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Utilization of *Acacia auriculiformis* seed (Akashmoni) oil – A new source for the synthesis of alkyd resin

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An alkyd resin is prepared from a renewable raw material *Acacia auriculiformis* seed oil (ASO), which has a high content of linoleic acid. Long oil alkyd resins have been synthesized by alcoholysis followed by the poly-esterification method using oil, glycerol, pentaerythritol (PE), sorbitol, phthalic anhydride (PA), and maleic anhydride (MA) in proportions according to alkyd formula calculation. The various physicochemical properties of ASO like acid value, saponification value, iodine value, etc, and fatty acid composition were determined. Physico-chemical properties of alkyd resins like viscosity, acid value, nonvolatile matter, etc. were determined and compared with soybean oil-based alkyd resins. The structural characterization of the resins was carried out using Fourier transform infrared (FTIR) and proton nuclear magnetic resonance (^{1}H NMR) spectroscopic techniques. These resins were cured by drier and their film properties were determined. The drying properties of the alkyd resins had been improved remarkably in the case of ASO-based resin. The performance of coating film produced by these alkyd resins were characterized by drying time, gloss measurement, hardness test, cross-cut adhesion test, and chemical resistance test. Analysis of these alkyds proved that ASO can be used as a new potential raw material for surface coating application.

Keywords: *Acacia auriculiformis*, Alkyd resin, Bio-renewable, Epoxy, Vernolic acid

Plant oils are renewable resources for coating applications, are now gaining interest because of the limited supply of petroleum-based products and their long-term effects on the environment[1]. Along with traditional oils such as soybean oil[2], castor oil[3], linseed oil[4], sunflower oil[5], and coconut oil[6], it has been reported that some non-traditional oils such as tobacco seed oil[7], nahar seed oil[8], rubber seed oil[9], karanja oil[10], karawila seed oil[11], jatropha seed oil[12], African locust bean seed oil[13] have been studied as prospective sources for alkyd resins synthesis. It has also been reported that acid oil, a by-product coming from chemical refining of vegetable oil[14], and deodorizer distillate-based fatty acids as a by-product of vegetable oil refineries can also be used to synthesis alkyd resin[15].

The importance of alkyd resins over other high-performance new resins developed in the coating industry is due to the availability of its raw materials, comparatively low raw material cost, low processing cost, biodegradability, durability, adhesion, flexibility, and ease of application.

The properties of the alkyd coating are strongly depending on the properties of vegetable oil. So, knowledge of the vegetable oil properties is essential to develop coating film properties that satisfy the requirements of a particular application. Also, to sustain in the market, the coating industry has required developing alkyd resin with low cost, better properties. All these facts raise the demand for other unconventional oils to replace some of the conventional oils and lowering their price.

Acacia auriculiformis (A. auriculiformis), a member of the family Fabaceae and subfamily Mimosoideae or Mimosaceae[16,17] is a medium-sized, straight, fast growth, evergreen tree normally found in the tropical region in the world where hot and humid conditions are normal.

Regarding the oil content in the seed of Acacia species, it has been reported that the oil content of acacia species varies from 2.1 to 21.3% on a dry weight basis. Most of them have oil content between 7 to 10% (dry wt%)[18]. Chowdhury *et al* and Gunstone *et al*. have reported oil contents were 6.8% and 6 % respectively on a dry weight basis[19,20]. These differences were probably due to genetic factors and the location factor since the chemical composition can vary with the crop varieties, soil, and climatic conditions of the area[21]. But the large volume and ease of availability can overcome the low oil content. It has been also reported that *A auriculiformis* content vernolic acid (cis-12-epoxyoctadeca-cis-9-enoic acid)[19,20,22]. Vernolic acid content epoxy ring in the fatty acid chain has an important impact in the industrial application in a plasticizer and stabilizer used in the polymer[23].

Literature reported that the seed oil of *Acacia auriculiformis* contains mainly linoleic and oleic acids. The oil is rich in PUFAs (polyunsaturated fatty acid) and contains fatty acids having an epoxy ring. An epoxy group provides good adhesion and chemical resistance and high unsaturation provides good air drying and hardness properties, therefore it can be regarded as potential raw material for alkyd resin synthesis with desirable performances.

The objective of this study was to prepare long oil alkyd resin with *Acacia auriculiformis* seed oil (ASO) and to compare its physicochemical and film performance with soybean oil-based long oil alkyd resin.

Experimental Section

Materials

Degummed soybean oil was obtained from Budge Budge Refineries Ltd., West Bengal, India. Akashmoni seeds were obtained from the local market (Kolkata).

Glycerol (99%), pentaerythritol (98%), sorbitol (70%), sodium hydroxide (98% AR), phthalic anhydride (PA) (98%), maleic anhydride (MA) (99%), are used to synthesize the alkyd resins were procured from Loba Chemie Pvt Ltd, Mumbai.

Solvents such as hexane, methanol, ethanol, *n*-Butanol, xylene were of analytical grade and procured from Merck India Ltd., Mumbai, and used as received.

Mineral turpentine oil (MTO), is a mixture of saturated aliphatic and alicyclic hydrocarbons having distillation range from 140-250°C, used as solvent was supplied by M/s Kamal Enterprises, Kolkata.

Driers (6% cobalt octoate, 24% lead octoate, and 10% calcium octoate) (Manufacturer: Patcham FZC) used in this study were procured from M/s Apex Chemicals India Pvt. Ltd., Kolkata, India.

Extraction of ASO from seed

Oil extraction was carried out in Soxhlet extractor using hexane as a solvent after cleaning and drying of the seeds accordingly ISO method 659:2009[24]. At least 10 cycles of extraction were allowed to happen until complete extraction was done. The extracting solvent was distilled off and the extracted oil was quantified gravimetrically.

Removal of phospholipids from the extracted oil

Gums are phosphatides present in the oil. It is required to remove gum from the oil before high-temperature processing or reaction. Degumming of ASO was done by phosphoric acid degumming process[25].

Characterization of SBO and ASO

Physical properties such as the acid value, saponification value, iodine value, moisture content, unsaponification value, refractive index, colour, acid value, specific gravity, etc. of SBO and ASO were determined by AOCS methods[26]. The fatty acid composition of SBO and ASO was determined by gas chromatography by Agilent Gas Chromatograph Model 6890N. The GC was fitted with a DB WAX capillary column (30 m × 0.32 mm × 0.25 µm) and a flame ionization detector. The rate of N_2, H_2, and airflow was kept at 1, 30, and 300 mL/min respectively. The inlet and detector temperatures were kept at 250°C and the oven temperature was programmed to increase from 150°C to 190°C at a rate of 15°C per min then to hold for 5 min and then to increase to 230°C at a rate of 4°C per min, and then to hold for 20 min.

Synthesis of alkyd resin from SBO and ASO

Alkyd resins from SBO and ASO were synthesized by using the monoglyceride method ie; two-stage alcoholysis followed by poly-esterification methods. A five-necked 1000 mL round-bottom flask was equipped with a mechanical stirrer, thermometer, nitrogen gas inlet, condenser with dean stark apparatus to remove the water of reaction azeotropically, and one unused neck to add or collect material between reaction process. The reactor was flushed with nitrogen before charging. Stoichiometric amounts of SBO or ASO (0.04 mol), glycerol (0.016 mol), pentaerythritol (0.0435 mol), 0.05% (w/w)(of the oil) sodium hydroxide were charged into the reactor with continuous stirring. Then the mixture was heated first at 160°C and raised to 260-265°C for 45–60 min until it formed monoglyceride. The first stage alcoholysis reaction was confirmed by complete solubility in methanol (reaction mixture: methanol = 1:3 v/v)[14]. The heating was stopped and the mixture was allowed to cool to 125ºC while stirring. Then the poly-esterification reaction was carried out at 240°C after introducing a stoichiometric amount of acid anhydrides (0.1087 mol) such as PA and MA, a polyol such as sorbitol (0.0108 mol) along with xylene (3%, w/w on raw materials) as azeotropic solvent[9]. The acid value was checked regularly after a 1 hour time interval until the acid value decreased below 16. The resins were cooled to 160-170°C and diluted with MTO to 60% solid content (Table 1).

Characterization of alkyd resins

Physicochemical properties such as specific gravity (ASTMD1475-08), non-volatile matter, and acid value (ASTMD1639-96) of the alkyd resins were determined. Viscosity is a measure of fluid's resistance to flow due to internal friction between two layers. The viscosity of both resins was measured by Brookfield viscometer as well as Gardner bubble tube viscometer. For the Brookfield viscometer, viscosity is determined by rotating a spindle attached to the viscometer in the liquid sample. The measured torque against the rotating spindle is transformed into viscosity units.

For the BYK-Gardner bubble viscometer, the kinematic viscosity of known liquids such as resins and varnishes are determined by comparing the time required to travel by air bubble through a specific tube filled by test sample against the standard sample. The viscosity of the liquid is directly proportional to the time taken by an air bubble to rise through the tube, which means the quick rise of the bubble indicates low viscosity. The sample tube is filled with liquid up to the mark specified, insert a cork, and then choose four standard tubes of nearest viscosity identified by "alphabet" and turn over them. Compare the rising time of the bubble in the sample with standard tubes which is closest. Calculate viscosity from the given viscosity of the standards data table.

Structural analysis by Fourier Transfer Infrared Spectroscopy (FTIR)

The main component of alkyd resin can be identified from the infrared spectrum. FTIR spectra of each of the alkyd samples were obtained using Agilent Cary 630 FTIR Spectrophotometer using ATR sampling accessory with 400–4000 cm^{-1}, taking acetone as solvent.

Structural analysis by ^{1}H NMR Spectroscopy

^{1}HNMR spectra of both alkyd resins were recorded in $CDCl_3$ at 300 MHz using Bruker Advance II 400 NMR spectrometer.

Preparation of dry films

Film properties were evaluated by making a 100g varnish by 86% resin, 3.225% driers (0.05% of 6% cobalt octoate, 0.5% of 24% lead octoate, and 0.2% of 6% calcium octoate based on metal content), and rest is made up by MTO and 0.2% of methyl ethyl ketoxime was added as an anti-skinning agent. After 5 min of continuous mixing, the resins were uniformly coated in the form of thin film on MS panel and cured at ambient conditions. After seven days of drying, film performance properties were examined.

Performance characteristics of the alkyd resins

The drying properties of alkyd resins were determined using the standard method as reported by Chiplunkar *et al.* 2016[14]. For both cases of alkyd, the tests were carried out under similar conditions.

After maturing the film for 7 days at ambient temperature the film performance of both alkyd films was measured in terms of physical and chemical resistance using different chemicals like acid, alkali, and water as per methods as reported by Momodu *et al.* 2011[27]. Visual observations like flow or leveling of clear, gloss were also observed. Pencil hardness was tested as per ASTM D3363-05, Adhesion was tested by using a cross-hatch cutter according to ASTM3359-09, and flexibility (ASTM D522-08) was tested.

Results and Discussion

Oil content of seed

The oil content of *A. auriculiformis* is found 7.1% on dry weight basis. The result is similar to the report showed in previous studies[18,19].

The colour of the oil found is reddish yellow.

Physico-chemical properties of SBO and ASO

The comparison of the physicochemical properties of SBO and ASO are shown in Table 2.

It is seen from Table 2 that the physico-chemical properties of ASO are quite similar to SBO except for the acid value which has a negligible impact on alkyd synthesis.

Table 1 — Compositions (in mol) of synthesized alkyd resins

Resins	SBO	ASO	PE	Glycerine	Sorbitol	PA	MA
SBO alkyd	0.04	-	0.0435	0.016	0.0108	0.106	0.0027
ASO alkyd	-	0.04	0.0435	0.016	0.0108	0.106	0.0027

Both oils have the same sap value indicating their comparable molecular weight and fatty acid chain length.

Iodine value indicates the degree of unsaturation present in the fatty acid part of oils. Iodine value is measured to predict the drying property of oils. ASO with an iodine value of 128.1 can be classified as semi-drying oil according to the classification of oil based on iodine value. It can be useful in surface coating applications like paints, resins, and printing inks. The iodine value of ASO compares with Soybean oils and also safflower, sunflower oils (with iodine values ranging between 112-148)[28]. In line with the iodine values, it can be used for the production of alkyd resin and in paint formulation.

The fatty acid composition of the ASO and SBO was determined by gas chromatography. The result indicates that both oils were mainly composed of unsaturated and saturated fatty acids (Table 3). Among the unsaturated fatty acid, linoleic acid and oleic acid were the most abundant while among saturated fatty acids, stearic acid and palmitic acid were the most abundant acids. The characteristic difference of ASO is the presence of vernolic acid (special fatty acid contains epoxy ring) which is absent in SBO also with high linoleic acid content than soybean oil. The result also supports previous studies[18,19].

Table 2 — Physicochemical properties of Soybean oil (SBO) and Akashmoni seed oil (ASO)

Properties	SBO	ASO
Acid value	1.10	5.77
Saponification value	193.28	193.8
Iodine value (g I_2 per 100 g)	126.6	128.1
Unsaponifiable matter (%)	1.05	1.5
Colour	Yellow	Reddish Yellow
Free fatty acid (% as oleic acid)	0.525	2.89
Refractive Index	1.466	1.461
Specific gravity	0.905	0.89

Table 3 — Fatty acid composition of Akashmoni seed oil (ASO) and soybean oil (SBO)

Type of fatty acid	Akashmoni seed oil (%, w/w)	Soybean oil (%, w/w)
Myristic acid, C14:0	1.13	-
Palmitic acid, C16:0	7.50	10.42
Stearic acid, C18:0	1.13	5.00
Oleic acid, C18:1	11.75	25.60
Linoleic acid, C18:2	71.71	51.51
Linolenic acid, C18:3	0.59	7.33
Vernolic acid, C18:1; 12 epoxy	5.04	-
Others	1.15	0.14

Both oils have a higher amount of unsaturated fatty acids (83%) and have iodine value greater than non-drying oil and therefore they are called semi-drying oil[29]. ASO has higher linoleic acid content than SBO. Due to high unsaturation, ASO can also be used for the synthesis of alkyd resin which must have good drying properties for coating application.

Synthesis of alkyd resins from SBO and ASO

Alkyd resins were produced by alcoholysis followed by a poly-esterification reaction. In this reaction, SBO or ASO reacted with glycerol and pentaerythritol (PE) at 260-265°C in the presence of sodium hydroxide as a catalyst. As the reaction proceed more monoglyceride is formed which increase the polarity of the reaction mixture. Based on this property completion of this step was identified by solubility in methanol (1:3 Sample: methanol by volume). The poly esterification reaction was then carried out at 220-240°C with PA and MA. Continuous evaporation of Xylene along with water as an azeotropic mixture indicates the progress of the reaction. The acid value of the reaction mixture was measured at 0.5 hr intervals of time for both the alkyds (Fig. 1). The reactions were carried out until the acid value was decreased below 16.

Variation of acid value with time

In both cases, the acid value decreases nearly at the same rate because of using the same anhydride which strongly influenced the rate of decrease of acid value[9]

It has been reported that the primary hydroxyl groups of the monoglyceride react more rapidly than the secondary hydroxyl groups with carboxyl groups of poly-basic acid[30]. Thus, the initial rapid decrease in acid value observed for all the alkyd samples may be due to rapid esterification with the primary hydroxyl group.

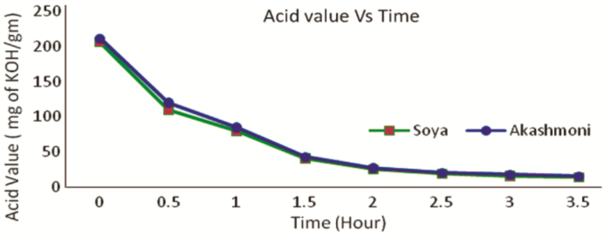

Fig. 1 — Plot of acid value vs time for (a) SBO and (b) ASO based alkyd

Amount of water removed vs time

Water was formed during esterification reaction initially at a high rate followed by a reduced rate as reaction procced. The reason was the high initial esterification rate, and also the presence of water in sorbitol which was used as raw material (Fig. 2). Similar results were reported by Oladipo *et al*. and Musa *et al*.[31,32].

Physico-chemical properties of alkydresins

The comparison between physic chemical properties of the two alkyd resins is shown in Table 4. The final acid value of SBO and ASO alkyds are maintained at 14.49 and 15.45 respectively(<16). The acid values obtained from the alkyd resins are in line with most of those obtained in the literature survey (acid value 14-16). An important parameter of alkyd is acid value which determines the chemical resistance of alkyd resin to various chemicals such as brine (NaCl), acid (H_2SO_4), alkali (NaOH), and water. Alkyd resins having low acid value are promising in formulating coatings like nitrocellulose lacquers for metal cans used in packaging foods, beverages, and drugs[9].

The colour of ASO alkyd is darker than SBO alkyd. This result is due to the initial dark colour of ASO. Both alkyds are darker than that of the respective precursor oil. The darkening in colour of the alkyd could be due to high temperatures of reaction, oxidation, and the catalyst[27].

The viscosity of alkyd resin that is friction between two adjacent liquid layers depends on hydrogen bonding, geometrical configuration, and close packing of resin molecule[11]. The viscosity of ASO alkyd at 30°C is higher than SBO alkyd. This could be a result of polymerization reactions at the reactive double bonds of unsaturation of the oil and increased molecular weights during alkyd synthesis.

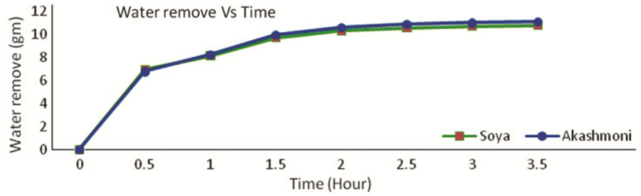

Fig. 2 — Plot of water remove vs time for (a) SBO and (b) ASO based alkyd

Table 4 — Physico-chemical properties of SBO and ASO alkyd

Properties	SBO alkyd	ASO alkyd
Viscosity in Brookfield@30°C	1195 cp	1887 cp
Viscosity in bubble tube@30°C	"X"	"Y"
Specific gravity@30°C	0.949	0.942
A.V	14.49	15.45
Sap. Value	295.4	286.7
% NVM	59.5	59.64

The specific gravity of the test alkyd resins at 30°C was observed to be relatively high compared to their corresponding values obtained in the seed oils. This could be a result of increased molecular weights during alkyd synthesis due to polymerization.

Structural analysis

The functional groups of SBO and ASO-based alkyd resins were confirmed by ATR-FTIR spectroscopy. The FTIR spectra of both resins have similar characteristics. The peak at 741, 745 cm^{-1} corresponds to the rocking vibration of the methylene group. Characteristics peak at 902 cm^{-1} found in ASO is for the epoxy group present in fatty acid which is absent in SBO. All peaks between 1073-1267 cm^{-1} confirm C-O-C stretching of ether. For SBO alkyd 1379 & 1464 cm^{-1} and for ASO alkyd 1360 & 1461 indicates symmetric and asymmetric bending vibration of C−H. The peak for aromatic C=C stretch appears at 1580 cm^{-1} for both resins. A sharp peak at 1736 and 1722 cm^{-1} indicates stretching of >C=O of the ester group. The peaks around 2855-2926 cm^{-1} correspond to an aliphatic C−H stretching. C−H stretching for alkene appears at 3011 cm^{-1} for both resins. The broad peak around 3526-3537 cm^{-1} confirms the presence of the −OH group in resin Table 5. The spectral data confirm the presence of hydroxyl group, ester group, olefinic double bonds, epoxy group in the alkyd resins[14,23].

^{1}H NMR spectra of the ASO and SBO alkyd also have similar characteristic peaks. Characteristic peaks at 0.833-1.005 ppm correspond to the protons of the terminal methyl group. Peaks at 1.13-1.306 ppm are for the protons of all the internal -CH_2 groups present in the fatty acid chains. For the protons of CH_2 group attached next to the above terminal methyl group, the peaks appear at 1.54-1.61 ppm. Characteristic peaks at 2.055-2.1 ppm are for allylic protons of –CH_2,

Table 5 — Characteristic peaks in FTIR spectra of alkyd resins

Wavenumber (cm^{-1})	Functionality
741, 745	Methylene rocking vibration
902	Epoxy group
1073-1267	C−O−C stretching of ether
1360, 1379, 1461, 1464	Symmetric asymmetric bending vibration of C−H
1580	C=C stretch of aromatic and alkene band
1722, 1736	C=O stretching of ester
2855, 2926	Aliphatic C-H stretching
3011	C−H stretching for alkene
3526- 3537	-O−H stretching

Table 6 — Characteristic peaks in ^{1}H NMR spectra of alkyd resins

Chemical shift (ppm)	Functionality
0.833–1.005	Terminal methyl group -CH_3
1.13–1.306	Internal -CH_2- of fatty acid chain
1.54–1.61	-$C(H)_2$- next to - CH_3
2.055–2.1	Allylic proton of -CH_2
2.19–2.35	α proton of Ester -HC–COOR
2.5 – 4.5	Protons of epoxy ring
5.361–5.386	-HC=CH- protons
7.24–7.28	Aromatic ring proton

at 2.19-2.35 ppm for α protons of ester groups, and at 2.5-4.5 ppm for protons in the epoxy ring comes from vernolic acid.

The peaks for protons of the –CH= CH moiety appears at 5.361-5.386 ppm. ^{1}H NMR spectra of the resins show characteristic peaks at 7.02-7.09 ppm are for the protons for CH of glycerol moiety. The peaks at 7.24-7.28 are observed for aromatic protons of PA present in resins (Table 6).

Film properties of alkyd resins

The resins were then cured by using driers such as cobalt octoate(6%)as primary drier, lead octoate(24%) as through drier and calcium octoate (10%) as an auxiliary drier to improve the oxidation of fatty acids through hydroperoxide decomposition and crosslinking between fatty acid chains.

In the alkyd coating films, unsaturation present in fatty acids, and MA undergo aerial oxidation, cross-linked with each other via hydroperoxide formation and decomposition, catalyzed by the metal driers in a complex process of radical polymerization[33]. Cross-linking occurs by free radical mechanism due to the unsaturation present in fatty acids and MA[34]. From the drying time measurement, it is observed that the drying time of ASO alkyd resin is shorter because of the higher drying index of ASO than SBO as ASO has high linoleic acid content.

Hardness occurs due to the chain flexibility and degree of crosslinking of the network. It depends on the type of substrate, the structural orientation of the monomer used for synthesis[35]. As both resins have the same kind of raw material used, their hardness was found comparable.

Alkyd resins have good adherent properties due to the presence of polar ester bonds. Table 7 shows that both resins have passed the adhesion test.

Gloss is an important physical property of coating film, which results from the interaction between light and the surface of the coating film. Gloss is found better for ASO-based alkyd resin.

Table 7 — Physical properties of film produced by alkyd resins

	Alkyd	
Film properties	SBO alkyd	ASO alkyd
Dry to touch	230 mins	200 mins
Pencil hardness	‘H’ pass	‘H’ pass
Bend test	Pass	Pass
Adhesion	5B	5B
Gloss visual	Inferior to ASO	Better than SBO

Table 8 — Chemical properties of film produced by alkyd resins

Chemical properties	SBO alkyd	ASO alkyd
Alkali resistance	Fail	Fail
Acid resistance	Good	Comparable to SBO
Water resistance	Good	Comparable to SBO

The film performance of resins under different chemical environments is listed in Table 8. The chemical resistance properties of both alkyd resins were comparable. Alkyd films of both resins are highly resistant to acid, water, but poor to alkali due to the presence of the ester group which is hydrolyzable in presence of alkali[36].

Conclusion

From the above study, it can be concluded that alkyd resin based on ASO with MA and PA as dibasic acid and glycerol, pentaerythritol, sorbitol as a polyol, have been successfully synthesized. It was observed that the resins acquire satisfying film properties such as drying time, gloss, hardness, adhesion and chemical resistance, etc., which may be suitable for surface coating and as a binder for composite, etc. The performance of the ASO-based alkyd resin was found to be comparable to SBO-based alkyd resins and in some cases, it is better like gloss and drying rate. *A. auriculiformis* is a fast-growing evergreen tree, having a life span of up to 30 years, found in tropical regions of the World. Though oil content is low but its capability to grow on a wide range of soil and its seed production rate could overcome the low oil content. Also, the seed of this tree is a by-product till today, which will make it cheap as it is a major source of wood.

Investigated research revealed that ASO, a minor seed oil can be utilized as a potential source of raw material for the surface coating industry like other vegetable oils. or it could be a promising alternative to soybean oil.

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