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Review

Sustainable Bio-Based Epoxy Technology Progress

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Abstract: Sustainable bio-based epoxy technology is developed by using bio-based epoxy materials instead of conventional fossil-derived ones. Significant progress in new bio-based epoxy material development on bio-based epoxy resins, curing agents, and additives, as well as bio-based epoxy formulated products, has been achieved recently not only in fundamental academic studies but also in industrial product development. There are mainly two types of bio-based epoxy resins: conventional epoxy resins and novel epoxy resins, depending on the epoxy resin building-block type used. Bio-based conventional epoxy resins are prepared by using the bio-based epichlorohydrin to replace conventional fossil-based epichlorohydrin. Bio-based novel epoxy resins are usually prepared from epoxidation of renewable precursors such as unsaturated vegetable oils, saccharides, tannins, cardanols, terpenes, rosins, and lignin. Typical bio-based curing agents are bio-based polyamines, polyamides, amidoamines, and cardanol-based phenalkamine-type curing agents. Cardanol is a typical bio-based reactive additive available commercially. Certain types of partially bio-based formulated epoxy products have been developed and supplied for use in bonding, coating, casting, composite, and laminating applications.

Keywords: sustainable; bio-based; epoxy resin; curing agent; additive

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1. Introduction

Epoxy resin forms very strong and durable bonds with most materials and is the most important thermoset, widely used as a coating, adhesive, and composite in various applications ranging from consumer goods, general industry, construction and building, electrical and electronics assembly, wind energy, and automobile production to aerospace and the defense market [1–10]. Epoxy systems are typically formulated in the form of a compound containing epoxy resin and a curing agent with modifiers and additives. Most of these epoxy resins, curing agents, and reactive additives used today still originate from fossil fuels.

Sustainable epoxy technology development has become more and more important nowadays to meet the expectations from global environmental protection, government regulation restriction, and customer requirements, focusing mainly on health and safety improvement, energy saving, and circular economy contribution [11–18]. Sustainable bio-based epoxy technology is developed primarily for circular economy contribution by using bio-based epoxy materials instead of conventional fossil-derived ones. Significant progress in new epoxy material development on bio-based epoxy resins, epoxy curing agents, and additives, as well as epoxy formulations, has been achieved recently not only in fundamental academic studies but also in industrial product development. The latest developments in fundamental academic studies have been introduced and reviewed very well [19–25], while the actual status in their industrial uses cannot be found easily in literature. Recent progress in bio-based epoxy technology development, with a focus on actual successful industrial uses, is introduced and summarized in this review article.

A bio-based material is a product whose raw materials are obtained entirely or partially from biomass. There are two methods for the bio-based content determination: the bio-based content and the bio-based carbon content. The bio-based content refers to the proportion of biomass in a product, taking into account the four main elements: carbon, hydrogen, oxygen, and nitrogen. The bio-based carbon content focuses solely on carbon and is generally expressed as a percentage of the carbon (organic carbon to total carbon) contained in the product. The bio-based carbon content is commonly used in epoxy technology to represent its bio content. The determination of the bio-based carbon content is carried out according to ASTM D6866 [26] and EN 16640 [27]. Bio-based carbon originating from biomass is radioactive, whereas carbon originating from fossils no longer is. The content of radioactive carbon and thus the bio-based carbon content can be determined and calculated using Accelerator Mass Spectrometry (AMS).

2. Bio-Based Epoxy Resins

There are mainly two types of bio-based epoxy resins available commercially or under commercial investigation: conventional epoxy resins and novel epoxy resins, depending on the epoxy resin building-block type used. Bio-based conventional epoxy resins are prepared by simply using the bio-based epichlorohydrin to replace conventional fossil-based epichlorohydrin in reaction with the same conventional building-block chemicals. Typical bio-based conventional epoxy resins are bio-based bisphenol A epoxy resins, bisphenol F epoxy resins, phenol novalac epoxy resins, and glycidyl amine-based epoxy resins. Since their preparation method and chemical structure are the same as those of conventional epoxy resins, bio-based conventional epoxy resins can be easily used in actual applications with no big concerns regarding its performance change. Bio-based novel epoxy resins are usually produced from the epoxidation of renewable precursors such as unsaturated vegetable oils, saccharides, tannins, cardanols, terpenes, rosins, and lignin [28–42]. Typical bio-based novel epoxy resins are vegetable-oil-based epoxy resins, cardanol-based epoxy resins, rosin-based epoxy resins, sorbitol- or isosorbide-based epoxy resins, lignin-based epoxy resins, and furan-based epoxy resins.

2.1. Bio-Based Epichlorohydrin

The majority of commercial epoxy resins are glycidyl-type epoxy resins synthesized by the reaction of active hydrogen in phenols, alcohols, amines, and acids with epichlorohydrin at certain well-controlled conditions. Most of these chemicals, including epichlorohydrin, originate from fossil fuels. Conventional epichlorohydrin is prepared by the reaction of propylene, originating from petroleum as well known, and chlorine. Recently, 100% bio-based epichlorohydrin has been developed and successfully commercialized. The bio-based epichlorohydrin is synthesized by the reaction of hydrochloric acid with renewable glycerol derived from vegetable oil as a byproduct in the preparation of biodiesel and oleochemical products. The preparation method of conventional fossil-based and new bio-based epichlorohydrin is shown in Figure 1 [43]. The bio-based epichlorohydrin has been used to replace conventional fossil-based epichlorohydrin in the commercial production of bio-based epoxy resins.

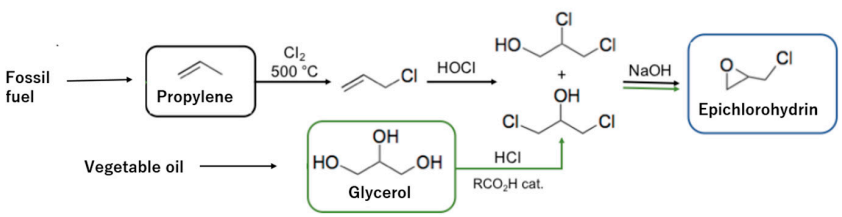


Figure 1. Preparation of bio-based and fossil-based epichlorohydrin.

2.2. Bio-Based Conventional Epoxy Resins

Bio-based conventional epoxy resins are typically glycidyl-type epoxy resins prepared using the bio-based epichlorohydrin to replace conventional fossil-based epichlorohydrin. Bio-based bisphenol A epoxy resin is prepared by the reaction of bisphenol A and bio-based epichlorohydrin, as shown in Figure 2 [44]. Th bio-based carbon content of standard-grade liquid bisphenol A epoxy resin is around 28%. The bio-based carbon content, EEW (epoxide equivalent weight), and viscosity of typical bio-based conventional epoxy resins are summarized in Table 1. These bio-based conventional epoxy resins have been commercially produced and supplied by several epoxy resin manufacturers [45–47]. Since their preparation method and chemical structure are the same as those of normal epoxy resin, bio-based conventional epoxy resins can be easily used in actual applications with no big concerns about its performance change. The preparation of 100% bio-based conventional epoxy resins, however, is a big challenge at the current stage as the resin building-block parts, such as bisphenol A, can only be produced from fossil sources.

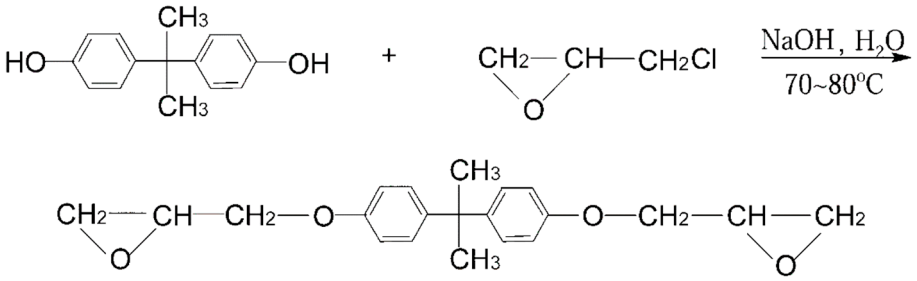


Figure 2. Synthesis of bisphenol A epoxy resin.

Table 1. Typical bio-based conventional epoxy resins.

Bio-Based Conventional Epoxy Resin	Bio-Based Carbon Content, %	EEW, g/Equ.	Viscosity, mPas@25 °C	Commercial Products
Liquid bis-A epoxy resin	28	190	12,000	Epotek YD 128G * Epilox A 1903G
Liquid bis-F epoxy resin	31	170	4000	Epotek YDF 170G Epilox F 1700G
Liquid novolac epoxy resin	30	172	13,000	Epotek YDF 173G
Phenol novolac epoxy resin	30	180	Semi-solid	Epotek YDF 173G
Tetra-functional epoxy resin	48	110	4500	Epotek YDM 441G
Diglycidyl ether of 1,6-hexanediol	50	150	20	Epilox P 1320G
Diglycidyl ether of 1,4-butanediol	60	135	17	Epilox P 1321G

* Epotek products supplied by Aditya Birla Chemicals; Epilox products supplied by Leuna-Harze GmbH.

2.3. Bio-Based Novel Epoxy Resins

Bio-based novel epoxy resins are usually produced from epoxidation of renewable precursors, such as unsaturated vegetable oils, saccharides, tannins, cardanols, terpenes, rosins, and lignin. Bio-based epoxy resins available commercially or under commercial investigation include vegetable-oil-based epoxy resins, cardanol-based epoxy resins, rosin-based epoxy resins, isosorbide-based epoxy resins, lignin-based epoxy resins, and furan-based epoxy resins [48,49]. Typical bio-based novel epoxy resins, their biomass source, and product available status are summarized in Table 2.

Table 2. Typical bio-based novel epoxy resins.

Bio-Based Novel Epoxy Resin	Biomass Source	Availability
Vegetable-oil-based epoxy resin	Vegetable oil	Aditya Birla Chemicals (Mumbai, India) Leuna-Harze GmbH (Leuna, Germany)
Cardanol-based epoxy resin	Cashew nut	Cardolite Corp. (Bristol, VA, USA)
Rosin-based epoxy resin	Pine, conifer	Ingevity Corp. (North Charleston, NC, USA)
Isosorbide-based epoxy resin	Starch	Huntsman Corp. (Woodlands, TX, USA) Nagase ChemteX, (Osaka, Japan)
Lignin-based epoxy resin	Wood	Aditya Birla Chemicals (Mumbai, India) Laboratory stage
Furan-based epoxy resins	Agricultural byproducts	Laboratory stage

2.3.1. Vegetable-Oil-Based Epoxy Resins

Vegetable-oil-based epoxy resins have been commercialized and supplied by several epoxy resin manufacturers [50–54]. By combination use of bio-based epichlorohydrin for product preparation, these vegetable-oil-based epoxy resins are real renewable materials with a maximum of 100% bio-based carbon content achievement. Typical commercialized vegetable-oil-based epoxy resins, their biomass source, EEW, and viscosity are summarized in Table 3.

Table 3. Typical vegetable-oil-based epoxy resins.

Bio-Based Epoxy Resin	Biomass Source	EEW, g/Equ.	Viscosity, mPas@25 °C
Glycidyl ether of C_{12}–C_{14} alcohol	Palm oil	300	10
Triglycidyl ether of castor oil	Castor oil	600	400
Triglycidyl ether of polyglycerol	Palm oil	170	1200
Glycidyl ether of dimeric acid	Tall oil	430	700

Glycidyl ether of C_{12}–C_{14} alcohol, derived from palm oil, has very low viscosity and is mainly used as a diluent. Its chemical structure is shown in Figure 3.

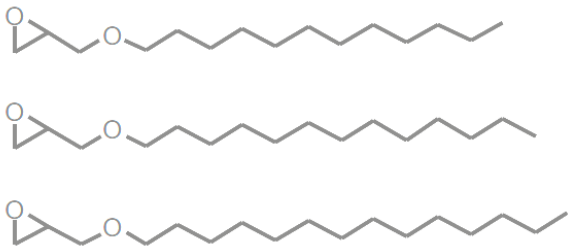


Figure 3. Chemical structure of glycidyl ether of C_{12}–C_{14} alcohol.

Triglycidyl ether of castor oil, derived from castor oil, is a tri-functional epoxy resin and can be used to improve flexibility and toughness. Its chemical structure is shown in Figure 4.

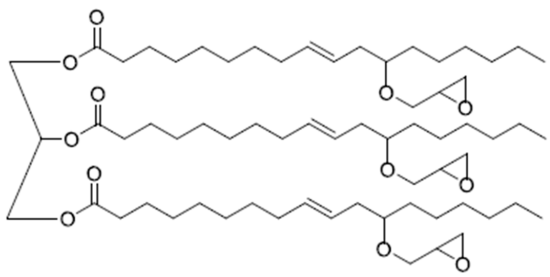


Figure 4. Chemical structure of triglycidyl ether of castor oil.

Triglycidyl ether of polyglycerol, derived also from palm oil, is a tri-functional epoxy resin and can be used to improve flexibility and toughness. Its chemical structure is shown in Figure 5.

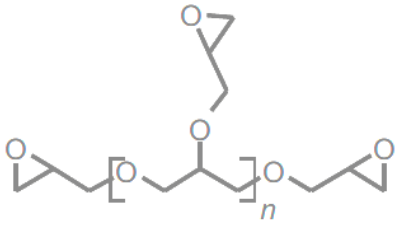


Figure 5. Chemical structure of triglycidyl ether of polyglycerol.

Glycidyl ether of dimeric acid, derived from tall oil, was commercialized a long time ago and has been used mainly to improve flexibility and toughness. Its chemical structure is shown in Figure 6.

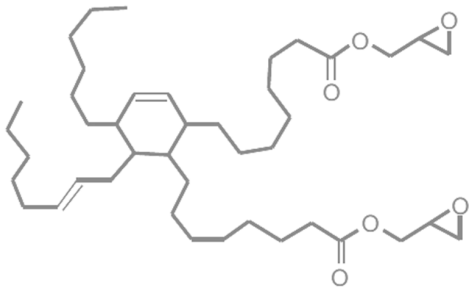
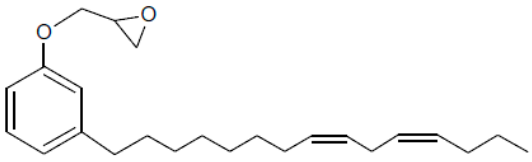


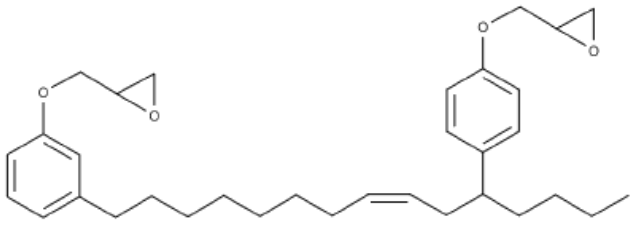
Figure 6. Chemical structure of glycidyl ether of dimeric acid.

2.3.2. Cardanol-Based Epoxy Resins

Cardanol is a phenolic liquid, derived from biomass cashew nuts. It is the main component of cashew nutshell liquid (CNSL) (https://en.wikipedia.org/wiki/Cashew_nutshell_liquid (accessed on 17 December 2024) Cardanol-based epoxy resins are produced by the reaction of cardanol with epichlorohydrin, and their bio-based carbon content can achieve 100%. Mono-functional, di-functional, and multi-functional cardanol-based epoxy resins have been commercialized by Cardolite Corporation [55]. Mono-functional cardanol-based epoxy resin has relatively low viscosity and can be used as a diluent. The chemical structure of cardanol-based epoxy resin is shown in Figure 7.



Mono-functional cardanol epoxy resin



Di-functional cardanol-based epoxy resin

Figure 7. Chemical structure of cardanol-based epoxy resin.

2.3.3. Rosin-Based Epoxy Resins

Rosin is a biomass solid material derived via bio-refinery from pine trees and other plants, such as conifers. Rosin contains resin acids, especially abietic aid. Rosin-based epoxy resin is a glycidyl-ester-type epoxy resin, prepared by the reaction of rosin acid with epichlorohydrin. Its bio-based carbon content can achieve nearly 100% with the use of bio-based epichlorohydrin. Rosin-based epoxy resins have been commercialized by Ingevity Corporation [56]. The chemical structure of tri-functional rosin-based epoxy resin is shown in Figure 8.

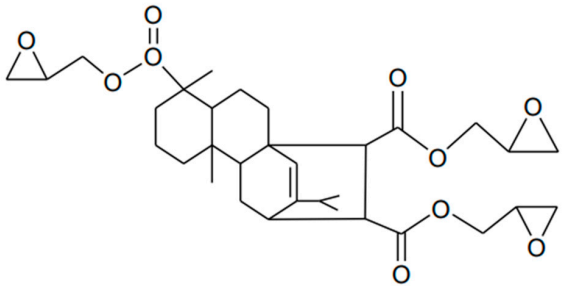


Figure 8. Chemical structure of tri-functional rosin-based epoxy resin.

2.3.4. Sorbitol, Isosorbide-Based Epoxy Resins

Sorbitol is a sugar alcohol, derived from the glucose in saccharides of biomass potato starch and corn starch. Isosorbide is prepared by the dehydration of sorbitol, as shown in Figure 9 [57].

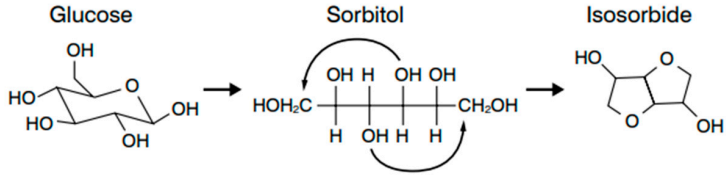


Figure 9. Preparation scheme of sorbitol and isosorbide from starch biomass.

Sorbitol-based epoxy resin can be prepared by the reaction of sorbitol with epichlorohydrin. Its bio-based carbon content can achieve nearly 100% by combination use of bio-based epichlorohydrin in the production. Sorbitol-based epoxy resin has been commercialized and supplied by several epoxy resin suppliers [58–61]. It is a multi-functional glycidyl-ether-type epoxy resin. The typical chemical structure of sorbitol-based epoxy resin is shown in Figure 10.

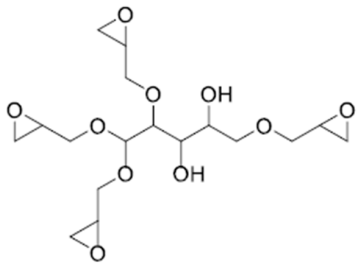


Figure 10. Chemical structure of sorbitol-based epoxy resin.

Isosorbide-based epoxy resin can be prepared by the reaction of isosorbide with epichlorohydrin. Its bio-based carbon content can also achieve nearly 100% by combination use of bio-based epichlorohydrin in the production. Isosorbide-based epoxy resin has been

commercialized and supplied by several epoxy resin suppliers [62,63]. It is a di-functional glycidyl-ether-type epoxy resin and can potentially be used as a base resin because of a relatively strong building-block structure. Typical chemical structure of isosorbide-based epoxy resin is shown in Figure 11.

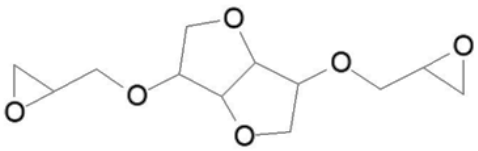


Figure 11. Chemical structure of isosorbide-based epoxy resin.

2.3.5. Lignin-Based Epoxy Resins

Lignin is one of the three main components of wood, together with cellulose and hemicellulose. Chemically, lignin is a polymer comprising cross-linked phenolic precursors. It is the second-most abundant biopolymer only behind cellulose and is the most abundant bio-aromatic compound [64]. Lots of studies have been carried out to prepare lignin-based epoxy resin and investigate its properties and performance, aiming to establish a useful level of bio-replacement on bisphenol A in epoxy resin technology [65–71]. Commercial preparation of bio-based lignin-based epoxy resins, however, is still at an early investigation stage, due to the complex structure of lignin material. Figure 12 illustrates one preparation route of lignin-based epoxy resin, synthesized by the reaction of lignin's phenolic hydroxy group with epichlorohydrin [72].

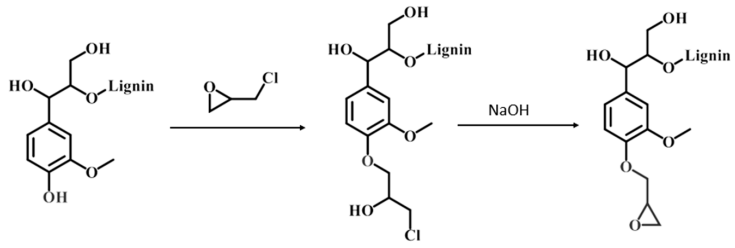


Figure 12. Preparation scheme of lignin-based epoxy resin.

2.3.6. Furan-Based Epoxy Resins

As illustrated in Figure 13, furfural compounds can be prepared from cellulose, the most abundant natural biopolymer derived from various plants. Furan-based epoxy resin can be prepared by the reaction of alcohol or acid furan compounds with epichlorohydrin. The maximum bio-based carbon content can achieve 100% with combination use of bio-based epichlorohydrin. Great efforts have been paid to develop and investigate furan-based epoxy resins, as they are potentially expected to replace fossil-derived bisphenol A due to its aromatic chrematistics [73–77]. Figure 14 shows the chemical structure of two di-functional furan-based epoxy resins.

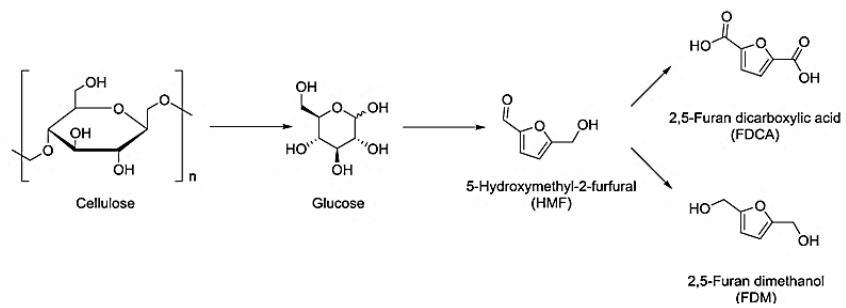
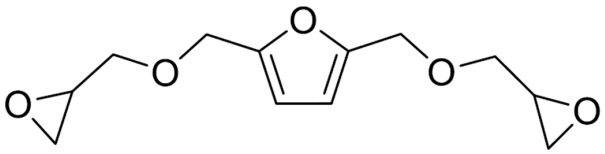
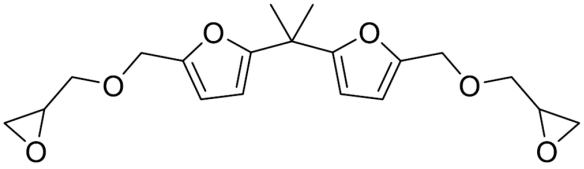


Figure 13. Preparation scheme of acid and alcohol furan.



2,5-bis[(2-oxiranylmethoxy) methyl]-furan (BOF)



bis-furan diepoxide (bBOF)

Figure 14. Chemical structure of furan-based epoxy resins.

3. Bio-Based Epoxy Curing Agents and Additives

Typical bio-based curing agents available commercially or under commercial investigation are bio-based polyamines, bio-based polyamides, bio-based amidoamines, cardanol-based phenalkamine-type curing agents [78–89]. IPDA (isophoronediamine), furan-based amine, and isosorbide-based amine curing agents are typical bio-based polyamine-type curing agents. Cardanol is a typical bio-based reactive additive.

3.1. Bio-Based Polyamine-Type Curing Agents

The majority of commercial polyamine-type curing agents are prepared from chemicals that originated from fossil fuels. Only a few of them are prepared from biomass material. As summarized in Table 4, IPDA, furan-based amine curing agents, and isosorbide-based amine curing agents are typical bio-based polyamine curing agents available commercially or under commercial investigation [90–92].

Table 4. Typical bio-based polyamine-type curing agents.

Bio-Based Polyamine	Biomass Source	Availability
Isophoronediamine (IPDA)	Starch, glucose	Evonik Industries (Essen, Germany)
Furan-based polyamine curing agent	Agricultural byproducts	Laboratory stage
Isosorbide-based polyamine curing agent	Starch	Laboratory stage

IPDA is one typical cycloaliphatic diamine-type curing agent offering low viscosity, good color stability, and high Tg suitable for high-temperature high-humidity-resistant applications. IPDA is prepared from isophorone, which is commercially obtained exclusively via the self-condensation of acetone. With the use of bio-based acetone derived from the fermentation of biomass material such as starch and glucose, up to 90% bio-based carbon content of IPDA has been commercialized. The chemical structure of IPDA is shown in Figure 15.

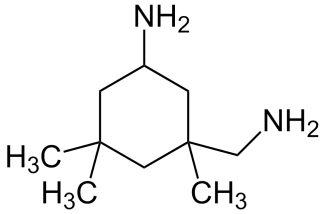


Figure 15. Chemical structure of IPDA (isophoronediamine).

Furan-based amine curing agents can be prepared by the amination reaction of alcohol furan compounds with ammonia. The maximum bio-based carbon content can

achieve nearly 100%. Figure 16 shows the chemical structure of [5-(aminomethyl)furan-2-yl]methanamine, a furan-based diamine.

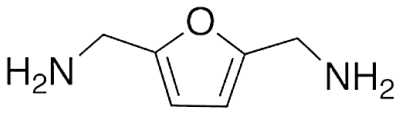


Figure 16. Chemical structure of [5-(aminomethyl)furan-2-yl]methanamine.

Isosorbide-based polyamine-type curing agents can be prepared by the amination reaction of isosorbide with ammonia. Its bio-based carbon content can also achieve nearly 100%. Figure 17 shows the chemical structure of diaminoisosorbide, an isosorbide-based diamine.

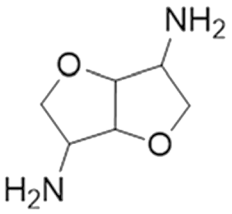


Figure 17. Chemical structure of diaminoisosorbide.

3.2. Bio-Based Polyamide-Type Curing Agents

Polyamide-type curing agents are prepared by the reaction of dimeric acid, derived from biomass tall oil, with excessive polyamine, as illustrated in Figure 18 [93]. Polyamide has higher viscosity, lower toxicity, and longer pot-life than polyamines. Polyamide curing agents show good adhesion, good flexibility, and toughness. Up to 70% bio-based carbon content can be achieved depending on the addition ratio of dimeric acid applied in its synthesis [94,95].

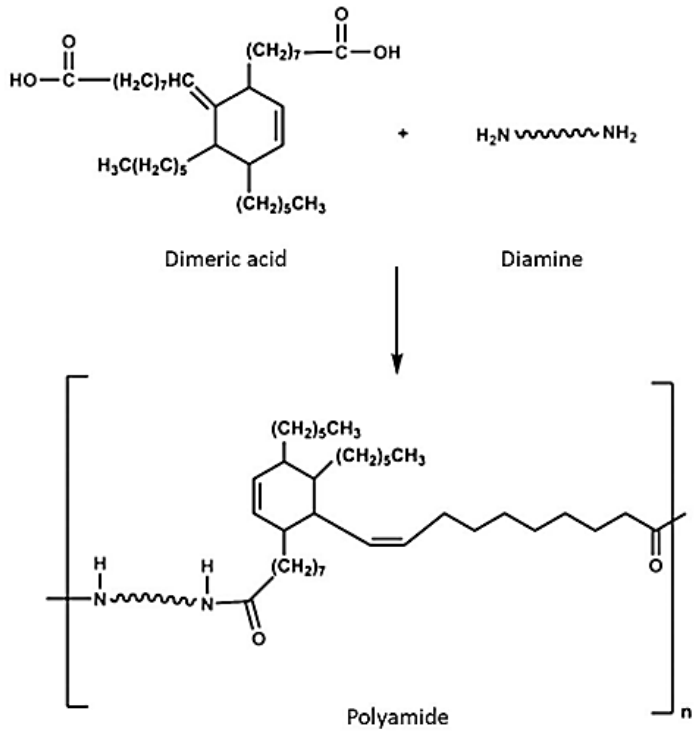


Figure 18. Preparation of polyamide-type curing agent.

3.3. Bio-Based Amidoamine-Type Curing Agents

Amidoamine-type curing agents are prepared by the reaction of monobasic fatty carboxylic acid, derived from biomass vegetable oil, with excessive polyamine. Amidoamine has

viscosity between polyamide and polyamine. Amidoamine-type curing agents show good handling property, good adhesion, and good chemical resistance. Up to 60% bio-based carbon content can be achieved depending on the addition ratio of fatty acid applied in its synthesis.

3.4. Bio-Based Phenalkamine-Type Curing Agents

Bio-based phenalkamine is a Mannich reaction product of polyamine with biomass-derived cardanol, and aldehyde, as illustrated in Figure 19 [96,97]. Bio-based phenalkamine-type curing agents offer fast curability and good chemical resistance. Up to nearly 90% bio-based carbon content can be achieved depending on the addition ratio of cardanol applied in its synthesis.

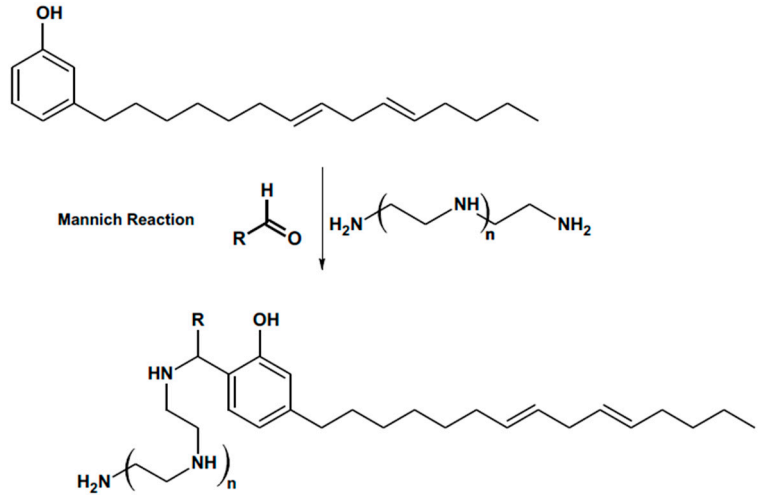


Figure 19. Preparation of bio-based phenalkamine.

3.5. Bio-Based Phenols

Cardanol is a 100% bio-based phenol, prepared by decarboxylated and distilled from cashew nutshell liquid, a renewable biomass as already described previously. Cardanol can be used as a reactive additive in epoxy formulation as one alternative to replace nonylphenol [98], a chemical with a high level of health and safety concerns. The hemical structure of cardanol is shown in Figure 20.

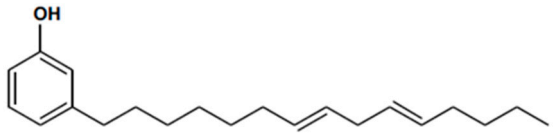


Figure 20. Chemical structure of cardanol.

4. Bio-Based Formulated Epoxy Materials

Bio-based epoxy materials are formulated by using, either partially or entirely, bio-based epoxy resins, bio-based curing agents, or bio-based additives to replace conventional fossil-derived raw materials [99–109]. Commercial bio-based formulated epoxy products have been already developed and supplied for bonding, coating, casting, composite, and laminating applications by several manufacturers. Table 5 lists a few bio-based formulated epoxy products available commercially, their product type, bio-based carbon content range, and target applications [110–114].

Table 5. Bio-based formulated epoxy products.

Bio-Based Formulated Epoxy Product	Product Type	Bio-Based Carbon Content	Typical Applications
Cardolite FormuLITE series	2-component	30–50%	Composites
Sicomin GreenPoxy series	2-component	28–56%	Laminating, bonding, coating, etc.
Polytek CPD series	2-component	20–35%	Bonding, casting, laminating
SikaBiresin CR75 series	2-component	38%	Composites

5. Conclusions

Significant progress in sustainable bio-based epoxy technology development has been achieved in recent years. Many bio-based epoxy resins, bio-based curing agents and additives, and bio-based formulated epoxy materials, as well as their performance, have been investigated. More importantly, certain types of bio-based epoxy resins, bio-based curing agents, and bio-based formulated epoxy products have been commercialized, and their use in actual applications has been realized. Continuous efforts in bio-based epoxy technology development have become increasingly important to realize a real sustainable and environmentally friendly society.

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