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Bio-Renewable Sources for Synthesis of Eco-Friendly Polyurethane Adhesives—Review

Ravindra V. Gadhave, Prakash A. Mahanwar, Pradeep T. Gadekar

Department Polymer and Surface Engineering, Institute of Chemical Technology, Mumbai, India
Email: ravi.gadhave3@gmail.com

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Abstract

Bio-renewable sources used during manufacturing of polyurethane (PU) adhesives have been used extensively from last few decades and replaced petrochemical based PU adhesive due to their lower environmental impact, easy availability, low cost and biodegradability. Bio-renewable sources, such as vegetable oils (like palm oil, castor oil, jatropha oil, soybean oil), lactic acid, potato starch and other bio-renewable sources, constitute a rich source for the synthesis of polyols which are being considered for the production of "eco-friendly" PU adhesives. Various bio-renewable sources for synthesis of bio-based PU adhesives and their potential applications are discussed in this review. This paper will focus on the progress of research in bio-based materials for adhesive application.

Keywords

Bio-Renewable, Polyol, Oil, Polyurethane, Adhesive

1. Introduction

Polyurethanes are up to date the most versatile polymers due to the flexibility of structure design at the application site. They are mainly used in footwear, packaging, automotive industry and furniture assembly in the form of rigid and flexible foams, coatings, adhesives, sealants, elastomers and binders. Increasing environmental awareness among producers and consumers has led to substantial interest and research in biomass resource stead of petrochemicals for PU synthesis [1]-[10].

Polyurethane as wood adhesives has developed a reputation for reliability and high performance [11] [12]. The performance and behavior of adhesive systems for wood depend on a wide range of variables, such as smoothness of substrate surfaces, pH, presence of extractives, and grain direction [13]. The bonding

mechanism of the adhesive to the wood substrate can include covalent bonding, weaker forces such as van der Waals forces and hydrogen bonding, or mechanical interlocking [14] [15]. Adhesives based on urea-formaldehyde, melamine formaldehyde, urea melamine formaldehyde and phenol-formaldehyde are commonly used [16] [17] [18], but are very sensitive to hydrolysis [19] [20] [21]. These adhesives also produce health hazards because of the formaldehyde they release [22]. To overcome such problems, scientists are trying to develop new polymeric adhesives [23] [24] [25]. PU adhesive has developed a reputation for reliability and high performance [25] [26].

An attempt has been made to develop PU adhesives that were at least partially made from the natural materials like natural vegetable oils [29]-[34], lignin, lactic acid and potato starch [27], and edible or non-edible plant-derived oils. Bio-renewable resources constitute a rich source of precursors for the synthesis of polyols and isocynates which are being considered for the production of "greener" PU adhesives [28] and reaction between polyol and isocynate is shown in **Figure 1**. Various bio-renewable sources for synthesis of bio-based PU adhesives and their potential applications are discussed in this review. This paper will focus on the progress of research in bio-renewable sources for adhesive application.

2. Vegetable Oil Based Polyurethane Adhesive

Polyurethanes are synthesized from vegetable oils obtained from various plant seeds such as castor, jatropha, palm, soybean etc [35]-[42]. Large quantity of plants oils are important renewable source to make soaps, surfactants, lubricants, diluents, plasticizers, inks, agrochemicals, composite materials, food industry etc. [43] [44] [45] [46]. The major components of vegetable oils are triglycerides which are esters of glycerol with three long chain fatty acids having varying composition depending on the source of oil [47]. Structure of triglyceride is shown in **Figure 2**. Polyols and polyisocyanates which are major raw materials for the synthesis of polyurethanes can be readily synthesized by utilizing these reactive sites [48]. Upon hydrolysis, triglycerides of vegetable oils give different fatty acids and glycerols. There are many factors which affect the properties of oil based coatings such as composition of various saturated and unsaturated fatty acids, extent of un-saturation, chain length of fatty acids, location and stereochemistry of the double bonds in fatty acid chains [49] [50]. Fatty acids can be a part of polyester polyols whereas hydroxyl functional vegetable oils are directly used in polyurethanes [51] [52] [53]. In past and present, various bio renewable PU are synthesized from vegetable oils as well as other bio renewable sources [54] [55] [56]-[93].

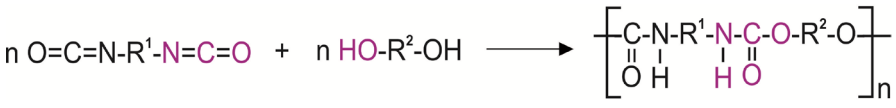

Figure 1. Reaction between polyisocynate and polyol.

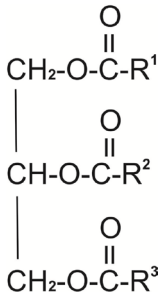

Figure 2. Structure of triglyceride.

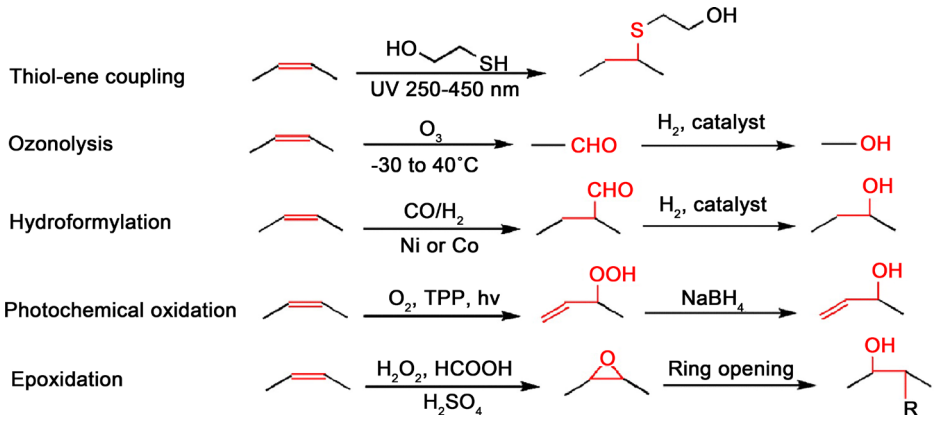

Figure 3. Synthesis method of vegetable-oil-based polyol.

wherein R^1, R^2 and R^3 are independently saturated or unsaturated aliphatic hydro-carbyl groups containing from about 8 to about 24 carbon atoms.

Synthesis of polyols

Many routes for the production of vegetable-oil-based polyols are reported including thiolene coupling reaction [94] [95], ozonolysis [96] [97], hydroformylation [98], photochemical oxidation [99], epoxidation [100] [101] [102] [103] followed by ring opening reactions as summarized in **Figure 3**. Main reaction route for synthesis of polyols is epoxidation and then reaction of the epoxy groups with different ring-opening reagents such as water [104] [105], alcohol [106], glycerol, 1,2-propanodiol [107] and acids [108] [109].

2.1. Castor Oil

Castor oil is a major candidate in these replacement efforts due to its inherent advantages over other vegetable oils [110]. Besides its renewability, low cost and easy availability in large quantities, castor oil is not edible, and does not compete with food, and has free secondary hydroxyl groups. Approximately 90% of fatty acids in castor oil are ricinoleic acid (C18:1), which have a hydroxyl functional group at the 12^{th} carbon. This provides a hydroxyl value of between 160 and 180 mg KOH g^{-1} [111] [112]. However, this low hydroxyl value along with the presence of secondary hydroxyls results in low functionality and low reactivity [113] [114], leading to low crosslinking density, which consequently produces semi-flexible and semi-rigid materials among other limitations [115].

PU adhesive was prepared using polyols obtained from castor oil modified by a trans-esterification reaction with pentaerythritol [116]. Other work describe the performance of castor oil based novel polyurethane adhesive system for wood to wood and metal to metal bonding by using polyester polyols, castor oil-polyester polyols and epoxy-polyester polyols with different isocyanate adducts having different NCO/OH ratio. Castor oil-polyester polyols were synthesized through the transesterification reaction between castor oil and polyester polyols [117]. Castor oil based polyester polyols were synthesized by the condensation polymerization of different dicarboxylic acids such as, maleic acid, fumaric acid, and oxalic acid with castor oil. The prepared polyester polyols were used in the preparation of wood adhesives [118]. Reaction of polyurethane wood adhesives from obtained polyester polyols based on castor oil is shown in Figure 4.

2.2. Jatropha Oil

Jatropha oil-based polyol was prepared using two different methods, that is, hydroxylation followed by either alcoholysis or epoxidation of jatropha oil. Epoxidation method of jatropha oil-based polyol had been described in the literature [122]. The synthesized polyol was treated with diisocyanate at 70˚C to generate the PU adhesive [119] [120] [121] [122] [123]. Reaction scheme is shown in Figure 5.

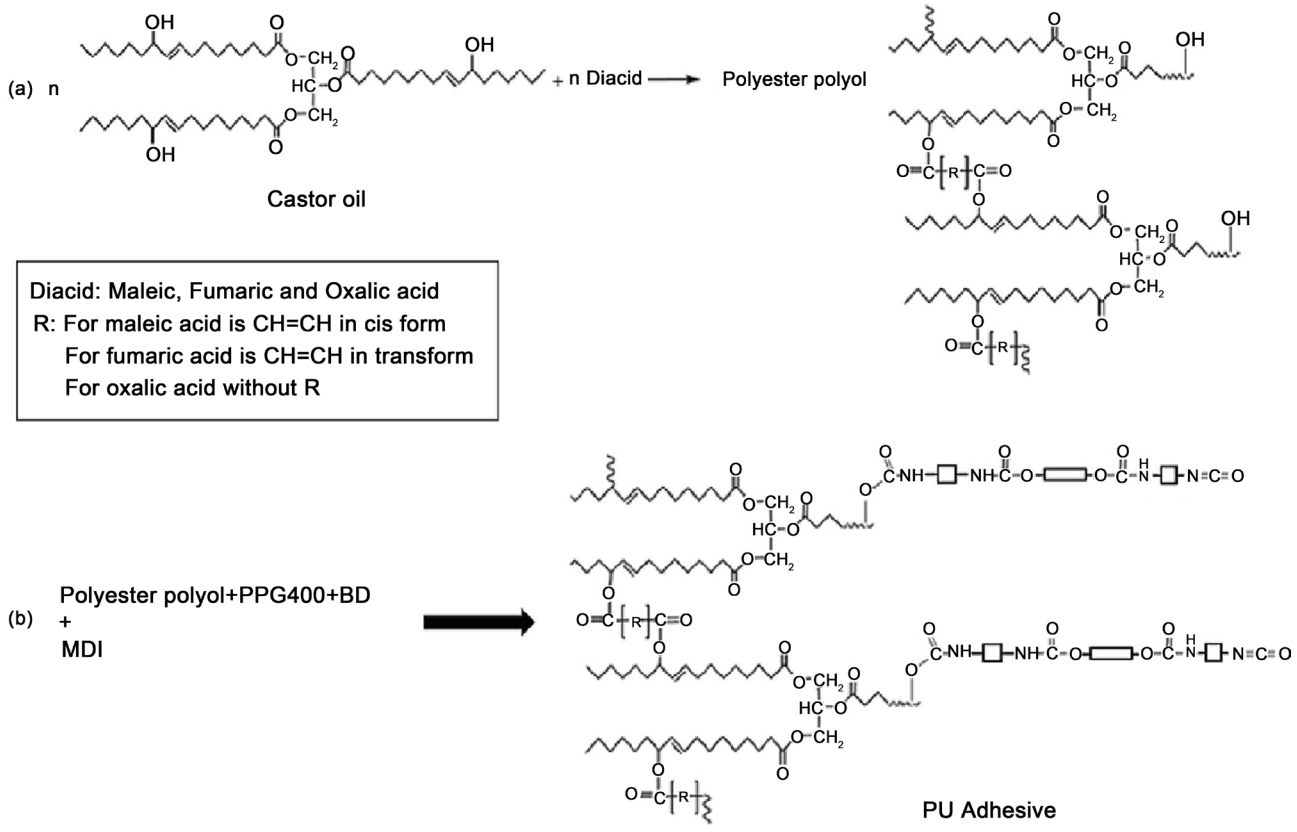

Figure 4. Synthetic scheme of polyurethane wood adhesives from obtained polyester polyols based on castor oil.

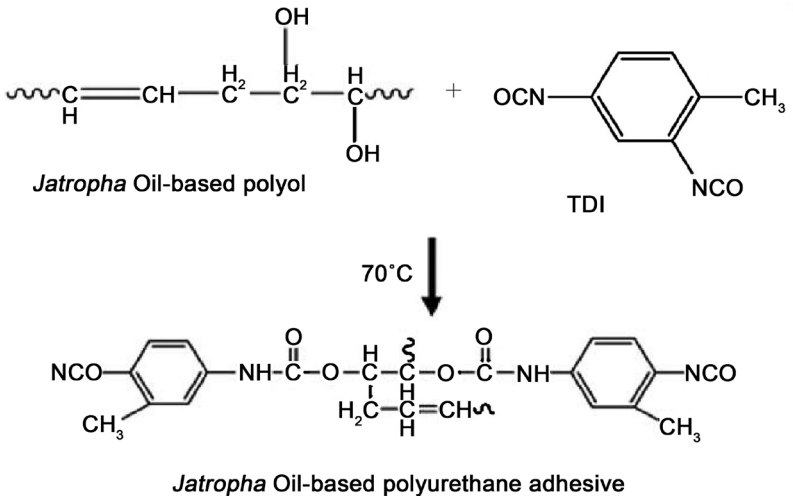

Figure 5. Hydroxylated polyol and prepolymer adhesive reaction with 2,4-toluene diisocyanates.

2.3. Multi-Hydroxy Soybean Oil (MHSBO)

The research was conducted to evaluate the possibility of using MHSBO/pMDI resin, obtained from soybean oil, as a wood adhesive. MHSBO was reacted with pMDI resin at different eq. mole ratios to prepare adhesives. These adhesives were used to investigate adhesive properties and bond performance in wood application [124] [125] [126]. Reaction of Synthesis of MHSBO and polyurethanes is shown in **Figure 6**.

2.4. Palm Oil

PU adhesive was prepared from palm oil-based polyester polyol [32] [131] that was synthesized by ring-opening reaction of epoxidized palm olein with phthalic acid. Phthalic acid was used to react with epoxidized palm olein to improve the hydrolytic stability of PU adhesive as cross-linking reaction increases the content of dangling fatty acid chains in the polyol to improve the hydrophobicity of PU adhesives [127]. Besides, the dangling fatty acid chains of the palm oil-based polyester polyols act as plasticizer that gives more flexible adhesive films [128] [129] [130]. Performance of this adhesive system was compared with commercial available wood adhesives. Reaction scheme is shown in **Figure 7**.

3. Polylactic Acid Based Polyurethane Adhesive

Polylactic acid was commercial thermoplastic polyester known for its biodegradability [132]. Preparing polyols from lactic acid (lactate polyols) as precursors for polyurethanes would be beneficial in several ways. Lactate polyols are polyester polyols containing lactic acid units. Introducing lactic acid units into a polyol structure can be done in different ways.

One route was the ring opening addition of lactide to hydroxyl groups. Other routes involve esterification of different polyols with lactic acid, or transesterification with esters of lactic acid (e.g., ethyl lactate, butyl lactate). The advantage

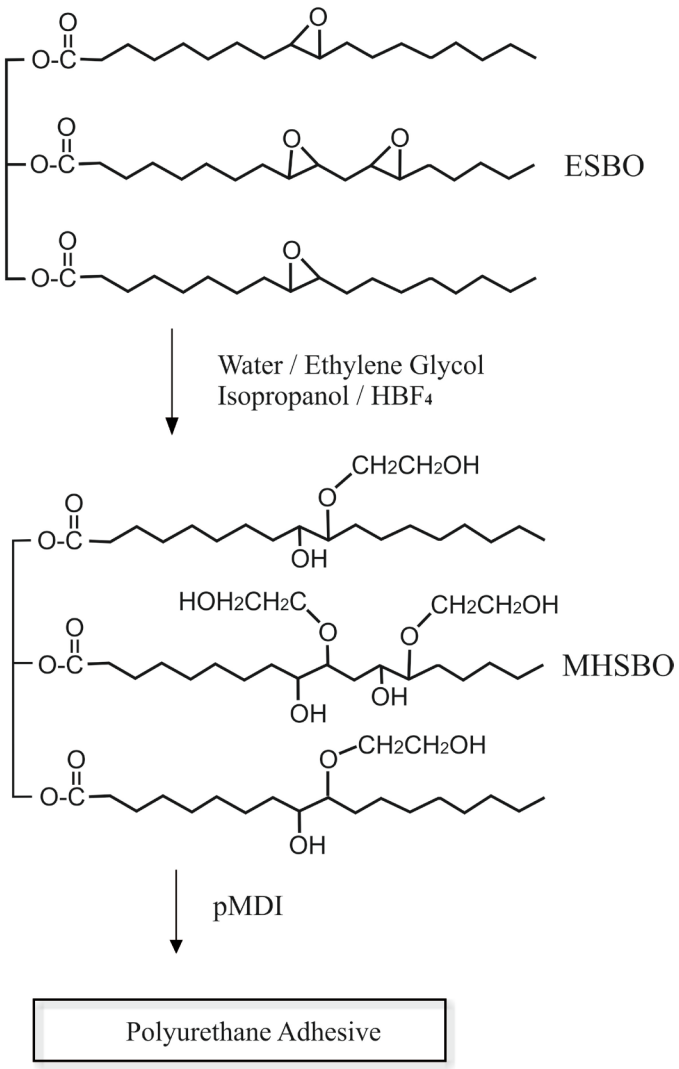

Figure 6. Synthesis of MHSBO and polyurethanes.

of the addition of lactide to polyols was in short reaction times and avoidance of the removal of low molecular weight compounds (water or alcohols). Lactate polyols can be prepared from 100% bio-renewable feedstock. Present work was to prepare high functionality polyester polyols containing lactic units (lactate polyols) suitable for preparation of rigid cast polyurethanes and foams. This PUs, apart from having high bio-based content, was expected to be biodegradable. The ring opening addition of L-lactide to hydroxyl groups was the reaction used for introducing lactic acid units into polyol structure. In order to obtain high functionality polyols with high bio-based content, different polyglycerols with high content of OH groups were used as starters. However, high concentration of OH groups in simple polyglycerol-lactate adducts and strong tendency to crystallization of lactate units result in strong intra-molecular hydrogen bonding, which makes these polyols immiscible with isocyanates. Consequently, this issue was addressed by incorporation of hydrophobic fatty acid segments in the lactate polyol structure, which are known to have good affinity for isocyanates.

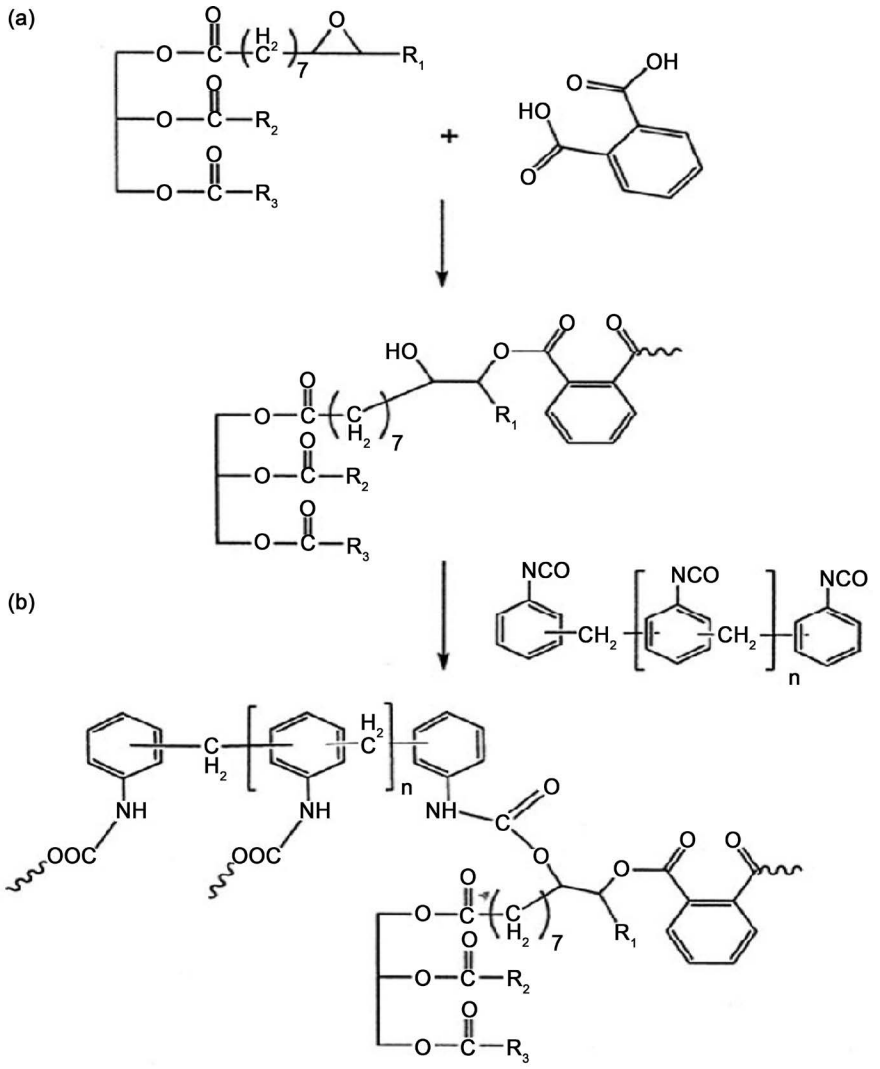

Figure 7. (a) Ring-opening reaction of EPOO by phthalic acid to form polyester polyol. (b) PU formation by reacting polyester polyol with pMDI.

Synthesized lactate polyols were reacted with isocyanate to obtain rigid cast polyurethanes [133] [134].

4. Potato Starch and Edible or Non-Edible, Plant Derived Oils

Development of a PU adhesive that is at least partially made from the natural materials potato starch and edible or non-edible, plant derived oils. Because the hydroxyl functionality of the polyol plays an important role in the formation of PU adhesives, polyols having different hydroxyl values were prepared. This was done by means of glycosylation of starch, followed by transesterification with oil to yield polyol [27].

1) Glycosylation of starch

Method of glycosylation was discussed here [135] [136]. The glycol glycoside so prepared, a mixture of a-D glycol glycoside and b-D glycol glycoside was used to synthesize polyols. The reaction is shown in **Figure 8**.

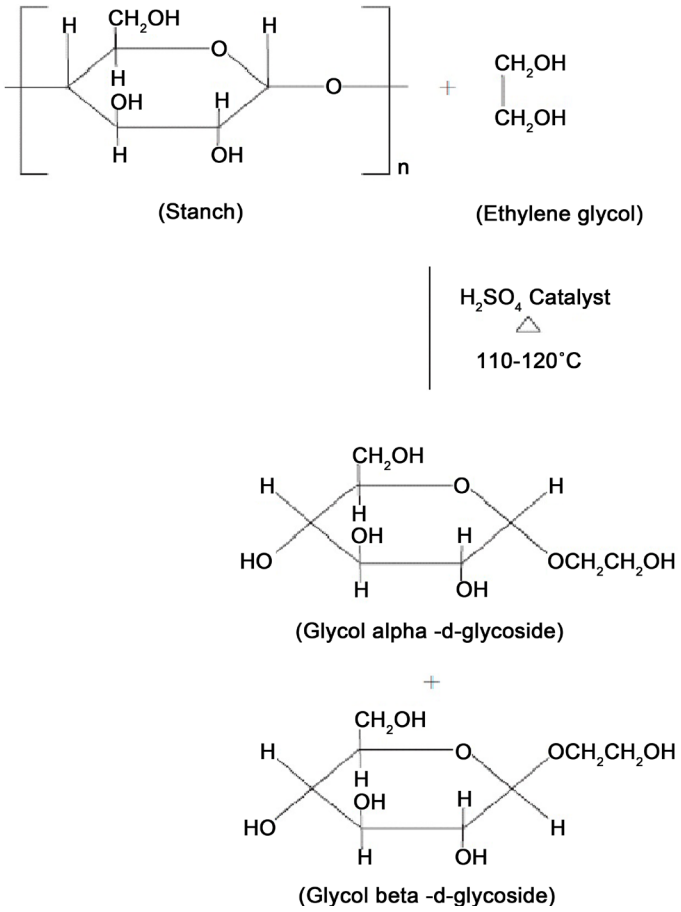

Figure 8. Glycosylation of starch.

2) Alcoholysis of oil with glycol glycoside

Polyols having varying hydroxyl values were synthesized by varying the ratio of glycol glycoside to oil. The reaction is shown in **Figure 9**.

5. Lignin Based Polyurethane Adhesive

Many tons of lignin is generated as by-products of industrial processes such as pulp and paper. Most of the lignin extracted from pulp and paper operations is burned during pulp-spent liquor treatment. This offers energy recovery and regeneration of pulping chemicals with less than 2% recovered for utilization as a chemical product [137] [138]. However, the amount of lignin produced exceeds the requirements for energy generation. The type of pulping process determines the type of lignin industrially available because it unavoidably modifies the lignin structure from that in the original feedstock.

To increase the potential applications of lignin in polymeric materials, some chemical modifications have been developed [139] [140], but these add stages to the process and/or raise their costs considerably. Therefore, the direct use of industrial lignin is the most favorable option because it is a relatively cheap raw material. Unmodified lignin has poor stability [141] and difficult melt processing [142], which make its direct use uncompetitive. However, many studies have

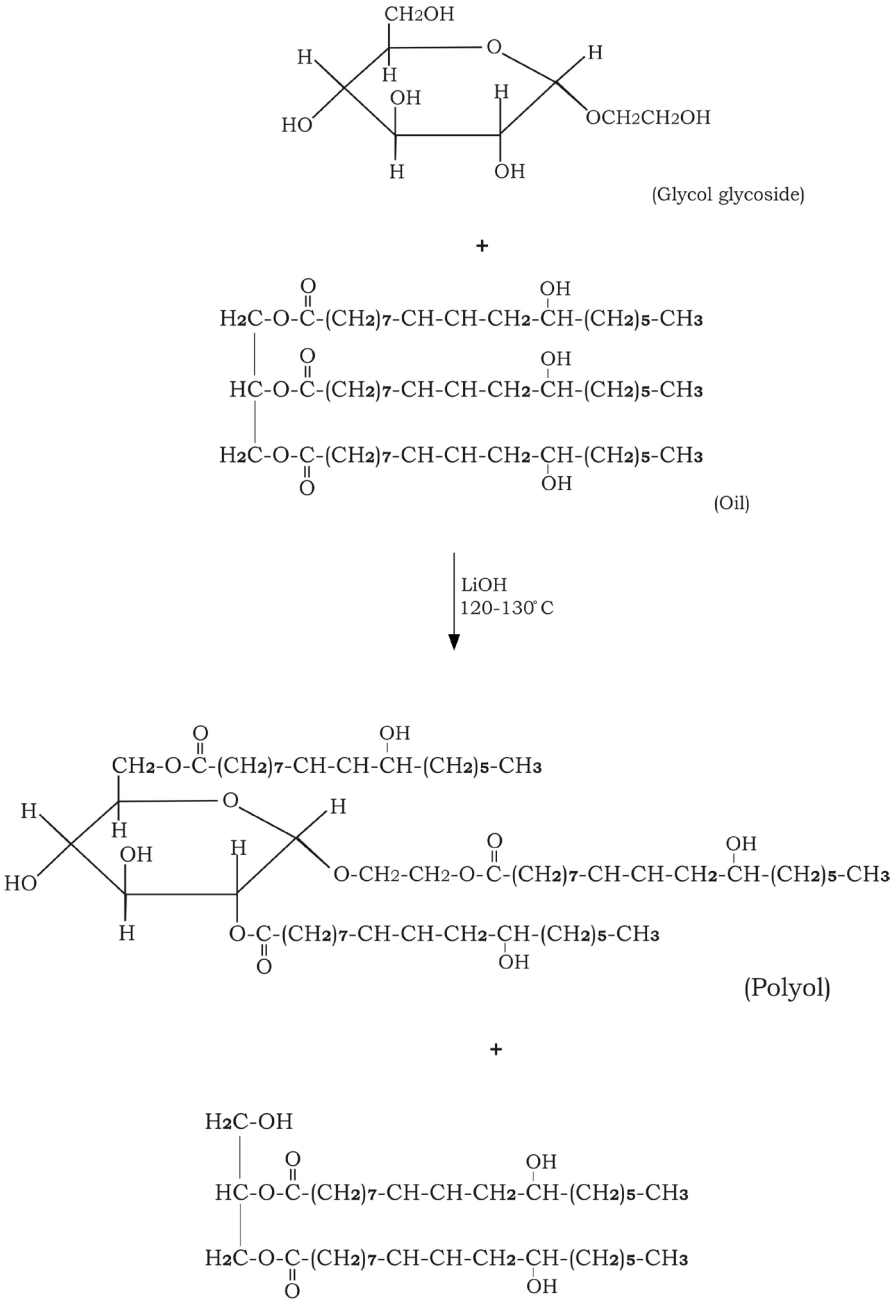

Figure 9. Alcoholysis of oil with glycol glycoside.

focused on the incorporation of lignin in polymer materials by blending it with synthetic or other bio-based polymers [143] [144] [145].

6. Conclusion and Future Perspectives

Production of bio renewable PU adhesives from bio resources like plants, trees and algae, is a new area of research due to declining of non-renewable feedstock. Epoxidation is the main route for the synthesis of polyols from vegetable oils (VOs) followed by ring opening reactions. Bio-based di-isocyanate synthesized from fatty acids gives PU with similar physical properties as PU derived from

petroleum based raw materials. On the other hand faced with declining reserves of fossil fuels, bio-based PU adhesives offer a solution for future challenges because in the future, new products for all industries like coating, paper, packaging, pharmaceutical and textile, for instance, could be created. Some of the chemical companies had already started the bio-based research years ago and other even position bio-based application as future strategy.

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