. and compared them with the degradation of PPO-based (polypropylene oxide) polyurethanes. It was found that oil-derived polyurethanes possess better initial thermal stability in the air as compared to PPO-based polyurethanes, while it was reversed when in nitrogen. Polyurethanes based on oils were more stable when higher conversion rate weight loss is the established criteria. The FTIR spectra corresponding to the non-degraded and the degraded polymer for 60 minutes are as given below (fig. 17).[109]

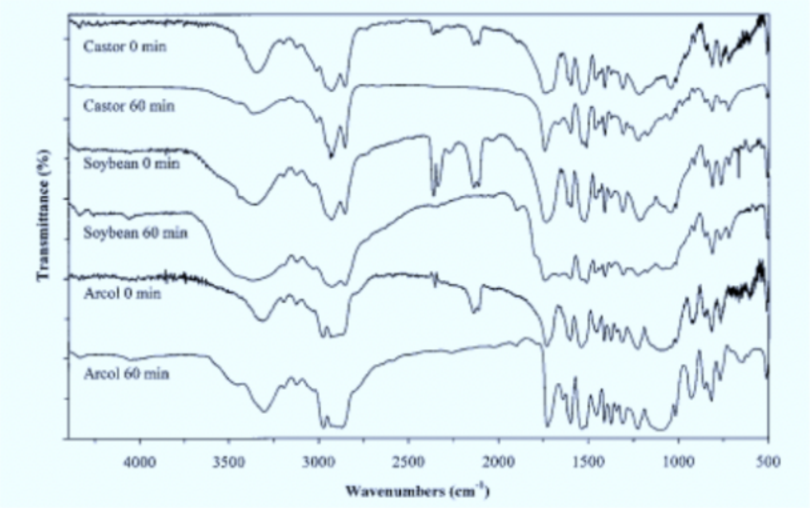


**Fig. 17: FTIR spectra for degraded and non-degraded polymers**

This low $T_{g,}$ however, also has its own remedies. Polyols yielded by coupling of epoxidized soybean oil and lactic acid can be used to prepare polyurethanes. These exhibit higher glass transition temperatures of up to 31 °C-96 °C. [5] Likewise, the inclusion of inorganic segments in the polyurethane proved to improve the $T_g$ as well. De Luca et al. synthesized new inorganic-organic hybrids via reaction between castor oil/epoxidized castor oil and tetraethoxysilane (TEOS). Along with the lower $T_g$, good adhesion was absorbed for all concentration ratios. An increase in TEOS concentration resulted in higher hardness and tensile strength while the homogeneity went down. These are used as creamer coatings. [110] Boron incorporation is another method to improve thermal stability and acid resistance in polyurethanes. [111] Other properties such as corrosion, chemical and water resistance of dried films are greatly influenced by the type and content of fatty acid oligomer along with the ionic composition in polyurethanes. [112,113]

Polyurethanes also have advanced and sophisticated applications like all the other resins discussed previously. Deka and Karak have found surface coating applications for high-performance hyperbranched polyurethanes. The hyperbranched polyurethane is synthesized using Mesua ferrea L. seed oil monoglyceride, 2,4-toluene diisocyanate, poly(- caprolactone)diol, and glycerol without a catalyst (see fig. below). While the physical, mechanical, and thermal performance depends on the segmental ratios, it was much superior when weighing against a traditional linear polymer. [114]

Soybean oil-based polyurethanes have recently found great application in the electrical insulation industry as they offer excellent thermal and electrical resistance. Epoxidized soybean oil was successfully converted into carbonated soybean oil, which was further converted into a soybean oil-based polyol to be used in polyurethane synthesis. The materials formed were considered formidable electrical insulators because of the obtained values of DC, DS, DF, solderability, moisture uptake, and solvent resistance. [115]

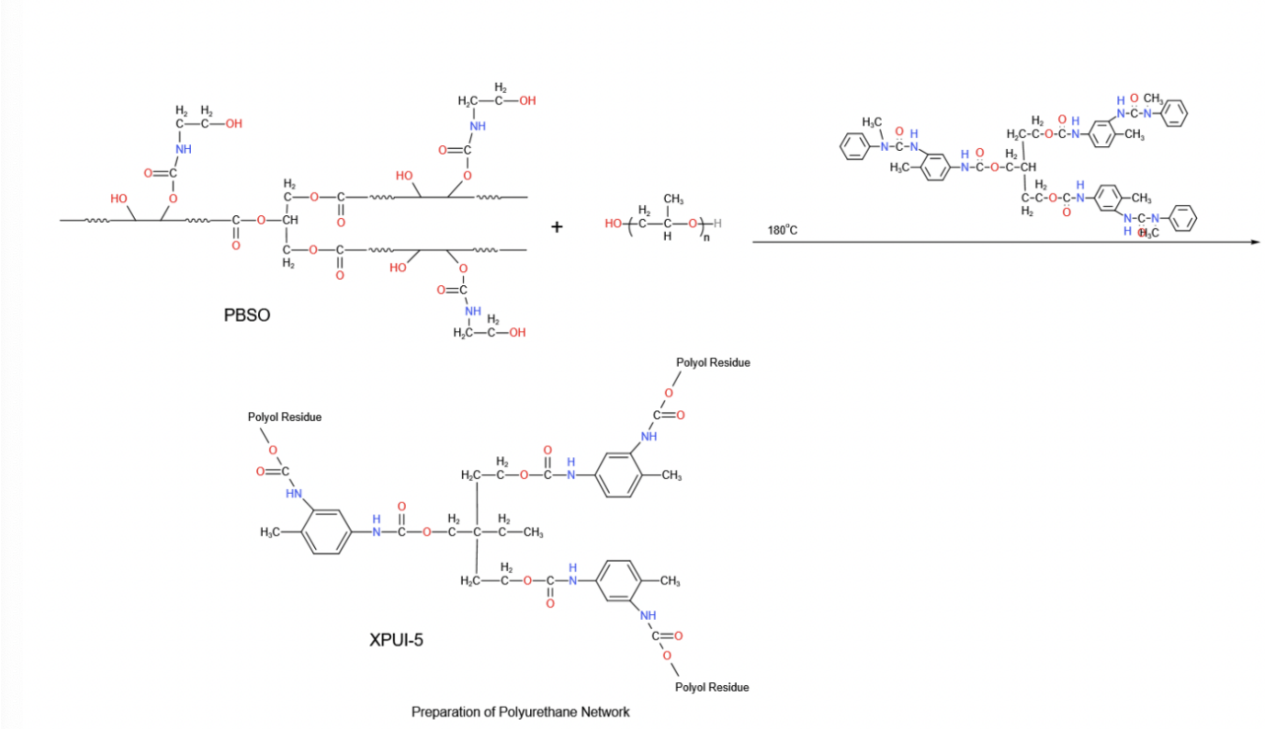


**Fig. 18: Synthesis and Preparation of Polyurethane Network**

Polyurethanes have been used in industrial maintenance as well. Two-pack polyurethane coatings based on acrylic copolymer with acid functionality and modified using alkyds were used as coatings for Industrial Maintenance. The alkyds were rubber seed oil-based and the coatings showed good chemical and thermal resistance. [116] Koprululu et al. synthesized urethane macromers based on triglycerides which are used for copolymerization. This urethane macromer was reacted with styrene to give a yellow coloured flexible transparent film. The film displayed good acid, but poor alkali resistance and the existence of styrene in the film was tantamount to the increased thermal stability and decreased hydrophilicity.[117] Insulating coatings of poly(ether-ester urethane)s were prepared using polyols made from PET (polyethene terephthalate), adipic acid, castor oil, and blocked isocyanates. Subsequent testing showed that these coatings were not only environmentally friendly, but they also gave a high performance in metal-insulator coatings and were referred to as 'Green Coatings.' They possessed excellent mechanical and electrical insulation properties. [118] Similar to polyester amide coatings discussed earlier, virgin polyurethanes, as well as polyurethanes with metal/metalloid components, have been proven to exhibit antimicrobial as well as anti-corrosive behaviour because of isocyanate, metal, and urethane parts present.

To summarise, a crucial part of society is fulfilled because of polyurethanes having a multitude of uses.

## Conclusion:

Vegetable oils are plentiful in nature, easily procured and cost-effective. They have a variety of properties and functional groups enabling them to find a multitude of applications. Various coatings with enhanced properties and better functionalities as compared to their counterparts can be designed using vegetable oils. Despite the use of vegetable oils in coatings being decades old, it is still prevalent and developing. With extensive and persistent research efforts, coatings based on vegetable oils will be able to establish themselves as the 'green alternative' to their petroleum-based counterparts and become the mainstream in almost every type of application.

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