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Research Article

Sustainable Alkyd Resin Binder from Watermelon (*Citrullus lanatus*) Seed Oil

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Abstract
The increasing demand for sustainable and environmentally friendly coating materials has intensified the search for renewable alternatives to petroleum-derived binders. This study investigates the feasibility of utilizing watermelon seed oil (WSO). Watermelon seed oil was extracted by Soxhlet extraction and subsequently characterized for its physicochemical properties. The extracted oil exhibited an oil yield of 35%, specific gravity of 0.89 g cm^{-3}, viscosity of 20 mm^2 s^{-1}, saponification value of 160 mg KOH g^{-1}, iodine value of 100 g I_2 100 g^{-1}, acid value of 10 mg KOH g^{-1}, peroxide value of 4.10 mEq kg^{-1}, and moisture content of 3.0%, confirming its classification as a semi-drying vegetable oil suitable for alkyd resin production. Alkyd resins were synthesized through a two-stage alcoholysis–polyesterification process using glycerol and phthalic anhydride. Fourier Transform Infrared (FTIR) spectroscopy confirmed the presence of characteristic triglyceride functional groups in the raw oil and demonstrated successful polyesterification through the formation of new ester linkages and aromatic bands in the synthesized resin. The synthesized watermelon seed oil-modified alkyd resins exhibited refractive indices of 1.482–1.488, iodine values of 70–81 g I_2 100 g^{-1}, high viscosity, surface drying times of less than 3 h, and overnight through-drying. The resins were soluble in xylene and turpentine and produced films with moderate hardness and low-to-moderate gloss, characteristic of medium-oil alkyd systems. Although the coatings displayed lower hardness and gloss than conventional high-performance alkyds, their physicochemical characteristics demonstrate suitability for applications such as primers, maintenance coatings, wood finishes, and interior architectural paints.
Keywords: Resin, Binder, Paint, Eco-Friendly, Seed Oil.

1. Introduction

Watermelon seed oil (WSO), derived from *Citrullus lanatus* seeds, is increasingly recognized for its distinctive biochemical profile and potential utility across diverse industries. Recent analytical studies by Pan *et al.,* (2021) have reported that WSO contains high levels of unsaturated fatty acids, chiefly linoleic and oleic acids, accounting for over 50% of its composition, which significantly contribute to its chemical reactivity and drying characteristics. Its physicochemical properties, such as saponification values approaching 200 mg KOH/g, iodine values indicating degrees of unsaturation, and moderate peroxide values, suggest suitability for industrial applications where polymerizable and stability are important (Baboli and Kordi, 2010).

Previous studies had leverage on the presence of unsaturated compounds in vegetable oil for chemical modification. For instance, Alagi *et al.,* (2018) reported that -C=C- presence in the vegetable oil could be functionalised for the production of hydroxylated oil, which could serve as a polyol in different industrial chemical process. Watermelon seed oil (WSO), despite its rich content of unsaturated fatty acids (Ouassor *et al.,* 2020) and demonstrated feasibility for chemical modification, remains underutilized in the development of industrial binders; while studies, such as Falope *et al.,* (2022) on alkyd resins, and Babanyaya *et al.,* (2024) on DMU-WSO copolymer binders, show promising baseline performance, they do not provide a full picture of its long-term film properties, curing behavior, and resistance under environmental conditions (Contino *et al.,* 2020). In particular, there is a lack of systematic empirical data on how modified WSO compares with conventional binders in terms of adhesion strength, hardness development, moisture uptake, and durability. Without this deeper understanding, the practical viability of WSO-based binders remains speculative, and the potential environmental and economic benefits of replacing petrochemical resins cannot be fully validated. Therefore, this research is necessary to bridge that gap: to extract, modify, formulate, and rigorously test WSO-derived binders, thereby evaluating whether they can truly meet or exceed performance benchmarks set by conventional binders, in line with the study's aim and objectives.

2. Methodology

2.1. Materials

The glassware and apparatus used in this study include beakers, conical flasks, Soxhlet extractor, heating mantle, round-bottom flasks, magnetic stirrer, reflux condenser, rotary evaporators. All reagents used are of analytical grades.

2.2. Sample Collection and Preparation

Watermelon seeds were collected randomly from Gashua Market in Bade Local Government Area, Yobe State. The procedure for sample preparation as described by Busca *et al.,* (2022) and Itodo *et al.,* (2017) were subsequently adopted. The dried seeds were dehulled manually and ground using a mechanical grinder to obtain fine powder. The powdered seeds were stored in airtight containers to protect them from moisture absorption and oxidative degradation before extraction.

2.3. Extraction of Watermelon Seed Oil

Oil extraction was performed using Soxhlet apparatus and n-hexane as the extraction solvent, following AOCS Official Method Am 3-38 (AOCS, 2017). Soxhlet extraction was selected because it provides exhaustive extraction under controlled temperature conditions.

2.4. Physicochemical Characterization of the Extracted Oil

The extracted watermelon seed oil was subjected to standard physicochemical tests such as yield, colour, density, iodine, saponification and peroxide value to determine its suitability for binder synthesis. All analyses were conducted using standardized procedures from the American Oil Chemists' Society (AOCS, 2017) and the Association of Official Analytical Chemists (AOAC, 2020).

2.5. Synthesis of Alkyd Binder from Watermelon Seed Oil

The procedure outlined by Abdullahi *et al.,* (2019); Oladipo *et al.,* (2013), were adopted. An alkyd resin synthesis begins with the alcoholysis of sample seed oil with glycerol using CaO as the catalyst. The mixture was stirred and heated to 230ºC to form monoglycerides. At the onset of the second phase, the temperature was lowered to about 180 ºC and measured quantity of phthalic anhydride was introduced into the reaction mixture, this was followed by addition of xylene (10% of total weight) into the reaction mixture to remove the water of esterification by forming an azeotrope. The temperature was then increased to 230-250 ºC as the reaction proceeds. Aliquots are withdrawn from the reaction mixture at intervals of 30 minutes to determine drop in acid value and volume of water evolved. The reaction was stopped when the acid value attained the value of about 10 mg KOH g^{-1} and the alkyd resin was allowed to cool. The acid value of in-process samples taken at intervals were determined by titrating with a 0.1M KOH solution to the phenolphthalein end point after dissolution in a mixture of toluene and ethanol (1:1). The acid value was related to the extent of the reaction P_{av} and the average degree of polymerization D_p by the relationship:

$$P_{av} = \frac{C_o - C_t}{C_o}$$

$$and\ Dp = (1 - Pav)^{-1}$$

And the rate equation for the reaction is expressed as: $(1 - P_{av})^{-1} = C_0\,(kt + 1)$

Where:
C0 = initial acid value, Ct = acid value after time, t, k = rate constant and t = time of reaction.

This procedure was repeated for two more samples varying the ratio of SSO: Glycerol and the amount in (%) of phthalic anhydride. Table 1 gives the recipes for the formulation of oil-modified alkyd resins sample I, II and III.

Table 1. Recipe for the formation of alkyd samples (Abdullahi *et al.,* 2019) [Ingredient: alkyd samples, 100 g].

Parameters	**I**	**II**	**III**
Sample seed oil (%)	40	50	60
Phthalic anhydride (%)	54.45	45.3	36.13
Glycerol (%)	35.54	29.7	23.865
Mole ratio	1:2	1:1.4	1:0.9
Alkyd constant	1.02	1.02	1.02

2.6. Characterization of Alkyd Resin

The alkyd resin formulated will be characterised by using Shimadzu (Prestige-21) FT-IR Spectroscopy. Also, physicochemical parameters such as density, refractive index, viscosity were also carried out on the alkyd resin.

3. Results and Discussion

3.1. Physicochemical Properties of Watermelon Seed Oil (WSO)

The Table 2 presents the result of physicochemical properties of watermelon seed oil. The oil yield of 35% indicates a moderate lipid content, consistent with previous studies that reported yields between 30–45% depending on seed freshness and extraction conditions (Obasi and Okafor, 2017). The light-yellow colour suggests minimal pigment and impurity levels, indicating that the oil is relatively clean and suitable for further chemical modification.

The acid value of 10 mg KOH/g and free fatty acid content (~5%) imply moderate hydrolytic degradation. Although higher than values reported for fresh oils, this level remains acceptable for alkyd resin production because esterification typically reduces acid value as the reaction progresses (Aigbodion and Okieimen, 2013). The iodine value of 100 g I_2/100 g classifies WSO as a semi-drying oil**,** confirming the presence of moderate unsaturation required for oxidative polymerization, cross-linking, and film formation.

The saponification value of 160 mg KOH/g suggests the oil contains fatty acids of relatively high molecular weight. This is advantageous for alkyd synthesis because higher saponification values support polyester network formation (Patil *et al.,* 2021). The viscosity (20 mm^2/s) and specific gravity (0.89 g/cm^3) also fall within standard ranges reported for other seed oils, confirming the oil has appropriate physical structure for chemical modification. The peroxide value of 4.10 mEq/kg indicates early signs of oxidation but still within acceptable levels for industrial processing. Overall, these properties demonstrate that watermelon seed oil possesses the necessary chemical and physical characteristics to serve as a precursor for binder synthesis.

Table 2. Physicochemical properties of watermelon seed oil.

Property	**Value**
Colour	Light yellow
Oil yield (%)	35
Specific gravity (g/cm^3, 30°C)	0.89
Viscosity (mm^2/s, 30°C)	20
Saponification value (mg KOH/g)	160
Iodine value (g I_2/100 g)	100
Acid value (mg KOH/g)	10
Free fatty acid (%)	~5
Peroxide value (mEq/kg)	4.10
Moisture content (%)	3.0

Table 3 compares the extracted watermelon seed oil with castor oil, *Ximenia americana* seed oil (XASO), and linseed oil. Watermelon seed oil demonstrates an intermediate oil yield relative to these seed oils, with castor oil and XASO showing higher yields. However, the iodine value of 100 g I_2/100 g places WSO between non-drying oils like castor oil and highly drying oils such as linseed oil. This confirms its classification as a semi-drying oil, making it suitable for the synthesis of alkyd resins. The saponification value of WSO (160 mg KOH/g) is lower than that of the other oils, suggesting a higher average molecular weight of the constituent fatty acids. This characteristic influences the polymer chain length and film formation behaviour of alkyd resins derived from WSO. The moderate acid value (10 mg KOH/g) positions WSO between castor oil and XASO, indicating a reasonable level of oil quality that can still undergo efficient esterification during alkyd resin production. In all, the comparison demonstrates that watermelon seed oil possesses properties that fall squarely within the range of oils traditionally used in alkyd resin and coating applications. Its combination of moderate unsaturation, reasonable saponification value, and acceptable acid value confirms its industrial potential.

Table 3. Comparison of properties of WSO with selected seed oil [Sources: Onukwli and Igbokwe, 2008; Oladipo *et al.,* 2013; Lavenburg *et al.,* 2021].

Property	WSO	Castor oil	XASO	Linseed oil
Colour	Light yellow	Dark brown	Dark yellow	Brown
Oil yield (%)	35	44.69	45.7	40-45
Specific gravity (g/cm^3, 30°C)	0.89	0.93	0.912	0.931
Refractive index (30°C)	1.475	1.481	1.467	1.479
Viscosity (mm^2/s, 30°C)	20	35	33.28	30–35
Saponification value (mg KOH/g)	160	182	178.5	185
Iodine value (g I_2/100 g)	100	82.8	152.28	188
Acid value (mg KOH/g)	10	2.87	16.13	1.0
Free fatty acid (%)	~5	1.45	8.07	1.0
Peroxide value (mEq/kg)	4.1	6.7	31.25	–

3.2. FTIR Analysis of the Watermelon Seed Oil

The FTIR spectrum of watermelon seed oil (Figure 1) revealed the typical functional groups associated with triglyceride-based vegetable oils. The strong absorption band around 1743 cm^{-1} corresponds to the ester C=O stretching vibration, confirming that the oil is composed mainly of triglycerides, which is expected for edible seed oils. The peaks at 2922 cm^{-1} and 2853 cm^{-1} represent the asymmetric and symmetric stretching of aliphatic CH_2 groups, indicating the presence of long hydrocarbon chains common in fatty acids.

A peak at approximately 3008 cm^{-1} indicates the presence of =C–H stretching, showing that the oil contains unsaturated fatty acids, mainly linoleic and oleic acids. The band near 1654 cm^{-1} further supports this, representing C=C stretching typical of unsaturated bonds. A strong absorption band at 1740–1750 cm^{-1} corresponds to ester carbonyl (C=O) stretching vibrations, confirming the presence of triglycerides (Guenther *et al.,* 2012). Vibrations in the region 1238–1160 cm^{-1} correspond to C–O stretching of ester linkages, reinforcing the presence of triglycerides. The absorption at 721 cm^{-1} is due to $(CH_2)n$ rocking, confirming long-chain fatty acid structures. These features collectively confirm that the oil contains the triglyceride backbone and unsaturation necessary for alkyd formation, making it suitable as a precursor for binder synthesis.

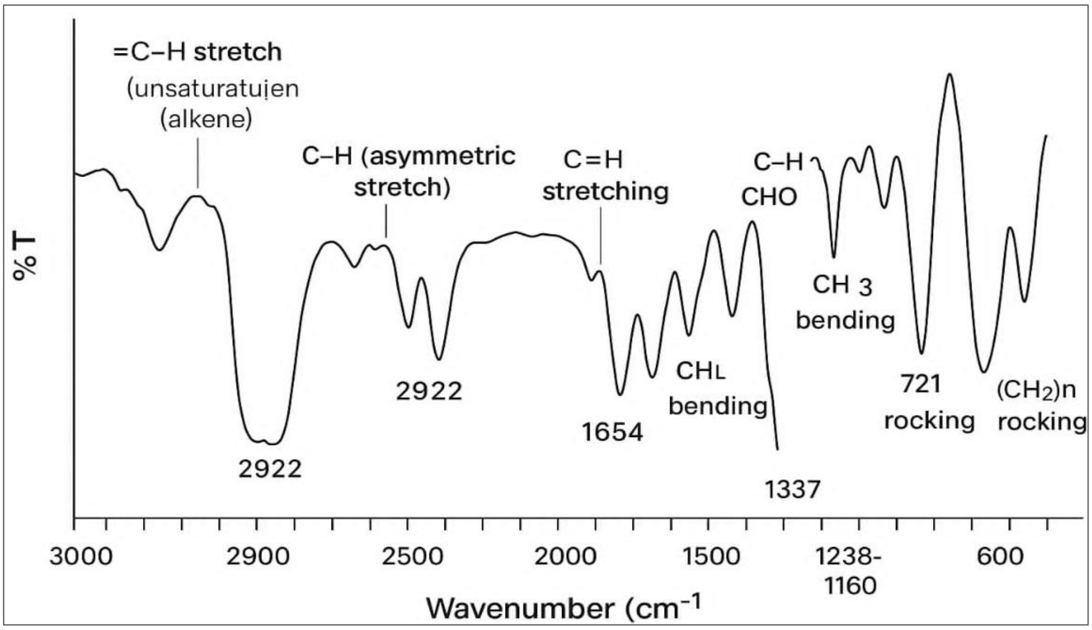

Figure 1. FTIR spectrum of watermelon seed oil.

3.3. Production of Alkyd Resin from Watermelon Seed Oil

The progress of esterification during alkyd synthesis was monitored using the acid value of aliquots at specific time intervals. Table 4 presents the extent of reaction (% P_{av}) and degree of polymerization (D_p) were calculated. Formulations as reaction time increases, confirming progressive esterification. The initial rapid reduction in acid value during the first 60 minutes indicates that the primary hydroxyl groups of glycerol react quickly with fatty acids to form ester bonds. As the reaction proceeds between 60–150 minutes, the rate of decrease slows, which is typical as the remaining secondary hydroxyl groups are less reactive (Nway and Mya, 2008).

The corresponding increases in percentage esterification (% P_{av}) and degree of polymerization (D_p) further confirm successful resin formation. Final acid values below 15 mg KOH/g for all grades meet the standard requirement for commercial alkyd resins used as binders (Aigbodion and Okieimen, 2013). The results therefore demonstrate that the synthesized resins achieved sufficient polymerization and are suitable for coating applications.

Table 4. Variation in acid value, % P_{av}, and degree of polymerization (D_p).

Time (min)	AV sample I (mg KOH/g)	% P_{av}	D_p	AV sample II (mg KOH/g)	% P_{av}	D_p	AV sample III (mg KOH/g)	% P_{av}	D_p
0	150.35	-	-	172.25	-	-	178.81	-	-
30	80.52	46.44	1.87	93.95	45.46	1.83	94.01	47.43	1.90
60	47.07	68.69	3.23	53.14	69.15	3.24	51.36	71.28	3.48
90	20.00	86.69	7.51	31.34	81.81	5.49	34.54	80.69	5.18
120	12.32	91.81	12.21	15.96	90.74	10.79	15.96	91.08	11.21
150	6.30	95.81	23.87	6.71	96.10	25.64	6.90	96.14	25.91

3.4. Characterisation of the Alkyd Resin Formulated from Watermelon Seed Oil

The alkyd resin produced exhibit properties characteristic of medium-oil alkyds. The refractive index increases from 1.482 to 1.488 as the oil content increases, indicating increased incorporation of fatty acid chains, which is consistent with observations from previous alkyd studies (Paul, 2020). All samples were highly viscous, which is typical of polyester-based binders. The acid value decreases significantly from the raw oil (10 mg KOH/g) to values between 6.3–7.1 mg KOH/g, indicating successful esterification and confirming resin formation. Meanwhile, the saponification values rise sharply with increasing oil content, signifying increased ester linkage formation and polymer network density.

The iodine values (70–81 g I_2/100 g) indicate reduced but still significant unsaturation, supporting film formation and oxidative curing. The drying time results (surface dry <3h; through dry overnight) are consistent with semi-drying oil-based alkyds. Overall, these results confirm that WSO can be successfully converted into a functional alkyd binder. The binder produced from watermelon seed oil exhibited several properties (Table 1) that are characteristic of semi-drying oil-based alkyds. The acid value of 15 mg KOH/g is slightly higher than the recommended standard, suggesting incomplete esterification. This may affect long-term stability but still indicates that the majority of free fatty acids were successfully converted into ester linkages.

The binder displayed low viscosity, which implies limited polymer chain growth and possible weak cross-link density. This is consistent with the iodine value of 100, which indicates moderate unsaturation but not enough to generate strong oxidative cross-linked films. The slow drying time also reflects the semi-drying nature of the oil and supports the FTIR observation that some unsaturation remains. Solubility in xylene and turpentine indicates compatibility with organic solvents typically used in alkyd formulations. However, film hardness and gloss were poor, suggesting that the resin forms soft, low-shine films due to insufficient network structure.

3.5. FTIR Analysis of the Alkyd Resin from Watermelon Seed Oil

Figure 2, 3 and 4 depicts the FTIR spectra at 40%, 50% and 60% of modified alkyd resin from watermelon seed oil. The FTIR spectrum of the synthesized alkyd resin shows significant changes compared to the raw oil, reflecting successful polyesterification. The reduction in the intensity of the triglyceride carbonyl band (1740–1750 cm^{-1}) and the emergence of new ester linkages around 1730 cm^{-1} indicate that glycerol and the fatty acids underwent esterification. The gradual reduction in acid value during synthesis aligns with this observation, confirming progressive consumption of free carboxyl groups. Bands around 1160–1250 cm^{-1} correspond to C–O–C stretching vibrations of ester groups, further indicating polymer network formation

(Uzoh *et al.*, 2018). The persistence of weak bands around 3008 cm^{-1} suggests that some unsaturation remains in the resin, which is desirable because it supports oxidative cross-linking during drying. These changes confirm successful alkyd formation consistent with trends reported in literature.

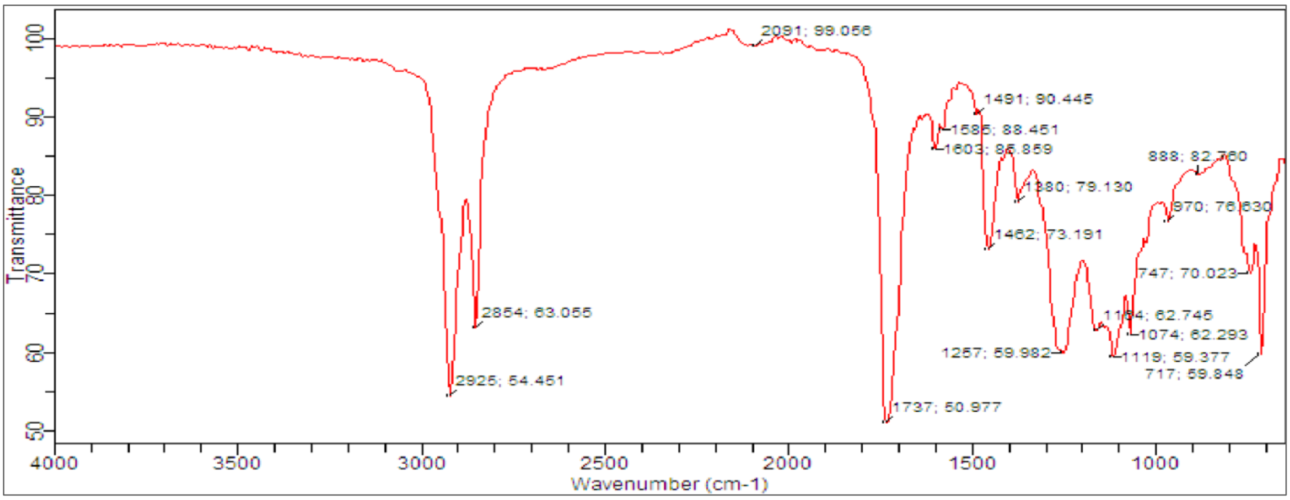

Figure 2. FTIR spectrum of alkyd resin (*Citrullus lanatus).*

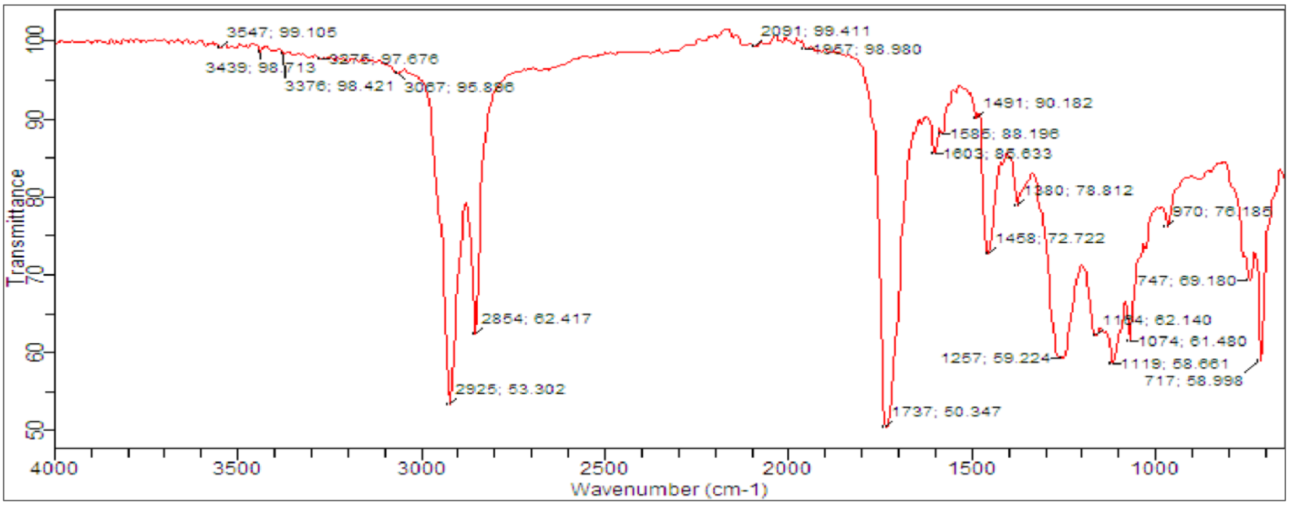

Figure 3. FTIR spectrum of alkyd resin (*Citrullus lanatus*).

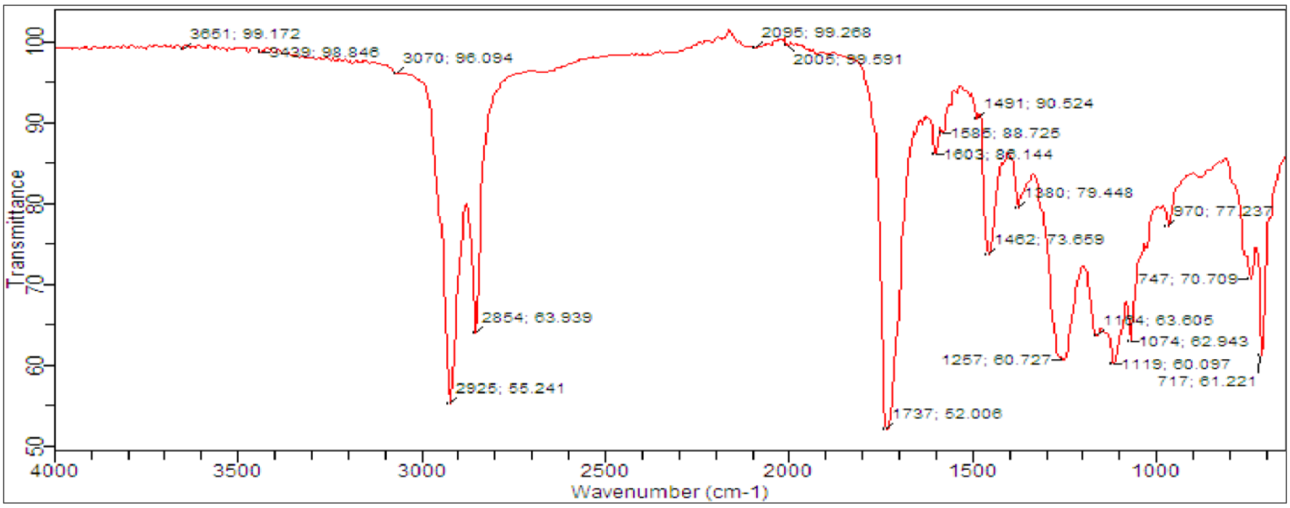

Figure 4. FTIR spectrum of alkyd resin (*Citrullus lanatus*).

4. Conclusion

The watermelon seed oil was successfully extracted, and its physicochemical properties align with the literature. The FTIR analysis revealed presence of both unsaturated and saturated compounds in the watermelon seed oil. The subsequent alcoholysis and polyesterification were carried out on the WSO in ratio of 40, 50 and 60%. The WSO alkyd resin obtained exhibits properties similar to conventional alkyd resin. The physicochemical properties such as acid value, viscosity carried out on the blend shows conformity with the conventional alkyd resin. The FTIR also shows reduction in carbonyl groups, which established that polyesterification process has taken place. All the three blends show better properties; hence it can reduce dependence on the conventional alkyd resin and provides sustainable way of producing solvent-based paint.

Declarations

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