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# Cardanol and Its Derivatives: Innovations in Waterborne Coating Technologies

Muhammad D. Saad [1] , Tushar Singh [1] , Kunal D. Wazarkar [1]

1. Functional Resins, Asian Paints Research and Technology Centre, Navi Mumbai, IND

**Corresponding author:** Kunal D. Wazarkar, kunal.wazarkar@asianpaints.com

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## Abstract

Utilization of bio-renewable materials to replace petroleum-based building blocks for advanced coatings has been a major focus area for researchers in the development of eco-friendly and sustainable products. Additionally, the growing restrictions on volatile organic compounds and hazardous air pollutants have forced industries to opt for waterborne systems. Therefore, considerable focus has been directed toward bio-based waterborne systems. Over the years, bio-based waterborne systems have evolved and been established with superior advantages such as fast curing, chemical resistance, excellent adhesion, and enhanced performance. Various biomaterials have been explored in the past for waterborne systems such as vegetable oils, lignin, tannin, cellulose, sugar alcohols, cashew nutshell liquid, and others. Among them, cardanol, an agricultural non-edible byproduct of cashew nut industry, has emerged as a promising candidate for waterborne systems due to its unique structural characteristics, abundant availability, low cost, inedibility, and biodegradability. Owing to its phenolic nature and presence of unsaturated alkyl chain at meta position, cardanol provides excellent balance of hardness and flexibility along with superior corrosion resistance. This review highlights recent advancements in cardanol-based waterborne systems as well as its application in surface coatings. In addition, many research groups have modified cardanol and its derivatives through variety of chemistries such as polyurethane dispersions, epoxy curing agents, and acrylic latexes for waterborne systems, which is also summarized in this review. Studies revealed that these coatings exhibited excellent performance properties offering tranquillity to the concern regarding the inferiority of cardanol-based waterborne coatings.

**Categories:** Advanced Materials, Environmental Engineering and Sustainability, Polymer Science and Engineering
**Keywords:** cardanol, waterborne, volatile organic compounds, biobased, environment friendly

## Introduction And Background

Approximately 8.1 billion people live in the world and continue to increase at a rate of 0.8% every year. The increasing population leads to an increase in the number of consumers, and every sector seems to have a huge surge in demand, which restricts us to limited non-renewable resource. The increased demand is directly related to the depletion of non-renewable resources and a rise in cost [1-3].

Our dependency on limited non-renewable resources drives the concern and consciousness regarding the environment and available resources. To overcome these concerns, big forums like G20 have their prior initiatives toward sustainability, and worldwide changes are happening to achieve sustainability, forcing every single firm and consumer to shift toward sustainable products and, hence, bio-based materials. Considering the coating industry, bio-based coating market is valued at US$ 13.56 billion in 2024 and is forecasted to expand at a Cumulative Annual Growth Rate of 8.6% to reach US$ 30.83 billion by 2034, reflecting the transformation of the world to green products and less dependency on fossil fuels or non-renewable resources [4,5].

Incorporating or using bio-based raw materials in place of existing petroleum ones is a challenge to replace them in terms of cost and performance properties. However, overcoming these challenges and adapting bio-based raw materials provide several benefits, including sustainability, reduced Volatile Organic Compounds (VOC) emissions and Hazardous Air Pollutants, and a lower carbon footprint. The coating industry, which has a huge dependency on petroleum-based products, is persistently exploring new avenues for the replacement of these raw materials by bio-based materials to step forward to achieve breakthrough of sustainable products with superior properties and cost benefits. Successful incorporation of bio-based raw materials will bring down the dependency on fossil fuels, implying minimum environmental impact hence diminishing the carbon footprint. Renewability is the biggest breakthrough in this, as it provides a never-ending supply through the agriculture and plants, and it will indirectly help to grow agriculture in rural areas. Products based on bio-based materials will provide inherent safety in terms of the toxicity of materials as well as reducing the VOC content [6,7].

Several sources are responsible for VOC generation and the largest being petrochemical industries and numerous chemical industries which accounts for 40% of total VOC [7]. These VOCs are organic gases, mostly composing of aromatic hydrocarbon gases like ethylbenzene, xylene, toluene, and others. Most

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common resources that are accountable for VOC generation are paint, varnish, adhesives, cleaning products, cosmetics, disinfectants, biocides, and so on. Studies on risk related to human health reveal that acute exposure of VOC can cause throat, nose, and eye irritation, headache, loss of coordination while chronic exposure could be responsible for very serious diseases like cancer. Contamination of VOC in water is the other reason for health hazard. Therefore, reduction of VOC content is highly important owing to its harmful effects on human health, living organism, and environment. Pollution control board and other regulatory authorities have defined various strategies and acts to control individual industries to minimize the VOC [8]. Coating industries have taken the great challenge of reducing VOC by cutting down the concentration of petroleum-based raw materials by bio-based raw materials and water incorporation as a solvent for the products, which are making huge impact toward sustainability and green products [9].

Polymer research field has accomplished the remarkable achievement of synthesizing waterborne polymers based on bio-based raw materials, such as vegetable oils [10], isosorbide [11], lignin [12], furfural [13], amylose [14], rosin [15], itaconic acid [16], cellulose [17], succinic acid [18], sorbitol [19], and glycerine [20]. Among them, cardanol, a non-edible byproduct of cashew nut industry, has gained tremendous attention recently for the preparation of waterborne polymers/oligomers and cross-linkers in surface coatings. In this regard, various waterborne chemistries based on cardanol are explored and summarized in this review.

## Cardanol extraction and composition

Cashew tree, scientifically named as “Anacardium occidentale” is originated in South America; however, it is abundantly available in Asia and Africa [21]. The largest cashew-producing country, Brazil, caters to 60% of global demand, followed by India and Vietnam [22]. Cashew nuts, which are used as an ingredient in food, have several health benefits. Along with this, cashew nuts generate very important byproduct cashew nutshell liquid (CNSL) that is used as bio-based resource to produce environment-friendly products. Extracted CNSL has different uses in different fields like coatings, pharmaceuticals, and other chemical industries. The stem of cashew tree is used for the preparation of juices and cashew apple is used to produce ethanol, jam, juice, syrup, and others. For the coating industry, CNSL has a very important role to play. Natural CNSL contains anacardic acid (74-77%) as a main component followed by cardol (15-20%), cardanol (1-9%), and 2-methyl cardol (1-3%) (as shown in Figure *1*, see ref. [5]). Amongst all, cardanol is the most popular and attractive molecule owing to its peculiar structure composing a phenolic hydroxy moiety and a meta positioned hydrocarbon chain (mixed saturated and unsaturated C15 chain with saturated 5-8%, monoene 48-49%, diene 16-17%, triene 29-30%) that provides excellent balance of flexibility, hydrophobicity, and hardness. In addition, the available functionalities can be modified with a variety of chemistries like etherification, esterification, nitration, hydrogenation, epoxidation, bromination, ethoxylation, allylation, hydrosilylation, phenolation, metathesis, and others [23,24] using suitable catalysts and reactants. The current global cardanol market is around 29-32 million dollars and has projected the value of 58-78 million dollars by 2030. Low odor, non-toxicity, low vapor pressure, high boiling point, easy availability at cheaper prices, and sustainability make it a very strong replacement for petroleum-based compounds. Owing to its significance, natural CNSL is converted into technical grade, which is reddish brown in appearance and contains cardanol as the main component (as shown in Table *1*), the and rest includes cardol, 2-methyl cardol, anacardic acid, and polymeric material depending upon the extraction process [25-28].

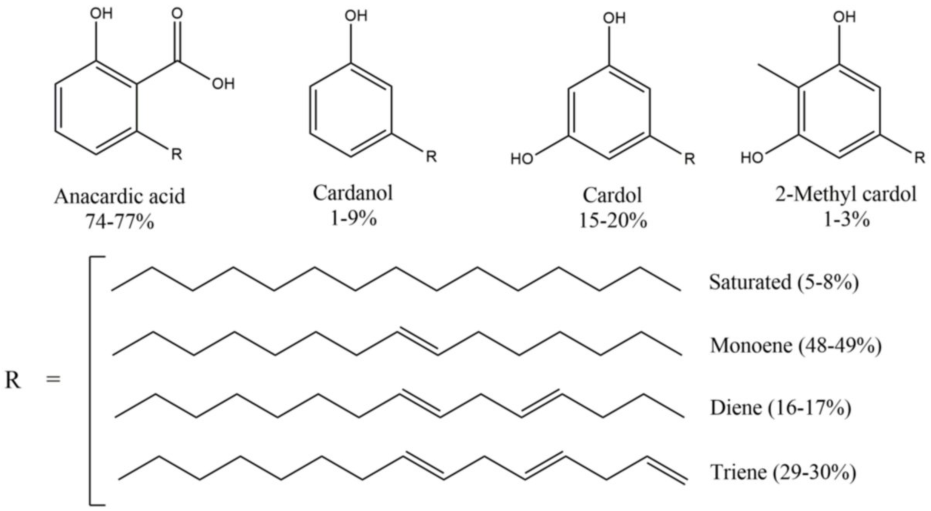


FIGURE 1: Derivatives of cashew nutshell liquid (CNSL)

| Type | % Component breakup | | | |
|---|---|---|---|---|
| | Anacardic Acid | Cardanol | Cardol | 2-methyl cardol |
| Natural CNSL | 60-65 | 10-15 | 20 | Traces |
| Technical CNSL | -- | 60-65 | 15-20 | Traces, 10% polymeric materials |

TABLE 1: Components in natural and technical CNSL

CNSL, cashew nutshell liquid

Commercially CNSL is extracted by various techniques including mechanical, thermal, roasting, steam processing, solvent extraction, quick roasting, and cold press, which involve extraction by pressing, extrusion, and so on. Cold pressed or solvent extracted CNSL generally results in natural CNSL and roasting of cashew nutshells yields technical CNSL. During extraction process, CNSL is heated up to 170-175℃ at reduced pressure of 30-35 mm of Hg, which ensures the decarboxylation of anacardic acid into cardanol. Furthermore, distillation applied to treated CNSL at 200-240℃ at 2-2.5 mm hg of pressure twice in same condition of temperature and pressure, gives cardanol at 205-210℃ and cardol at 230-235℃ as distillates. This results in two grades of cardanol: single-distilled and double-distilled, distinguished by the composition of cardanol [29,30].

| Parameter | Single distilled | Double distilled |
|---|---|---|
| Composition | Cardanol (90-94%) cardol, 2-methyl cardol, anacardic acid (6-10%) | Cardanol (94-98%) cardol, 2-methyl cardol, anacardic acid (2-6%) |
| Color (Gardner scale) | 3-4 (freshly distilled) | 2-3 (freshly distilled) |
| Moisture | Negligible | Negligible |
| Specific gravity | 0.92-0.93 | 0.92-0.925 |
| Iodine value (g of $I_2$/100 g) | Minimum 275 (Wij's method) | Minimum 275 (Wij's method) |
| Ash content | Negligible | Negligible |
| Brookfield viscosity@ 30℃ (cps) | 50-55 | 45-50 |

**TABLE 2: Composition and characteristics of single- and double-distilled cardanol**

Composition (Table 2) shows that double distillation increases the cardanol percentage and hence reduces the other content which causes the low color value of cardanol. Freshly distilled cardanol has the appearance of pale-yellow liquid and changes to reddish brown rapidly due to the presence of cardol. Providing less amount of cardol in comparison with single-distilled to double-distilled cardanol provides color value of 7-8 and 6-8, respectively, after 30 days of storage. Sometimes, cardanol is also reacted with bases of alkaline and alkali earth metal or aldehyde and amines or boric acid to obtain color-stable cardanol. These compounds react with cardol, which is responsible for color change, thus providing color stability to the cardanol [23].

From polymer and coating perspective, cardanol can be modified in several ways to make it suitable for the use in preparation of different types of polymers. Inherently, it has the phenolic hydroxyl available to replace phenolic compound and can be used to synthesize phenolic resin, but the ability to undergo variety of chemical reactions to generate different functionalities opens a window of opportunities to incorporate cardanol in different polymers [31,32]. For example, epoxidized/hydrolysed cardanol can be used in alkyd, polyester, acrylic resins through ring opening/esterification with acid functional groups and polyurethane by reacting hydroxy functional cardanol with isocyanates. Apart from this, cardanol derived phenalkamines and cationic, nonionic, and anionic surfactants are other possibilities. Cardanol-derived polymers provide sustainability as well as numerous benefits like excellent electrical resistance, water, alkali and acid resistance, flexibility, organic solvent solubility, excellent corrosion resistance, and antimicrobial properties, which attracted every industry to research and incorporate cardanol into their product as a versatile green raw material. Figure 2 represents structure property relationship as well as possible modification of available functionalities in cardanol [33,34].

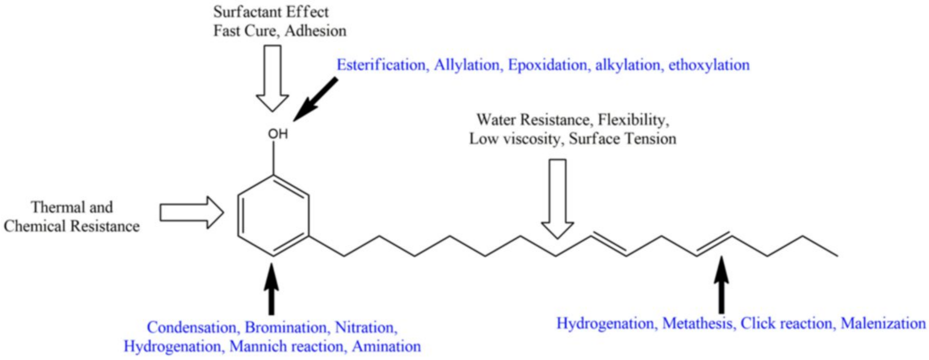


**FIGURE 2: Cardanol: possible chemistries and structure-property relationship**

# Review

## Water-based systems based on Cardanol and its derivatives

Cardanol and its modification through variety of chemistries are well known, and being water insoluble, these products mainly find applications in solvent borne coatings. There are a number of review articles published in the past, and new ones are published every year, focusing on the utilization of cardanol in solvent borne applications. However, there is not much literature on exploration of cardanol and its derivatives in water-based systems. Therefore, looking to the advancements and surge in the research works of this field, an attempt has been made to provide insights of the current trends in this paper. Present article includes various strategies for the modification of cardanol and its derivatives for its suitability in water-based systems, namely polyurethane dispersions (PUDs), epoxy curing agents as well as acrylic emulsions. The article highlights the effect on mechanical, chemical, thermal, and anticorrosive behavior of the coatings and establishes structure-property relationships of the same.

### *Polyurethane Dispersions*

Polyurethane is an important class of polymers with superior properties and a variety of applications in foams, elastomers, sealants, adhesives, and paints. When polyurethanes are finely dispersed in water as a continuous medium, a binary colloidal system known as PUDs is obtained. In the last few decades, waterborne PUDs have successfully superseded conventional solvent borne dispersions [35]. Today, waterborne PUD coatings are giving tough competition to their solvent borne counterparts with even superior properties. Not only because they eliminate the solvent emission but also because they are suitable for a wide range of applications as compared to solvent borne polyurethane coatings [36]. Most solvent borne PUD coatings are thermoplastics, and lack in cross-linking and higher molecular weights, whereas waterborne PUDs attain higher molecular weights even at low viscosities. Dispersed phase of PUD can be soft, ductile, or hard, and the particle size lies in the range of 100-600 nm [37,38]. They are broadly classified based on the charge of the dispersing moiety i.e., cationic and anionic PUD. Dimethylol propionic acid (DMPA) and dimethylol butanoic acid are widely used reactive dispersing agents for this purpose [39].

Recently, various cardanol-based polyols were developed by researchers and incorporated in PUDs. For example, Mishra et al. synthesized epoxy cardanol by reacting cardanol with epichlorohydrin in basic medium. Further, epoxidized cardanol (in excess) was reacted with 1 mole of itaconic acid in the presence of triphenyl phosphine (TPP) as catalyst to yield cardanol-based polyol at temperature of 150-160°C. A prepolymer consisted of 30% cardanol as a bio-based material was formed when isophorone diisocyanate (IPDI) was slowly added to the mixture of DMPA and cardanol-based polyol in the presence of a dibutyltin dilaurate (DBTDL) catalyst. Hydroxyethyl Methacrylate was used for end capping of this prepolymer until free isocyanate content reached nearly 0%, followed by neutralization with triethylamine and finally dispersed in deionized water. To evaluate the effect of UV/oxidative curing on coating properties, three coatings were formulated. For oxidative curing, PUDs were mixed with metal driers, a photo initiator, and amine synergists were used for UV-cured system and combination of above was utilized for dual-cured system. DMA studies revealed that the highest cross-link density was obtained with dual-cure system followed by UV-cured system, and the least for oxidative-cured system and similar trend was observed in the case of glass transition temperature of films. The behavior could be attributed to dual cross-linking of unsaturation present in substituted alkyl chain of cardanol that enhanced overall cross-link density of films. Overall, dual UV/oxidative cured PUD coatings had higher cross-linking density, which resulted in excellent thermal, mechanical properties, and superior resistance to chemical, water, and organic solvents as shown in Table *3* [40].

| Properties | Curing process | | |
|---|---|---|---|
| | Oxidative cured | UV cured | Dual cured |
| Tensile strength (MPa) | 2.6 | 4.6 | 5.7 |
| Elongation at break (%) | 116 | 42.7 | 50.3 |
| Gel fraction (wt%) | 72.4 | 85.1 | 93.3 |
| Water contact angle ($^0$) | 81.6 | 86.4 | 89.7 |
| Water absorption (wt%) | 7.4 | 5.3 | 3.4 |
| Adhesion (cross-hatch) | 5B | 5B | 5B |
| Flexibility (1/8’’ mandrel) | Passes | Passes | Minor crack observed |
| Pencil hardness | 2H | 4H | 5H |
| Scratch hardness (kg) | 1.8 | 2.4 | >3.0 |
| Impact strength (cm) | 80 | 80 | 70 |
| Alkali resistance | Film softens | Whitening | Mild whitening |
| Salt water resistance | Unaffected | Unaffected | Unaffected |
| Acid resistance | Slight color change | Unaffected | Unaffected |
| Solvent resistance (MEK double rub) | <100 | >200 | >200 |

**TABLE 3: Performance and chemical properties of UV-PUD films with different curing process**

UV-PUD, ultraviolet-cured polyurethane dispersion; MEK, methyl ethyl ketone

Another study carried out by Patel et al. allowed cardanol to directly react with diglycidyl ether of bisphenol A using ethyl-triphenylphosphonium bromide as a catalyst to form cardanol epoxy condensation polyol (CE). This CE was further characterized by Fourier Transform Infrared (FTIR) spectroscopy and Gel Permeation Chromatography for structural and molecular weight confirmation, respectively. Reaction of CE, DMPA with slow addition of IPDI in the presence of DBTDL catalyst resulted in NCO-terminated prepolymer, which was neutralized and dispersed in deionized water using high-speed disperser. Experimental studies showed that cardanol-modified polyurethane dispersions were of low particle size, attained higher molecular weight at low viscosities, and contained high renewable content. In addition, PUD films cured via oxidative drying exhibited higher cross-linking density and corrosion resistance [41]. Figure *3* describes various polyols synthesized from cardanol.

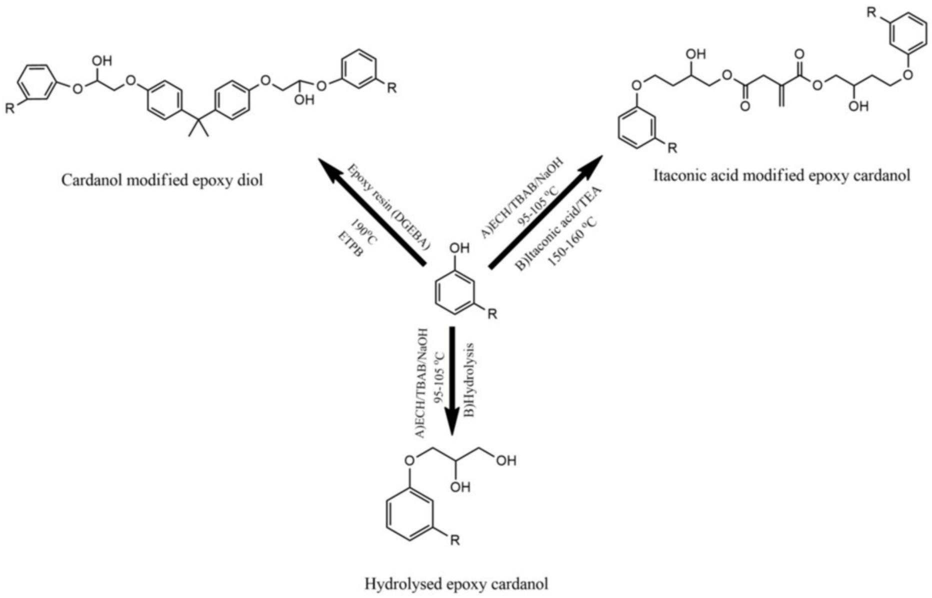


**FIGURE 3: Cardanol-based polyols for polyurethane dispersions**

Kattimuttathu et al. carried out epoxidation of cardanol with epichlorohydrin in alkaline medium followed by the hydrolysis of epoxy ring under acidic conditions to obtain cardanol diol. The synthesized diol was further utilized for the preparation of PUDs. Three dispersions were designed by varying the type of chain extenders and their concentrations, such as butanediol 9 wt%, cardanol diol at 13 wt%, and 26 wt% loading. It was observed that butanediol as chain extender, reduced the average particle size and polydispersity index as compared to cardanol diol in PUDs. Cardanol diol resulted in broad particle size distribution; however, higher concentration of the same reduced the particle size. The lowest polydispersity index was observed with cardanol diol at 13 wt%, which also showed 35% bigger particle size. Study demonstrated that ease of film formation in PUDs was better with cardanol diol as compared to butane diol. Films prepared with cardanol diol were observed to be soft and flexible while brittle films were obtained with butane diol. This could be related to the higher molecular weight of cardanol and dangling alkyl chain that resulted in low crystallinity as compared to butane diol. With 26 wt% of cardanol diol as chain extender, no breakage was observed even at elongation of 1600%. In addition, contact angle increased with increase in cardanol diol concentration and the lowest contact angle was observed with butane diol. Furthermore, thermal decomposition profiles showed that addition of cardanol diol increased thermal stability of the films marginally [42].

In another work, Mestry et al. synthesized novel cardanol diol dispersant that acted as a polyol as well as dispersant for PUDs. Authors first sulfonated the cardanol in the presence of oleum followed by epoxidation and subsequently hydrolyzed to obtain sulfonated cardanol diol (as shown in Figure *4*). The synthesized multifunctional cardanol dispersant was used as a replacement to traditional DMPA in PUD formulation. Two PUDs based on DMPA (waterborne polyurethane (WPU)-DMPA) and sulfonated cardanol diol (WPU-ethyl sulfonated (ESC)) were designed and evaluated for performance properties. It was observed that WPU-ESC showed lower particle size than the WPU-DMPA, due to higher dispersion capability of sulfonyl groups in the water. The particle size of WPU-ESC was 317.8 nm while WPU-DMPA had 375.9 nm. Incorporation of sulfonated cardanol as dispersing agent in water-based polyurethane increased the Tg of film, as sulfonated group along with phenyl group hindered the chain mobility of polymer backbone restricting the segmental motion of polymer chains to a great extent. Further, presence of sulfur also improved thermal stability of WPU-ESC as compared to WPU-DMPA as seen by Thermogravimetric Analysis. The char yield was also higher for WPU-ESC (15.7%) as against WPU-DMPA (2.8%), indicating significant improvement in thermal stability. In terms of mechanical properties, WPU-ESC exhibited low hardness, flexibility, and impact resistance as compared to WPU-DMPA. This could be attributed to the presence of aromatic ring and sulfur atom in sulfonated diol that resulted in brittle films. Incorporation of bulky group in WPU-ESC introduced voids that reduced the effective cross-linking density and water resistance. Similar trend was observed in gel content as WPU-DMPA displayed 7.8% higher gel content than WPU-ESC counterpart, which again proved that cross-linking density remains lower in the case of WPU-ESC [43].

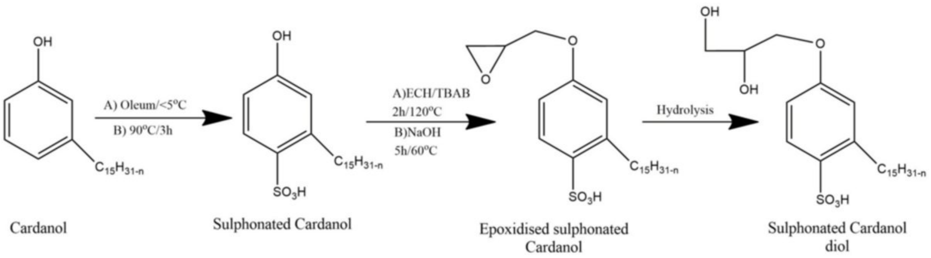


**FIGURE 4: Synthesis of waterborne polyol dispersing agent from cardanol**

In a similar line of work, Xia et al. employed sulfonated cardanol modified with varying molecular weights of polyethylene glycol (PEG) units. Further, the cardanol intermediate (SCP) was mixed in various ratios with poly(butylene adipate)glycol-3000 (PBA) and reacted with IPDI in the presence of DBTL catalyst followed by chain extension with ethylene diamine to yield PUDs. It was observed that with an increase in molecular weight of PEG units, a stable dispersion having particle size close to 300 nm was achieved. This behavior could be related to the improved hydrophilicity of polymer backbone due to higher molecular weight of PEG units that enhanced emulsification resulting in smaller size particles. Hence, PUDs with PEG-600 showed particle size of 11.9 μ as against PEG-2000 with 294 nm particle size. Similarly, reducing the PBA/SCP ratio also reduced the particle size that improved long-term stability of dispersions. In contrast, tensile strength and % elongation reduced with increase in molecular weight of PEG units, owing to the reduction in hard segments in PU backbone. To further improve tensile strength and other properties, authors exposed these films to UV radiation for 5 s to 15 s and as expected, 53% increase in tensile strength was observed for PU films based on PEG-1500 after 10 s of UV exposure. Authors concluded that UV cross-linking of PU films improved thermal stability, water resistance, and water vapor barrier properties of the coatings [44].

In another work, Balgude et al. reacted cardanol-based diglycidyl ether (NC-514) with various multifunctional acids including citric acid, tartaric acid, and adipic acid, individually as well as in combinations to incorporate hydrophilic moieties into the polymer backbone. Multifunctional acids opened the oxirane ring of NC-514 in the presence of TPP to yield hydrophilically modified polyols (as shown in Figure *5*). The designed polyols had hydroxyl values in the range of 97 mg KOH/g (100% adipic acid) to 275 mg KOH/g (100% tartaric acid) and acid values in the range of 89 mg KOH/g (100% tartaric acid) and 168 mg KOH/g (100% citric acid). These hydrophilically modified polyols were partially diluted in propylene glycol monomethyl ether and neutralized with triethyl amine followed by the addition of water under stirring to obtain polyol dispersions. The developed polyol dispersions were then cured with water-based polyisocyanate cross-linkers based on IPDI and hexamethylene diisocyanate (HDI), and evaluated for coating properties such as mechanical, solvent and chemical resistance, thermal, anti-corrosive, and others.

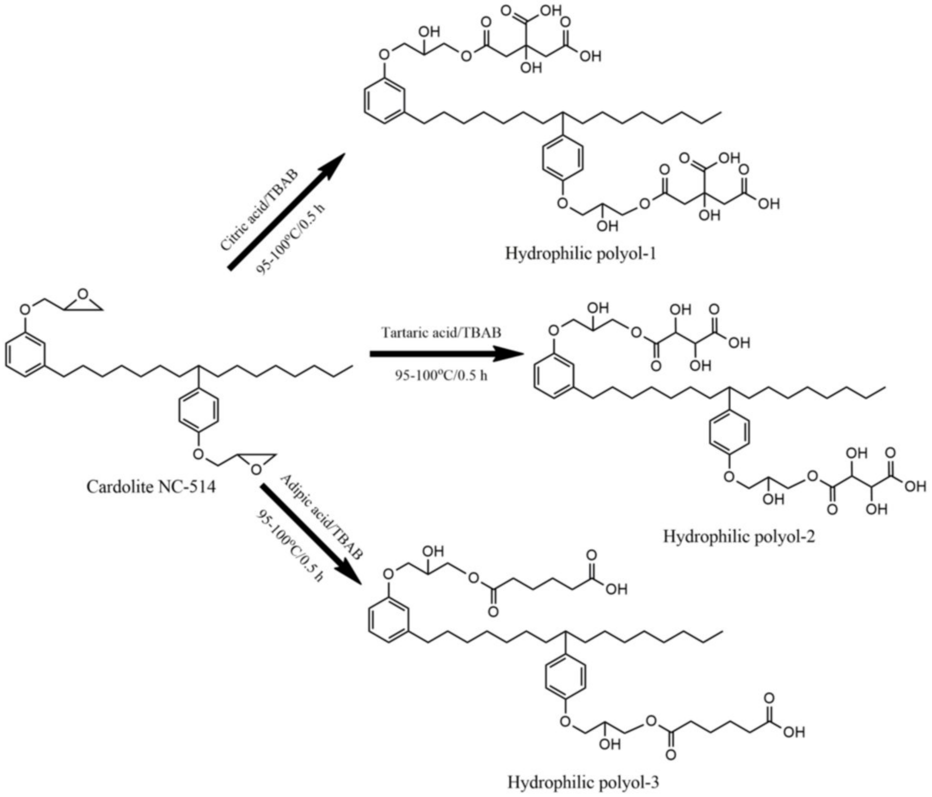


FIGURE 5: Cardanol-based hydrophilically modified polyols

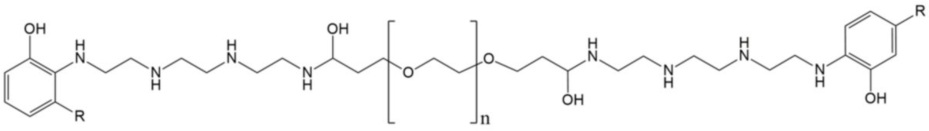
It was observed that the presence of cycloaliphatic ring in IPDI was responsible for better gloss than HDI-based coatings. IPDI-based PUDs having higher cross-linking density and high hydroxyl content led to better rigidity, pencil, and scratch hardness as compared to HDI-based counterpart. Long aliphatic chain in cardanol imparted chemical stability to PUDs thus improving chemical resistance of all coating films. Overall, polyols with increased hydroxyl content resulted in higher cross-linking density, which enhanced corrosion resistance of the coatings. Owing to its rigid structure, IPDI-based PUDs exhibited higher Tg and tensile strength but suffered low flexural strength and elongation than HDI-based PUDs [45].

*Epoxy Curing Agents*

Conventionally, cardanol based phenalkamines and phenalkamides are used as curing agents for epoxy coatings. To avoid their high VOC emission, recently, CNSL-based waterborne polyamines that are stable in water without the need of other higher VOC co-solvents were developed by Cardolite Corp. USA [46]. Experimental studies were carried out by Xu et al. and performance of three different types of waterborne curing agents was assessed in epoxy coatings. Three waterborne curing agents were mainly based on modified polyamine (CA-1), CNSL modified polyamine (CA-2), and first generation waterborne CNSL curing agent (CA-3). These curing agents were formulated with five different solid epoxy resin dispersions individually and characterized for performance properties. Results revealed that modified polyamine and CNSL modified polyamine curing agents were easy to dilute to lower viscosities even with small addition of water as compared to pure CNSL-based curing agents. For example, viscosity of CA-1 and CA-2 dropped significantly as compared to CA-3, which remained at 20000 cPs even after the addition of 25% water. Moreover, lower viscosities and pot life of >3.5 h of CA-1 and CA-2 were enough to make them application friendly. Their study also demonstrated that CA-3 exhibited similar pot life and less hard dry time as compared to commercially available curing agents as well as CA-1 and CA-2. To evaluate the anti-corrosion properties, these curing agents were formulated with single solid epoxy dispersion of 53% solid content and application done on different metal substrates with varying dry film thicknesses. Salt spray studies conducted on different metallic panels like blasted steel, cold rolled steel, galvanized steel, and stainless-steel suggested that severe blistering was observed on galvanized steel panels when exposed to 1000 h as compared to other metallic panels exposed for the same duration. CA-1 and CA-3 formulated primers displayed better wet adhesion properties as compared to commercially available curing agent-based primers and CA-2. Overall, these zero VOC waterborne CNSL curing agents provided superior properties such as fast curing, better adhesion, mechanical strength, corrosion resistance, and water resistance to the epoxy coatings [47].

In a similar line of work, Nadkarni et al. disclosed preparation method of water thinable phenalkamines based on CNSL. The first step of synthesis involved diels alder reaction between unsaturation of itaconic acid with that of unsaturation present in meta substituted side chain of CNSL at 200°C for 5 h. The resultant acid functional intermediate with acid value 73 mg KOH/g was further reacted with polyamines and paraformaldehyde at 70-80°C for 1 h and later at 130-140°C for 4-5 h, to yield water thinable phenalkamine with amine value of 280 mg KOH/g. The synthesized phenalkamine could be thinned with water up to 50 wt% and evaluated in liquid epoxy paint for performance properties. Experimental studies revealed that coatings cured with water-based phenalkamine exhibited touch dry time of 5 h and 18 h as well as hard dry time of 9 h and 40 h at 25°C and 5°C, respectively. Salt spray exposure to 400 h was also observed to be excellent along with outstanding overcoat adhesion with epoxy and polyurethane coatings [48].

Another method for developing waterborne epoxy curing agent was to carry out the Mannich reaction, in which cardanol reacts with the triethylenetetramine and formaldehyde to yield cardanol tetramine (CTA) as an intermediate which was shown by Juan Wang and coworkers. In the second step, diepoxy poly (ethylene glycol) also known as D-PEG undergo ring opening reaction with CTA to form waterborne epoxy curing agent (as shown in Figure *6*) containing water up to 30%. Furthermore, the structure of waterborne curing agent was confirmed by FTIR and Nuclear Magnetic Resonance (NMR) spectra through two-step reaction. The synthesized waterborne curing agent demonstrated better compatibility with epoxy resin as well as graphene oxide that was dispersed later to enhance the barrier and corrosion properties of the coating film. Since cross-linking of epoxy resin occurred at high temperature, resultant coatings based on cardanol curing agents displayed longer cracks, fractured surface, and micro pores. However, the addition of graphene oxide in appropriate amount facilitated the denser, shorter cracks, and flat surface on coatings. Incorporating 0.25 to 0.1 wt.% graphene oxide as nanocomposite in waterborne epoxy coatings enhanced corrosion resistance, barrier properties, and shielding effect as evaluated by open circuit potential and electrochemical impedance spectroscopy [49].



Cardanol based waterbased epoxy curing agent

**FIGURE 6: Cardanol-derived water-based curing agent for epoxy resin**

*Acrylic Emulsions*

Emulsion polymerization plays a vital role in developing these water-based coatings as water itself acts as a dispersion medium. However, synthesizing fully bio-based emulsion is still challenging [50]. Monomers are major constituent of water-based latexes which are derived from petroleum sources, among which, styrene is one of the most widely used monomers in emulsion that is toxic, carcinogenic, and mutagenic in some contexts. Replacing these monomers by some non-harmful bio-based resources will reduce the carbon footprint by great extent [51]. Cardanol seems to have a scope in synthesis of emulsifiable condensation monomers due to its unique structural features like phenolic hydroxyl, aromatic ring, and unsaturated alkenyl side chain.

In this regard, Ladmiral et al. prepared cardanol methacrylate by reacting cardanol with excess of epichlorohydrin followed by reaction with methacrylic acid in the presence of triphenylphosphine and hydroquinone to yield cardanol methacrylate (CAMA) monomer (as shown in Figure 7). Structural confirmation from NMR spectroscopy clearly displayed the functionalization of the phenolic moiety. Further, CAMA was homopolymerized in the presence of ammonium persulfate as thermal initiator and sodium dodecyl sulfate as surfactant. It was observed that conversion of CAMA in polymer was greater than 90% and coagulum was found to be less than 2%. This P-CAMA (Polymer of cardanol methacrylate) showed sufficient polydispersity with particle size ranging from 50 to 110 nm. In another variant, CAMA copolymerized with Methyl Methacrylate (MMA), in which copolymerizability of both monomers was monitored by using NMR spectroscopy. MMA being highly reactive than CAMA, found to be consumed earlier in the polymer. Owing to the steric hindrance offered by the pendant group on cardanol, after 2.5 h MMA consumption was 90% as against 80% of CAMA. Differential Scanning Calorimetry curves represented considerable shift in Tg from 30℃ at 25 wt.% to 40℃ at 100 wt.% of CAMA in monomer feed. P-CAMA latexes contained small amounts of dithiols that helped in photo cross-linking and film formation. Overall, synthesis and utilization of CAMA, not only as a sole monomer but as a comonomer with MMA in latexes resulted in high conversion, less coagulum, high stability, good particle size, and wide range in film forming temperature. Authors concluded that cardanol methacrylate monomer can be a good alternate to conventionally used styrene in emulsion [52].

Another route to synthesize CAMA is via hydroxyethylation of cardanol, which was carried out most recently by Li et al. Cardanol, along with cyclic carbonate, mostly ethylene carbonate, is reacted in the presence of 1,5-Diazabicyclo(4.3.0)non-5-ene (DBN) catalyst at a temperature of 180℃ for 4 h. As phenolic hydroxyl group of cardanol is very much resonance stabilized due to aromatic ring, it does not not allow cardanol to take part in reactions occurring at normal conditions. Therefore, authors performed hydroxyethylation to move the phenolic hydroxyl group away from the aromatic ring to form hydroxyethylated cardanol followed by esterification with excess of methacrylic anhydride to yield CAMA. Authors carried out solution, emulsion, and miniemulsion polymerization using CAMA as a sole monomer or in combination with other comonomers to study its conversion in final polymer. In solution polymerization, copolymerizability of CAMA and MMA monomer observed to be nearly equal and no significant compositional drift was detected in conversion vs. time curve. When homopolymer of CAMA and copolymer of CAMA-MMA were compared in thermogravimetric analysis, P-CAMA demonstrated higher thermal stability than P-(CAMA-co-MMA). Approximately 15% residue was observed for P-CAMA as against 0% for P-(CAMA-co-MMA) at 450℃. In emulsion polymerization, around 81% conversion of the CAMA monomer was recorded when emulsion polymerization was carried out by conventional method. Resulting latex from the copolymerization of CAMA and MMA showed poor colloidal stability and low conversion of monomer even at higher cycle time due to low solubility of cardanol in water as compared to MMA. To improve stability of emulsion and conversion of CAMA, miniemulsion polymerization was carried out in the presence of MMA. Different monomer feed of CAMA and MMA were taken to study the influence of cardanol methacrylate on physical and chemical properties of the resulting films. Cycle time of miniemulsion copolymerization was faster than solution copolymerization and the rate of polymerization increased with increase in the wt.% of MMA in CAMA-MMA monomer feed. Like solution polymerization, homopolymerization of CAMA was slower than copolymerization and, in both cases, redox initiators polymerized the unreacted monomer making the conversion approximately upto 98%. Performance properties revealed that films copolymerized with 25 wt.% CAMA and 75 wt.% MMA were brittle in nature with no tackiness and increasing the CAMA content in film increased tackiness and softness. Thermal stability of final polymer was observed to be directly proportional to wt.% of CAMA in polymer. P-CAMA presented lower decomposition of mass as compared to P-(CAMA75-co-MMA25), and first degradation loss curve was observed at 200℃ for P-CAMA as against 150℃ for P(CAMA75-co-MMA25) [53].

Dechmuk et al. used high oleic soyabean (HOS) based monomer as a comonomer with CAMA in miniemulsion polymerization. CAMA was synthesized in a similar fashion as explained earlier, while HOS was synthesized by transesterification reaction of crude oleic soyabean oil with N-hydroxyethyl acrylamide in the presence of NaOH catalyst. Due to natural differences between their structure both monomers had different levels of unsaturation as represented by their iodine values 202 gI2/100g for CAMA and 123 gI2/100g for HOS. CAMA and HOS copolymerized at different monomer feed ratio resulted in different wt.% in polymer due to their different reactivities. CAMA ratio in monomer feed was inversely proportional to the molecular weight of final polymer in miniemulsion due to its high unsaturation alongside chain, which favored more chain transfer reactions. Degree of unsaturation in CAMA and HOS monomers provided a suitable pathway to cross-linking after polymerization, which ultimately increased its thermomechanical properties. Cross-linking density showed direct relationship with the wt. % of CAMA in monomer feed due to the higher level of unsaturation in CAMA. In addition, Young's modulus and toughness were also improved with higher concentration of CAMA. Overall, cross-linking density, glass transition temperature, Young's modulus, and toughness of emulsion films improved directly as concentration of CAMA in monomer feed was increased and best results were obtained when CAMA was homopolymerized in miniemulsion process [54].

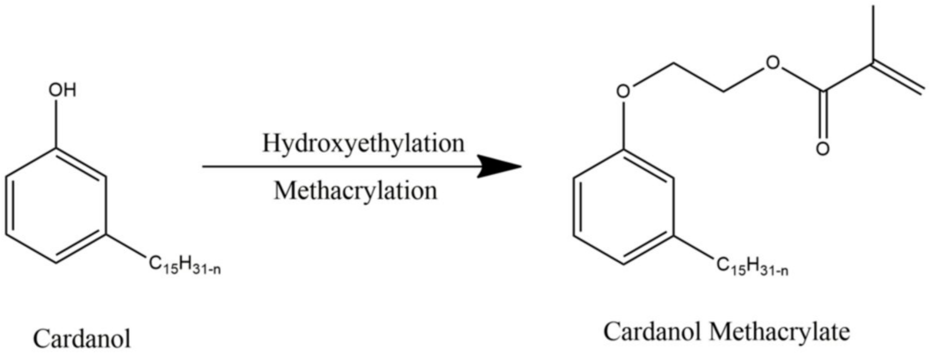


**FIGURE 7: Synthesis of cardanol methacrylate (CAMA)**

*Economical and Ecological Aspects*

The CNSL market is facing significant growth due to the increasing demand of renewable and environment friendly products. However, the industry faces various challenges such as market volatility, shortage of cashew nuts, and stringent regulations that needs immediate attention. CNSL market is known for its volatility influenced by factors such as weather conditions, global demand, and geopolitical events. This uncertainty poses challenge to cashew supply chain and provide stability and fair compensation to all stakeholders. Secondly, the composition and quality of CNSL varies depending on the region and method of extraction, and hence it becomes difficult to maintain consistent quality standards for CNSL as well as allied industry.

Considering environmental impact, cashew cultivation requires deforestation to produce cashew orchards in high yields. This raises concerns about soil erosion, biodiversity loss, and overall ecological balance. In addition, harvesting and processing cashew nuts involve labor-intensive tasks that are hazardous such as extraction of nut from shell due to the presence of caustic oil. Therefore, strategies for sustainable cashew farming, reforestation efforts, automation, innovations in technology, and fair labor policies are required for streamlining processes and crucial for mitigating environmental impact.

### *Challenges*

Greater utilization of cardanol and its derivatives in polymer and coatings is an attractive proposal due to its low cost, abundant availability, and other multifunctional attributes. However, certain challenges and disadvantages limit its utilization in a variety of applications. One of the major disadvantages of cardanol is its dark color. During extraction of cardanol, it is difficult to remove entire cardol, which is responsible for imparting dark brown color to cardanol. This dark color of cardanol restricts applications in decorative coatings and enamels where aesthetics and shade play a major role. With advancements in technology, dark color can be bleached to obtain colorless cardanol, but the process is tedious, and the cost involved is too high. Another common issue with cardanol is the purity and composition of saturation and unsaturation chains present in it. Cardanol contains four types of C15 aliphatic chains in its structure, and composition of these chains varies depending on region, extraction, isolation and purification methods, chemical composition of CNSL, and so on. The changes in composition of saturated and unsaturated chains in cardanol result in products with inconsistent quality and hence proper quality control measures need to be placed to resolve such issues.

## Conclusions

Cardanol is an agricultural byproduct of cashew nut industry. Its easy availability, low cost, and inedibility make it a promising candidate for wide variety of applications across various industries. The seamless modification of available functionalities in cardanol further enables scientists and researchers to explore cardanol in various domains. The global production of cardanol has already reached 800 MT/annum and continues to increase in coming years. Cardanol and its derivatives-based waterborne systems are gaining a lot of attention nowadays as it addresses two of the most significant points, i.e., sustainability and reduction of VOC. Therefore, Cardolite Corp., the largest developer, manufacturer, and marketer of CNSL, has started exploring CNSL as well as cardanol in waterborne systems long back and recently commercialized a few waterborne curing agents for epoxies and surfactants for water-based emulsions. In India, with increase in demand of cardanol, several companies were involved in extraction of cardanol from CNSL such as Satya Cashew Chemicals Ltd., Adarsh Industrial Chemicals, Golden Cashew Products, Akshar Cashew Industries, and others.

Cardanol is a versatile material with huge potential in the development of waterborne polymers and resins which are yet to be fully explored. In coming years, a lot of research is dedicated to the development of cardanol-based waterborne systems and its cost-effective solutions.

## Additional Information

### Author Contributions

All authors have reviewed the final version to be published and agreed to be accountable for all aspects of the work.

**Concept and design:** Kunal D. Wazarkar

**Acquisition, analysis, or interpretation of data:** Kunal D. Wazarkar, Tushar Singh, Muhammad D. Saad

**Drafting of the manuscript:** Kunal D. Wazarkar, Tushar Singh, Muhammad D. Saad

**Critical review of the manuscript for important intellectual content:** Kunal D. Wazarkar

**Supervision:** Kunal D. Wazarkar

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