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Synthesis, characterization, and performance characteristics of alkyd resins based on *Ricinodendron heudelotii* oil and their blending with epoxy resins

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ABSTRACT

Ricinodendron heudelotii oil is a good source of polyunsaturated fatty acid, mainly α-eleostearic, a triple conjugated double bonds diene fatty acid chain. Three sets of long oil alkyd resins based on *R. heudelotii* oil have been synthesized by a two stage alcoholysis-polyesterification process at relatively low temperatures (190 °C) with different molar ratio of phthalic and maleic anhydride. The synthesized alkyd resins were characterized by FT-IR, ^{1}H-NMR, and ^{13}C-NMR spectroscopy and gel permeation chromatography (GPC). The synthesized alkyd resins were blended with epoxy resin and methyl-ethyl ketone peroxide (MEKP) as initiator, cobalt-octoate as accelerator and poly(amido amine) as hardener. The blends were cured at room temperature and also at 50 °C. The cured resins exhibited improved performance characteristics in term of drying time at room temperature, adhesion and pencil hardness, gloss, and chemical resistance. The cured resins exhibited good tensile strength, improved elongation at break and thermally stable up to 400 °C.

1. Introduction

The growing use of polymer materials coupled with the depletion of petroleum-based resources demands for the replacement of some synthetic raw materials with renewable and inexpensive natural resources in order to produce eco-friendly environmental products (Oprea and Doroftei, 2011). The advantages of these bio-based polymeric materials are their renewability and biodegradability (Li and Larock, 2003). Amongst wide range of polymeric materials derived from renewable resources, alkyd resins have gained more interest due to their versatility and ease compatibility with others.

Alkyd resins are prepared from the condensation of polyhydric alcohols, polybasic acids, and monobasic fatty acids or oils (Odetoye et al., 2013). Variations in the amount and types of components give enormous varieties of resins with different properties. Alkyd resins are used in the formulation of paint, surface-coating, adhesive, lacquer, and constitute a very high proportion of conventional binders used in the coating industry. Other unique properties of alkyd resin are high gloss, gloss retention, good adhesion, good heat, solvent resistance, flexibility, and durability.

However, alkyd resins lack from important properties such as alkali resistance, low mechanical strength and hardness, and fair thermal stability. These drawbacks can be improved by blending alkyd resins with others resins such as epoxy resin, amino resin, silicon resin, and ketonic resin (Gogoi et al., 2014; Bora et al., 2014; Patel et al., 2010). Long chain fatty acid groups of alkyd resins impart non-polar characteristics and promote compatibility with other non-polar materials such as vinyl resins. Hydroxyl functional groups of the alkyd resin can react with acid and epoxy groups of the other resins. Moreover, unsaturated double bonds in the fatty acid chain can be used to graft conventional polymer chains such as polystyrene (Oyman et al., 2005).

Various vegetable oils based alkyd resins that have been synthesized are: Tung (*Aleurites fordii*) or Chinese wood oil (Thanamongkollit et al., 2012; Xiao, 2008), tall oil fatty acid (Uschanov et al., 2008; Rämänen and Maunu, 2014), soya bean fatty acid (Yin et al., 2014), yellow oleander (*Thevitia peruviana*) seed oil (Bora et al., 2014), *Parinari polyandra* benth seed oil (Odetoye et al., 2013), *Jatropha curcas* (Boruah et al., 2012), *Albizia* benth (Akintayo and Adebowale, 2004), Nahar (*Mesua ferrea*) seed oil (Dutta et al., 2004), Rubber seed oil (Aigbodion and Pillai, 2001), and Castor oil (Hlaing and Oo, 2008).

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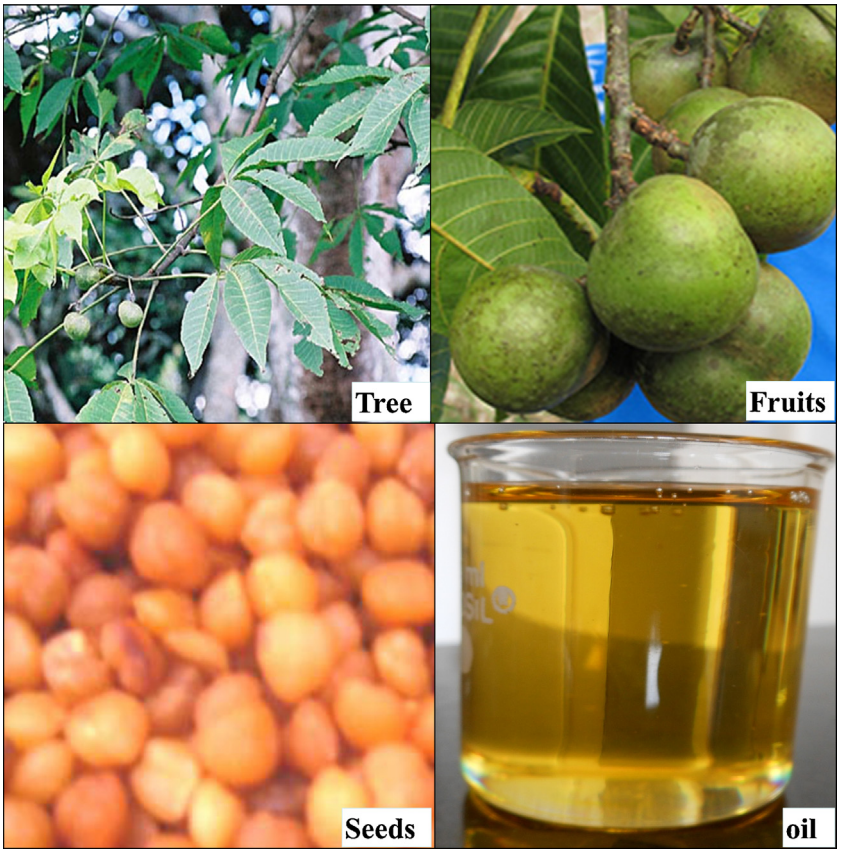

Fig. 1. Tree, fruits, seed, and oil of *R. heudelotii.*

Ricinodendron heudelotii (*R. heudelotii*) is a fast growing tree belonging to the Euphorbiaceae family. It grows naturally in tropical forest in Africa and can reach up to 40 m in height and 120 cm in diameter. *R. heudelotii* is a common species known as Akpi in Côte d'Ivoire (Ivory Coast), Njansang in Cameroon, Okwe in south east Nigeria, Bomoko in Central Africa Republic, and Betratra in Madagascar. Fruits are green or yellow when ripe and can bear up to four seeds (Fig. 1). Its distribution area in Africa is from West Africa (Côte d'Ivoire, Ghana, and Nigeria) to Central Africa (Cameroon, Gabon, Congo, and Central Africa Republic) and also in Madagascar (Fondoun et al., 1999). *R. heudelotii* tree is considered as a plant having multiple end usage. The seed is known for its unique taste in soup and stews and is often used as a thickener or flavoring in soup and rice particularly in Côte d'Ivoire, Cameroon, and Nigeria. The tree's bark is used in traditional pharmacopoeia. It is used against coughs and as an anti-dote to poison and increases breast milk. The barks are widely used for sexual and fertility problems as well as to ease pain associated with menstruation or child birth (Izundu and Nnacho, 2011). The latex and leaves are used as purgative and for guinea-worm extraction (Izundu and Nnacho, 2011). The oil content of the seed is varied from 45 to 67% (Kapseu and Tchiegang, 1995). *R. heudelotii* oil is a mixture of triglycerides with polyunsaturated chains up to 80.5% of the total fatty acid. The oil is a good source of α-eleostearic acid (9*cis*, 11*trans*, and 13*trans*); a triple conjugated fatty diene (52.6%). Others constituents of the oil are linoleic acid (27.9%), oleic acid (6.4%), stearic acid (6.5%), and palmitic acid (6.6%) (Leudeu et al., 2009). The iodine and saponification value are 157 and 190, respectively (Tchiegang et al., 1997).

In recent years, intense research is in progress on the valorization of the seeds of *R. heudelotii* and its domestication (Ayuk et al., 1999). But these researches were only focused on the valorization of seeds as food purpose and the mechanization of the extraction of the seeds (Mbosso et al., 2013). Due to the high level of polyunsaturated acid (α-eleostearic acid) content and iodine value, *R. heudelotii* oil is a drying oil and comparable to Tung oil (*A. fordii*) and, therefore, *R. heudelotii* oil can substitute Tung oil as raw renewable material for the formulation of surface coating system, paints, and adhesives.

As far as we know, there are no reports on the synthesis of alkyd resins using *R. heudelotii* oil in spite of its high degree of unsaturated fatty acids mainly α-eleostearic. The present study aims to investigate the preparation, characterization of the alkyd resins based on *R. heudelotii* oil and their blending with epoxy resin. The performance characteristics like, drying time, chemical resistance, pencil hardness, thermal and mechanical properties of the cured blends were evaluated.

2. Materials and methods

2.1. Materials

Oil from the seeds of *R. heudelotii* from Côte d'Ivoire (Ivory Coast) have been extracted with the help of soxhlet apparatus with hexane (Merck), dried over anhydrous sodium sulfate (Rankem) and kept under nitrogen at 4 °C for further use. The comparison of physico-chemical properties and fatty acid composition of *R. heudelotii* are given in Table 1 and Table 2, respectively. Analytical grade phthalic anhydride (PA), glycerol and potassium hydroxide pellets (Qualigen), maleic anhydric (MA) (G.S. Chemical), lithium hydroxide (Merck) were used without further purification. Epoxy resin (Epoxy equivalent weight:170–180 g/eq) and poly(amido amine), methyl-ethyl ketone peroxide (MEKP), commercial grade Co-octoate (Kumud Enterprise, Kharagpur, West Bengal, India) were used as received.