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Use of a Cashew Nut Shell Liquid Resin as a Potential Replacement for Phenolic Resins in the Preparation of Panels – A Review

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The Cashew Nut Shell Liquid (CNSL) can be considered as a versatile raw material with wide applications in the form of surface coatings, paints and varnishes, as well as the production of polymers. Within this context, the chemical constituents of CNSL (anarcadic acid, cardanol, 2-cardol and methylcardol) become promising in the development of new materials components. Once separated, CNSL can be used in the research and development of additives, surfactants, pharmaceuticals, pesticides, polymers, resins and others. Being a byproduct, CNSL used in the preparation of new materials is characterized as a truly technological innovation.

Keywords: Biocomposites; cashew nut shell liquid; phenolic resins

1. Cashew Nut Shell Liquid - CNSL

The cashew (*Anacardium occidentale* L.) is an exotic plant species with glabrous leaves, crooked trunks and male and hermaphrodite flowers. Its stalk is overdeveloped and is widely used in the preparation of juices and is confused with the fruit. The pseudo fruit can make juices, jellies, jams, wines and syrups. The main producer countries of cashews are in Asia, Africa and South America [1–6].

The shell, which is around 50 wt.% of cashew nuts, produces a dark, heavy liquid with a lingering scent. One can remove 25% liquid caustic, flammable having characteristics. This material is called cashew nut shell liquid (CNSL) as it is known internationally [6].

Currently, the plant is widespread in many countries like Brazil, India, Mozambique, Tanzania, Kenya and more recently Vietnam, Indonesia and Thailand [7]. In northeastern Brazil, there is a large concentration of nut processing industries, due to extensive plantations and favorable climate. Ceará state in Brazil owns about 70% of installed capacity in the region for production and processing nuts. About 20 thousand people are involved in the process, and provide thousands of jobs in the field [7].

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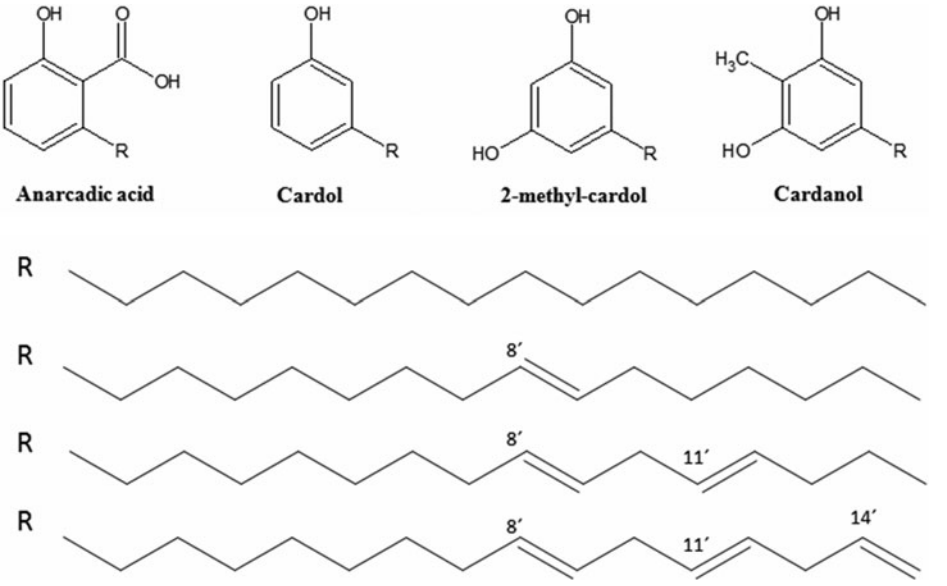

Figure 1. Major components of the CNSL.

2. Chemistry of CNSL

The liquid from cashew (CNSL) is considered a byproduct of the cashew agribusiness, having very low value. The liquid is a source of phenolic non-isoprenoid lipids of natural origin containing as anacardic acid, cardanol, cardol and isomers. Its main chemical composition can be seen in [Figure 1] [8–10]. The oil has many applications in the fine chemicals industry according to the functionalization of the isolated products [11–21]. From this fact, one can understand the importance of effective extraction and its relation with the best possible performance of phenolic compounds.

3. Production of CNSL

The traditional extraction processes nowadays available can be used to obtain the CNSL through cold extraction action presses, solvent extraction and thermal-mechanical processes (hot oil process) at 190°C [22]. At high temperatures, the outer shell can break and release alkylphenols present in the mesocarp, with subsequent removal of the inner bark. This allows the recovery of almonds. Another widespread process available nowadays is by supercritical fluid extraction using CO_2 under supercritical conditions to obtain nonpolar components. The efficiency of the process reaches values up to 100% [22–24]. Even at high temperatures, anacardic acid can suffer a decarboxylation reaction. This reaction leads to the formation of cardanol, producing a fluid called nut technical cashew widely used in the preparation of new materials [Figure 2] [11–13].

The natural CNSL can contain a large amount of anacardic acid and shows polymeric material in its composition. However, so-called technical CNSL can provide a high percentage of cardanol, and polymeric material, as the process occurs at high temperature [7]. The presence of the polymeric material reduces the quality of CNSL since there is the need for further purification processes that can make unfeasible product industrially.

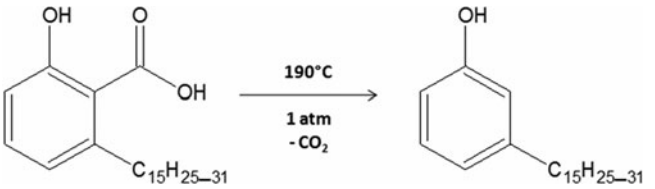

Figure 2. Production of cardanol from anarcadic acid.

The cardanol is fundamental in CNSL, because it is one of the most important and promising components. After their separation it can be used in diverse industries such as fine chemicals. In this sector, the costs and prices are exorbitant. It can be mentioned the production of plastics additives, surfactants, pharmaceuticals, pesticides, resins and others. Considering that the oil and its components are byproducts, any research and development with this material, can lead to the development of new products and improvements of the existing ones. This path may lead to development of new and innovative materials [7].

In the chemistry of phenolic compounds, the double bonds have an important role, being responsible for the large chemicals and physical-chemicals characteristics. The position of the double bond (C8, C11′ and / or C14′), is called in accordance with the isomer, allows numerous functionalizations, beyond the possibilities envisioned with the double bonds of the aromatic ring [see Figure 2]. Various activities are associated with pairs, such as antioxidant, flame resistance, hydrophobicity compounds, etc. [5–7].

One of the great advantages in the development of new materials is that the cardanol has no aggressive smell and can be worked on an industrial scale without causing cross contamination. Also has low volatilization and higher boiling point than the other phenolic compounds derived from petroleum. This point is important, since many oil products cause harm to the human health and the environment [5–7].

Several reactions can be carried out, by improving the ability of developing new products. One example is the possibility to produce prepolymers from the oligomerization of phosphorylated cardanol. The unsaturations of its side chain are essential in this process. This opportunity may lead to development of flame retardant additives, essential in the development of new housing and automotive materials. The union of these possibilities with elastomers and plastics, such as polyethylene (PE), polyvinyl chloride (PVC); natural rubber; cellulose and polyurethane enlarge the chance of developing new products [7]. Besides the possibility of developing new materials, cardanol is a renewable raw material, and is considered an important intermediate chemical non-toxic to the environmental [7]. The cardol is present composing 18% of the liquid from cashew nuts. Despite its considerable level, is one of the least studied constituents. Its aromatic system and the long side carbon chain may arouse interest in its use as a precursor in many organic syntheses [3, 7].

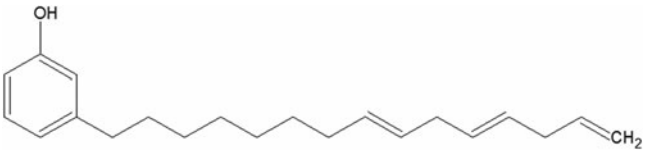

Figure 3. Carbon double bonds of the side chain of cardanol molecule.

Given these opportunities, the purification of the active components of the liquid from the cashew nut can provide a source of new precursors of new molecules and new materials, including industrial scale. Various processes and developments are being reported in the literature, but finding difficulties related to the low percentage yields of pure cardanol, since many processes have high costs of raw materials and reagents, using analytical techniques such as chromatography [25–30]. Besides this issue, the contamination of the substances of interest in CNSL with polymeric material can occur, preventing its getting on an industrial scale.

Research and development on a laboratory scale allows the use of chromatographic techniques in low-pressure column, which allows the complete separation of cardanol the other components of technical CNSL. Several studies reported the possibility of obtaining up to 81% yield of cardanol by chromatography [27, 29]. However, its industrial application is not economically viable. Several tests of scale-up have yet to be performed, but these tests require further investments [25–30]. Among the several studies seeking possibilities that can resolve the problems mentioned here, some studies report on the possibility of cardol complexation and 2-methylcardol using diethylenetriamine. The results were good as it can achieve 25% cardanol, cardol with lower concentrations and 2-methycardol [25].

Other studies have shown the possibility of obtaining processes using liquid-liquid [26] were extracted, but contamination with other substances are observed. There are no reports of using green solvents in processes of liquid-liquid extraction.

Chitosan microspheres can be used in association with chromatographic processes promoting complexation reactions with components of CNSL acting as complexing agents of di-hydroxylated CNSL [30, 31].

On an industrial scale, more efficient unit operations must be considered, and vacuum distillation may be an option; however, this process does not guarantee the purification of the cardanol [32–37]. Contamination is still found by polymers that decrease the yield of the process. Some patents suggest the use of amines for the purification of cardanol [36, 37]. Based on the above data, it appears that for most substances that have interests in the purified CNSL, these components can be separated without contamination only in laboratory scale. There is a need for more investments in industrial scale up and new techniques to obtain these compounds. Chromatographic systems require exorbitant investments and solvent extraction processes are against the principles of sustainability. There is the need to improve processes and yields that meet the technical and economic demands. Partnerships between academia and the areas of R&D from private companies should be consolidated, allowing agribusiness to this new and appropriate approach, especially the possibility of adding value to CNSL is still little explored.

4. Phenolic Resins and CNSL

The world economy currently revolves around the petrochemical industry, which created a network of dependency in fossil based energy and materials. However, research under new concepts are bringing new possibilities to reduce this dependency. In recent decades, studies have been done in search of replacing fossil resources due to its limitations and disadvantages, such as volatile prices and effects harmful to the environment [52]. As a solution, the use of natural resources like biomass becomes a great option because it provides energy and a variety of opportunities for obtaining new materials. Biomass is available across the globe and its use is driving economies and contributing to the reduction of emissions of greenhouse gases and pollutants.

The CNSL is an option for this application. Several studies and surveys have been conducted seeking to develop new materials from biomass, including CNSL [11, 16, 38, 39]. One of the most important applications, which is critical in the production of resins and polymeric derivatives, is the substitution of petroleum, much more expensive and toxic [40–42].

The polymers of CNSL can be obtained by polycondensation process with electrophiles (formaldehyde), by polymerization of unsaturation present in the side chain with acid catalysts or from reactions of the hydroxyl group, followed by oligomerization. Thus, it can be important to obtain functionalized prepolymers [7].

A monomer bifunctionalized HPPDP (4-[(4-hydroxy-2-pentadecenylphenyl) diazenyl] phenol) was obtained from cardanol, and polymerizations were performed to obtain a copolyester [43]. These polymers can be used in ion exchange resins, anticorrosive paints, waterproof materials, flame retardants, coating of surfaces, friction materials and rubber modifiers [7]. Polymers with flame retardant properties are used aiming to improve the heat resistance of other polymers, depending basically on its compatibility and miscibility.

Through the oligomerization of cardanol side chain can be phosphorylated obtain prepolymers. These prepolymers can act as flame retardant additives in addition to various materials such as plastics and elastomers such as polyethylene (PE), polyvinyl chloride (PVC), natural rubber, cellulose and polyurethane [50].

Various resins obtained from CNSL can be part of the composition of surface coatings, epoxy resins, wood composites, adhesives and in the manufacture of anionic and non-ionic agents [53]. In the field of polymers, CNSL has been studied as a modifier for phenol formaldehyde resins, due to their structural similarity with phenol [52]. These alternatives are the most promising using the unprocessed CNSL.

The resins obtained from the CNSL are more malleable and are soluble in organic solvents different from other resins that are more rigid and this is a limitation in certain applications [7]. Because they are hydrophobic resins, have greater resistance to the action of acids and bases. The aliphatic chain containing 15 carbons in the *meta* position of the phenolic ring is responsible for the hydrophobicity.

Soluble aromatic polyimides can be prepared from a derivative of hydrogenated cardanol diamine. This new material has allowed the development of transparent and flexible compound in chloroform solution [51], suitable for various applications.

Analysis by thermogravimetric analysis (TGA) showed that when adding the crude CNSL into phenol formaldehyde resin, the thermal stability of the new resin may be altered. Materials such as plywood, when produced with a blend of resins CSNL has a higher decomposition temperature which makes it a material resistant to thermal decomposition. One can use up to 20% (w) CNSL in place of phenol resin - formaldehyde reducing the emission of free formaldehyde [52].

In mixtures of phenolic resins, the CNSL can be used benzoxazine resins. These resins are widely used in adhesives, phenolic resins that also employs the base. The use of CNSL showed better thermal characteristics, allowing the development of more resistant resins flame, adding flexibility to new materials, allowing low moisture absorption and low shrinkage rate [53–57].

Other studies show that a new benzoxazine-based monomer containing an oxazine cardanol in its structure has been prepared. This was possible, since the rings present in the benzoxazine react with the aromatic ring of cardanol [58]. Addition of CNSL resins with benzoxazine can substantially reduce the liquefaction temperature, the gel time, cure temperature and the activation energy of these resins, opening new opportunities for the use of these materials [59]. These resins were used in tests with wood composites. Due

to the low viscosity of the mixture BA-a/CNSL, the material showed good compatibility with fibers mainly bamboo. According to the results, there was a significant reduction in moisture absorption and swelling of the material due to moisture [60].

Several phenolic resins can be developed to condense cardanol with hydrogen-rich compounds, such as resols and novolac [61–69]. These resins may be modified to develop epoxy resins with epichlorohydrin, to improve the performance of new materials [70–73]. The introduction of an unsaturation at the end of the epoxy resin by reaction with monomer acrylic acid / methacrylic acid can produce an important eco-friendly vinyl [74]. Vinyl ester resins are obtained by reacting an epoxy resin and an ethylenically unsaturated carboxylic acid. This results in a polymer with chain end unsaturation. Therefore, various epoxy resins such as diglycidyl ether of bisphenol A or higher homologues, phenol novolac resin - formaldehyde and diepoxide epoxidized polypropylene oxides, etc., are being developed for use in new materials [75]. In preparing these new resins and acrylic methacrylic acids are being used. The reaction of acids with epoxides is simple and can be catalyzed by ternary amines, phosphines or alkalis [75–77]. These resins can be widely used as thermosetting matrices to produce a variety of reinforced structures including pipes, tanks, scrubbers, panels and pipes. In addition to these applications, vinyl esters have also been used as coating materials, adhesives, molding compounds panels, structural laminates, electrical insulators [74].

Results of analysis indicated that it is possible to optimize the esterification reaction of epoxidized novolac resins with methacrylic acid and cardanol basis. With this reaction can produce a vinyl ester resin-based cardanol. The data showed that cardanol-based epoxidized novolak is reacted with methacrylic acid in a molar ratio of 1: 0.9, at a temperature of 89.96 °C for 5 h in the presence of triphenylphosphine catalyst (1.49%). Thus, a new resin can be designed to be used using components of CNSL in the study of new materials [74].

Novolak phenolic resins based on cardanol were synthesized with different molar ratios of cardanol-to-formaldehyde: 1: 0.6, 1: 0.7, 1: 0.8. These were epoxidized novolac resins with epichlorohydrin molar to 120°C in excess basic medium. Epoxidized novolac resins were separately mixed with different weight ratios of polybutadiene ranging from 0 to 25% by weight, with a range of 5% [98]. All blends were cured at 150 °C with 40% by weight of polyamide. The tensile strength and elongation-at-break of the cured samples showed an increase of up to 15% by weight in the mixture [98].

The addition of CNSL resin and hemp fiber to obtain composites was studied. After getting into molds, the samples were analyzed by physico-mechanical methods: strength, porosity and surface topography. The use of CNSL improved the mechanical properties of the biosensor. The use of CNSL as binder contributed to a better interaction fibers - matrix, allowing the material to offer better physical and mechanical [78] properties. Sisal fibers treated with 5% NaOH and CNSL allowed the elaboration of a composite with improved thermal stability [80].

Some studies highlight the use of mixtures of hydrolyzed tannins, CNSL and urea formaldehyde in the preparation of panels with waste coffee pods. The results showed that the composite is more resistant to water and humidity. However, the thermal and mechanical properties were slightly higher than the data obtained using synthetic resins for biomaterials [79].

Phenalkamines can act as agents of commercial epoxy cure and are a series of Mannich bases obtained by reaction of cardanols, aldehyde compounds and polyamines [81]. The phenalkamines derived from cardanol have a fast cure characteristics at low temperature and allows the application at wet or damp surfaces [82, 83]. However, there was one drawback:

dark and unstable color. This condition limited the use of these resins, until Huanga and colleagues successfully addressed the issue of color and its stability [84].

Acid catalyzed, CNSL resins with formaldehyde and $CuCl_2$ were used in the preparation of adhesives for the production of pellets. CNSL-formaldehyde resins are widely used since the 80s and show excellent abrasion in new product development [86, 87]. Agglomerates developed from rice husk mixed with resin and CNSL with formaldehyde and alkali catalysts were created [88]. Fiberboards medium density were produced with coconut fibers and prepared with CNSL resin and formaldehyde giving good yields [89, 90].

Panels jute reinforced with a mixture of phenol resins-CNSL and formaldehyde were produced and tested [91]. Apart from coconut waste jute for preparation of materials for construction were tested. The CNSL was reacted with phosphoric acid and after neutralization, it was mixed with formaldehyde and HMTA [92].

The thermal behavior of biocomposites of CNSL-hemp and kenaf-CNSL were studied by DSC [93]. A large endothermic peak was observed for the composite fiber hemp-CNSL, while with the compound of the kenaf fiber-CNSL showed an endothermic peak and two exothermic peaks in their thermal analysis curve. The width of the endothermic peak observed for both compounds was between 40 and 150°C, indicating the presence of water adsorbed on the composite. The first exothermic peak, which was small in the case of composite kenaf fiber was 174.7°C, indicating incomplete cure of the resin, while the second at 275°C was associated with the fiber.

The physical-mechanical results showed that the epoxy resin - cardanol exhibits improved properties compared to the pure epoxy resin. An increase in tensile strength, elongation and reduction of water vapor transmission of the film was observed. Improvements in these properties indicate that the new material developed is more durable than other epoxy-based materials [94].

New materials formulated with resin were compared with their counterparts made of epoxy resins unmodified. Zinc powder, zinc phosphate, iron oxide and synthetic iron oxide were used as pigment, with fillers, additives and an aromatic polyamine as a hardener. Improvements in the physical and mechanical properties, chemical resistance and greater efficiency in corrosion protection were observed [94].

Other works propose the manufacture of roofing panels reinforced with sisal fibers using CNSL [95]. As synthetic resins are more expensive than the fibers in the preparation of a composite, the author suggests the use of CNSL in developing a new material that can replace fiberglass. Through a condensation polymerized with formaldehyde, created a natural resin that has been applied on a carpet fiber in a compression mold. It was found that the composites are suitable for roofing and other construction purposes [95].

In India, various materials have been developed with *Ipomoea carnea* species, and to improve their physical and mechanical characteristics, various materials are being tested, including the CNSL. The results showed that this type of coating has been applied successfully extending the life of wood in constructions under various conditions [96].

Studies report that CNSL can be used as resin in the preparation of adhesives, and may be used in the production of medium density fibreboard (MDF) panels. Thus, the developed composites can be used in panels and finishes, as well as covers of tables and cabinets, doors and shutters [97].

Resins derived from CNSL can be used as binders for agglomerates. The resin obtained by heating CNSL, phenol and hexamethylenetetramine showed good properties of tensile strength, strength and compressibility perpendicular traction. It was found that 15% of the produced resin has allowed the development of a board of acceptable quality. The used resin contained 20% of CNSL and no changes were observed in material properties [100].

In the development of resins, it is important to study the cure temperature. Tests have shown that temperatures of 150, 160, 170, 180 and 190°C can affect the action of resins on the properties of the panels. The results showed that at 150°C after 15 minutes already get a high degree of curing [101]. Levels above 15% resin, makes the material more resistant to traction. Thus, it is established that an ideal mixture of phenolic compounds: phenol is l: 2.9 and may use up to 50% of CNSL to ensure good compressibility. However, the content of CNSL can reduce the tensile strength [101].

5. Perspectives

The use of fossil fuels for the production of resins is responsible for about 4–5% of the world's oil consumption, with the prospect of increased demand in the future. The challenge is to promote the reduction of dependence on these fossil fuels and protect the environment by reducing CO_2 released, as well as preserve the environment against the harmful effects of the indiscriminate use of these fuels. A search for reductions in developing new materials stimulates a demand in developing new technologies and innovative materials with biodegradable and with Eco's characteristic. Alternatively, the shell liquid cashew (CNSL) is an agricultural byproduct abundantly available and is one of the main economic sources of natural phenols. Because it is a versatile raw material and with broad applications, the CNSL can be applied in developing a range of new materials in the areas of surface coatings, paints, varnishes, binders and resins, as well as in the production of biocomposites.

The molecules of the major components of CNSL, cardanol highlighting, become components of great interest. Several byproducts from the chestnut industry are available for materials improvements, which may mean the development of products with high added value and technological innovation. The cardanol molecule becomes a key player in developments with CNSL. Currently, companies already sell the CNSL and its derivatives, such as CASKYD [102], SENESEL [103], KANCO [104] and Aboissa [105] among many others. The developed products meet the markets of paints and varnishes, printing, insulation, rubber, corrosion, coatings, friction resins, etc. Moreover, firms produce resin with CNSL according to customers' desired cardanol specifications levels. The challenge is to find new applications, in addition to technology innovation of the existing processes.

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