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Research Article

Modification of Polyvinyl Acetate with Hydroxylated Avocado Seed Oil as a Copolymer Binder for Possible Application in Coating Industry

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Abstract: The study explored the modification of conventional polyvinyl acetate (PVAc) with hydroxylated oil for possible application in coating industry. Avocado seed oil was extracted, epoxidised and hydroxylated. The hydroxylated avocado seed oil (HASO) was blended with conventional polyvinyl acetate in different ratio of 10 to 60% of hydroxylated oil, to develop HASO/PVAc copolymer binder. The different ratio of blend is to ensure that no other properties of copolymer is jeopardized, at the expense of another, since polymer tends to have a molecular weight in which each property exhibit optimum value. The important physical parameters of the copolymer resin (HASO/PVAc) at different composition ratio were investigated.

The physical properties such as viscosity; density, turbidity, refractive index, gel time, moisture uptake, water solubility, elongation at break, and melting point were investigated. FT-IR analysis of copolymer resin (HASO/PVAc) showed chemical interaction between PVAc and HASO. Furthermore, moisture uptake, density, turbidity, viscosity and melting point are found to be decreased, as the concentration of HASO in the copolymer matrix increases, while elongation at break, water solubility, gel time, and refractive index increase with increase in concentration of HASO in copolymer matrix. The results shows improvement in flexibility, moisture absorptivity, and glossitivity, which are major setback of conventional PVAc.

Keywords: Extraction, Epoxidation, Hydroxylation, Copolymerisation, Polyvinyl acetate (PVAc).

I. Introduction

Paint is one of the oldest synthetic substances known, with a history stretching back into prehistoric times. It transcends from being a tools of covering to reflect the inner beauty and belief of individual. Painting and coatings are unique human activities that have helped the human race to contribute significantly in earning a better livelihood, building a better and more beautified world for ourselves. (Akinterinwa *et al.*, 2015; Osemeahon *et al.*, 2018).

Paint is any liquid, liquefiable, or mastic composition that after application to a substrate in a thin layer, converts to a solid film. It is most commonly used to protect, colour, or provide texture to objects. It is also the term used to describe a number of substances that consist of a pigment suspended in a liquid or paste vehicle such as oil or water with a brush, a roller or a spray gun. Paint is applied in a thin coat to various surfaces such as wood, metal or stone (Nkafamiya, 2017).

Paint is known to provide an economic protection, preservation and decoration, aesthetic and adds functionality to structures (Crowley *et al.,* 2008). As much as paint production and application are of matter of necessity and vital to life, it has also come with quite some limitations, as paint thinned with solvent, though prove to be of better qualities like good water resistance, glossiness, and durable among other things, but was found of efflorescing volatile organic compound (VOCs), which is inimical to safe environment, as VOC depletes the ozone layer of the atmosphere resulting to global warming and hence, has a negative consequence on environment.

Scientists never gave up on minimizing, or even eliminating these limitations in the quest to build a safe and healthy world of paints and coatings. However, emulsion/water thinned paints prove to be environmental friendly, though, inferior in qualities to oil thinned paint (Kumthekar and Kolekar, 2011).

The vehicle (that is the binder) is one of the most important ingredients in paint formulation because it is a film-forming material or oil (Gopalan, 2000). Polyvinyl Acetate (PVAc) is one of the convectional binder used in the production of emulsion paint. However, the setback usually experience in emulsion paint formulated by PVAc is largely due to deficiency in PVAc, as PVAc is found to be inflexible, poor water resistance and capable of fleeting. Therefore, in order to supplement for these inadequacies, PVAc is modified with hydroxylated oil.

II. Materials

PVAc, Avocado seed oil, NaOH, HCl, sodium dihydrogen phosphate, sulphuric acid, acetic acid, formic acid, Hydrogen peroxide, Kaolin, Butanol. Petri dishes, Beakers, Conical flasks, Measuring cylinders, three neck flask, hot plate, thermocouple, condenser, stirrer.

III. Methodology

A) Extraction of Avocado Seed Oil

Avocado seed oil was extracted manually, according to the method described by Evwierhoma and Ekop, 2016.

B) Epoxidation of Avocado Seed Oil

Epoxidation was carried out on the extracted avocado seed oil using the method described by Goud *et al*., 2007.

C) Hydroxylation of Epoxidised Avocado Seed Oil

Hydroxylation was carried out on the epoxidised avocado seed oil, using the procedure described by Petrovic *et al.*, 2003.

D) Copolymerisation of Pvac with HASO

The HASO/PVAc copolymer was prepared by carried out different blends (0-60%) of the HASO in PVAc. The mixture was stirred and then poured into a glass Petri dish for casting. The resin was also allowed to cure and set for seven days at (30°C). The physical properties of the resin films were investigated (Osemeahon *et al.*, 2013).

E) Determination of Moisture Uptake

The resin films moisture uptake was determined gravimetrically as described by Osemeahon and Archibong, 2011. Average value of triplicate determinations of each sample was recorded.

F) Determination of Viscosity and Gel Time

Viscosity and gel time was carried out according to method described by Osemeahon *et al.,* 2013. Average value for the triplicate measure was recorded. The gel point of the resin was determined by measuring the viscosity of the resin with time until a constant viscosity profile was obtained (Osemeahon *et al.*, 2013).

G) Water Solubility

Solubility of the resins in water was determined by mixing 1ml of the resin with 5ml of distilled water at room temperature (30°C).

H) Density, Turbidity, Melting Point and Refractive Index

Density was determined using density bottle of standard volume as described by Yelwa *et al.,* average value of five readings was taken for each sample.

The turbidity of the sample will be determined using Supertek digital turbidity meter (Model 033G).

The melting point of the film sample was determined by using Galenkamp melting point apparatus (Model MFB600-010F).

The refractive index of the sample was determined with Abbe refractometer. The above properties was determined according to standard methods.

IV) Determination of Elongation at Break

The elongation at a break was determined using Inston Tensile Machine (model 1026). Resin films of dimension 50mm long, 10mm wide and 0.15 mm thick was brought to rupture at a clamp rate of 20mm/min and at a full load of 20kg. Average value of triplicate was recorded.

V. Results and Discussion

A) FT-IR Spectra Analysis

FTIR spectroscopy has been widely used by many researchers to study the formation of blends (Satyamaiah *et al.*, 2014). FTIR spectroscopy is one of the powerful tools used in identifying and investigating the presence of functional groups in a molecule, as each specific chemical bond often has a unique energy absorption band, and can obtain structural and bond information on a complex to study the strength and the fraction of hydrogen bonding and miscibility (Kaniappan and Latha, 2011).

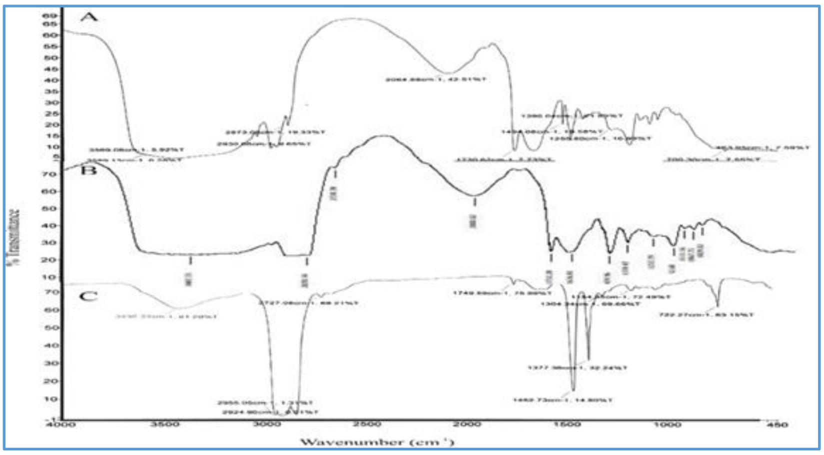

Figure 1. FT-IR spectra of A (PVAc), B (HASO/PVAc) and C (HASO)

Figure 1, shows the FT-IR spectra of A (PVAc), B (HASO/PVAc) and C (HASO). The FT-IR spectrum of PVAc, shows a dominant band at 3589.13 cm^{-1} , this was due to the O-H stretching vibration, different intense peaks such as the doublet of the $-CH_3$ and $-CH_2$ anti-symmetric stretching vibration at 2930.08 cm^{-1} and 2873.06 cm^{-1} were also detected (Yelwa *et al.*, 2017). C=O stretching vibration associated with acetate groups with a molecular vibration at 1730.62 cm^{-1} was detected (Sudharsan *et al.*, 2012).

Intense peaks at 1494.61 cm^{-1} and 1396 cm^{-1} was due to the $–CH_2$ and $–CH_3$ bending vibration. Another intense peak was detected at 2064.88cm^{-1}, and this was as a result of –C=C asymmetric stretching of vinyl acetate. The wavelengths absorption at 1255.80 and 700.30 cm^{-1} was due to the characteristic C-O-C stretching vibration of acetate and C-H bending vibration.

In the spectrum of the blend B (HASO/PVAc), the broad O-H peak at 3569.06cm^{-1} in PVAc appears at a lower frequency of 3407.73 cm^{-1} in the blend. This may be due to the hydrogen-bonded hydroxyl groups that contribute to the complex vibrational stretches associated with free inter- and intra-molecular bound hydroxyl groups (Shashidhara and Jayaram, 2010). This observation indicates a chemical reaction has taken place between HASO and PVAc.

The sharp bands at 2870.54 cm^{-1} which is characteristic of C-H stretches associated with the methane hydrogen atoms, was observed. The band at 1732.20 cm^{-1} was assigned to the C = O stretching of saturated ester. The broad absorption at 2088.82 connotes the presence of –C=C asymmetric stretching of vinyl acetate.

In the FT-IR spectrum of C (HASO), the broad bond stretching at 3436.32 cm^{-1} is due to –OH group present in the HASO, (Osemeahon, 2011). The dual band sharp transmitted at 2955.05 cm^{-1} and 2924.96 produced by stretching of the C-H group of alkane in the spectrum of HASO can be remarkably distinguished.

The peak at 1749.59 cm^{-1} indicates C=O stretching vibrations of the saturated ester present in the oil (Yelwa *et al.*, 2017). In addition, the region of 1377.36 and 1462.73 cm^{-1} of the IR spectrum show two bands that correspond to the bending vibration of C-H of alkane. The peak at 1154.35 cm^{-1} is due to C-O stretching mode of carboxylic acid (Patrick *et al.*, 1991).

B) Effect of HASO concentration on the Density of HASO/PVAc resin

The effect of HASO concentration on the density of HASO/PVAc resin is shown in figure 2. Increase in concentration of HASO lead to gradual decrease in the density of HASO/PVAc resin. Since pure PVAc has higher density than HASO, it is expected for the HASO/PVAc resin to experience decrease in density, as concentration of HASO is increasing (Blaise *et al.*, 2012).

This can also be as a result of increasing in the amount of soft segment, as physical properties of polymer depend on chain length and content of soft and hard segments (Mavani *et al.*, 2007). This behaviour is similar to the findings of Osemeahon and Barminas (2006) when polystyrene was blended with TMU resin.

Density is a physical property of matter that expresses a ratio of mass to volume (m/v), and is an important physical parameter in polymer engineering processes. The density of a paint binder in a coating industry has an important influence on factors such as pigment dispersion, brush ability of paint, flow, leveling and sagging (Kayz and Rekuviene, 2011).

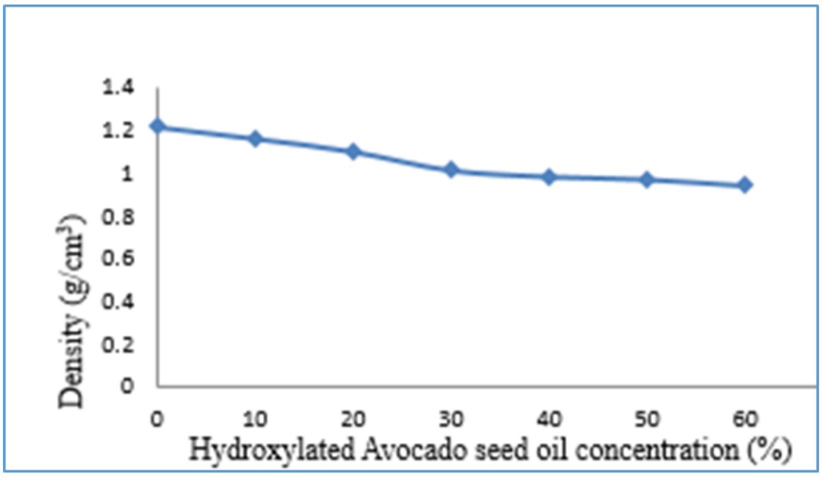

Figure 2. Effect of HASO concentration on the density HASO/PVAc resin

C) Effect of the HASO Concentration on the Refractive Index of HASO/PVAc Resin

The effect of HASO concentration on the refractive index of HASO/PVAc resin is shown in Figure 3. It is observed that, the refractive index increases with increase in HASO concentration. PVAc has lower refractive index than HASO. As the concentration of HASO increases, the optical property of HASO/PVAc resin, as regard refractive index increases. This explains the observed behaviour of the copolymer resin. The alteration of the crystallinity of the system with HASO loading give rise to higher crystallinity in HASO/PVAc resin, which also explains the above behaviour (Jain, 2008). Refractive index is one of the cogent factors that affect the gloss of a material. Gloss is an optical property which is based on the interaction of light with physical characteristic of surface. It is usually the ability of a surface to reflect light into a specific direction (Montemor, 2014). Gloss is a necessary coating property when the purposes is for surface aesthetic or decoration (Yumiko *et al.,* 2010). The refractive index of the binder is a crucial factor in the ultimate gloss property of the paint formulation (Kaygin and Akgun, 2009).

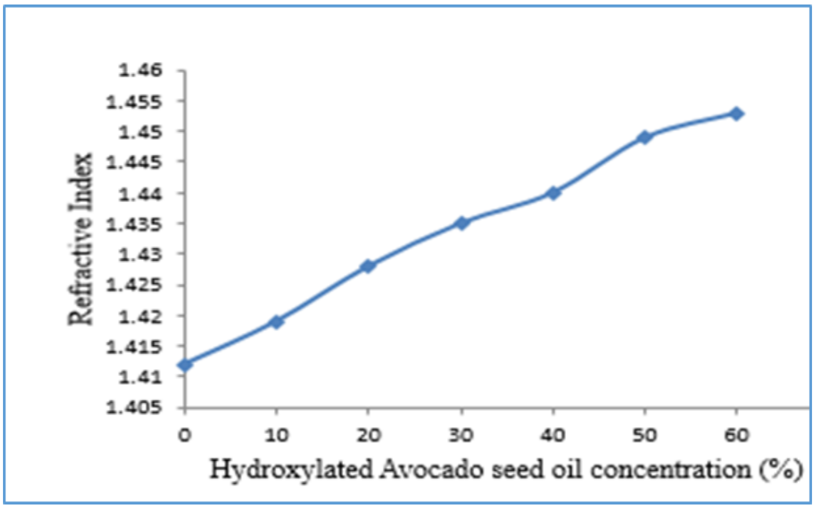

Figure 3. Effect of HASO concentration on the Refractive Index of HASO/PVAc resin

D) Effect of HASO concentration on the solubility of HASO/PVAc Resin

Table 1 shows the effect of HASO concentration on the solubility of HASO/PVAc copolymer composite. It is observed that increase in concentration of HASO causes a decrease in its water solubility. Results showed that at concentration of 10, 20, 30 and 40% of HASO, the resin is soluble. This is because the resultant hydrophilic effect of the PVAc in the

HASO/PVAc resin in these blends is higher. At concentrations of 50% HASO, resin is partially soluble, and this is because the hydrophobicity of HASO and hydrophilicity of the PVAc are roughly equal. Above 50% of HASO, the hydrophobicity of HASO dominates and the mixture becomes insoluble. Solubility is the property of a solid, liquid, or gaseous chemical substance called solute to dissolve in a solid, liquid, or gaseous solvent. The solubility of a substance fundamentally depends on the physical and chemical properties of the solute and solvent as well as on temperature, pressure and the pH of the solution (Akinterinwa *et al.*, 2015). Water solubility is an important parameter in the consideration of any resin as a binder for emulsion paint formulation. It helps us to know the ability of the resin to form emulsion in each blend.

Table 1. Effect of HASO concentration on the solubility of HASO/PVAc Resin

HASO Concentration (%)	Solubility
0	Soluble
10	Soluble
20	Soluble
30	Soluble
40	Soluble
50	Partially Soluble
60	Insoluble

E) Effect of HASO concentration on the turbidity of HASO/PVAc resin

Figure 4 shows the effect of HASO concentration on the turbidity of HASO/PVAc resin. Turbidity is relatively high at 0% HASO, because PVAc is cloudy in nature. However, on addition of HASO it becomes more homogenous, and the light scattering decreases. As the concentration of HASO increases, the suspended matter in PVAc is being reduce to particles of uniformly small and evenly distributed, the cloudy solution becomes more clearer, which lead to decrease in light scattering. Hence turbidity of the system can be used as an indicator of the level of inter chain cross-linking (Al-Manasir *et al.*, 2009). Turbidity is the degree of cloudiness or haziness of a fluid caused by large numbers of individual particles that are generally invisible to the naked eyes. Light interaction with a colloidal system is its turbidity. Turbidity actually derived its principle from light scattering. When we have homogeneity and few particles, there will be less scattering; hence, higher scattering is observed when we have a non-homogenous system with a lot of particles (Biresaw and Carriere, 2004).

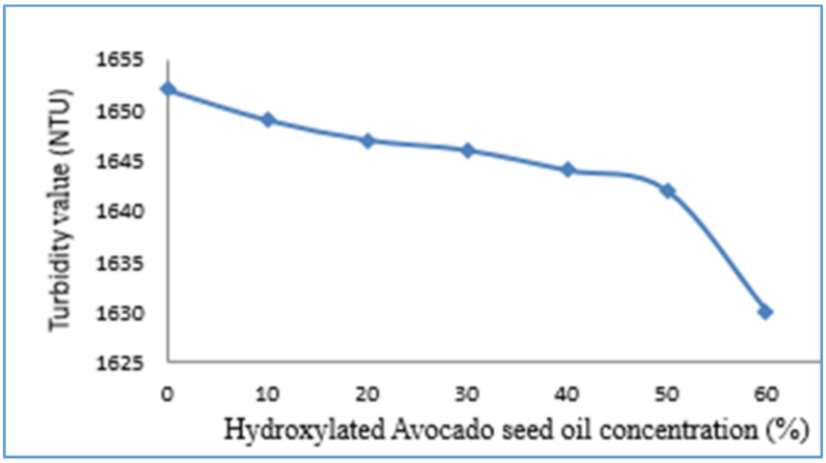

Figure 4. Effect of HASO concentration on the turbidity value of HASO/ PVAc resin

F) Effect of HASO on the viscosity of HASO/PVAc resin

Figure 4.6 shows the effect of HASO on the viscosity of HASO/PVAc resin. The result showed that, there is a steady decrease in viscosity on addition of 10, 20, and 30% of HASO, and a slight decrease at 40 and 50% of HASO. This arises as a result of variation in respective chain length (Yelwa *et al.*, 2017).

As the concentration of HASO is increased to 60%, a slight rise in viscosity is observed. This is due to increase in molecular weight which give rise to increase in viscosity due to increase in chain cross-linking between HASO molecules, and PVAc chains.

Viscosity is a fundamentally rheological parameter of macromolecular compounds (polymer) properties that defines resistance to flow and is related to the characteristics of composite materials widely used in many field of industry. The viscosity of a substance (liquid, gas or fluid) is simply its resistance to flow (Osemeahon *et al.*, 2013).

Studying rheological properties of fluids and gels are very important, since operation processes design depends on the way the product flows through a pipe, stirring in a mixer and packaging into containers.

Emulsion lattices has many sensory attributes which are related to their rheological properties, examples are creaminess, thickness, smoothness, spread- ability, flow -ability, brittleness and hardness (Hussain and Nasr, 2010).

Viscosity measurement allows the most sensitive or convenient way of detecting changes in colour, density, stability, solids, content and molecular weight (Biresaw and Carriere, 2004).

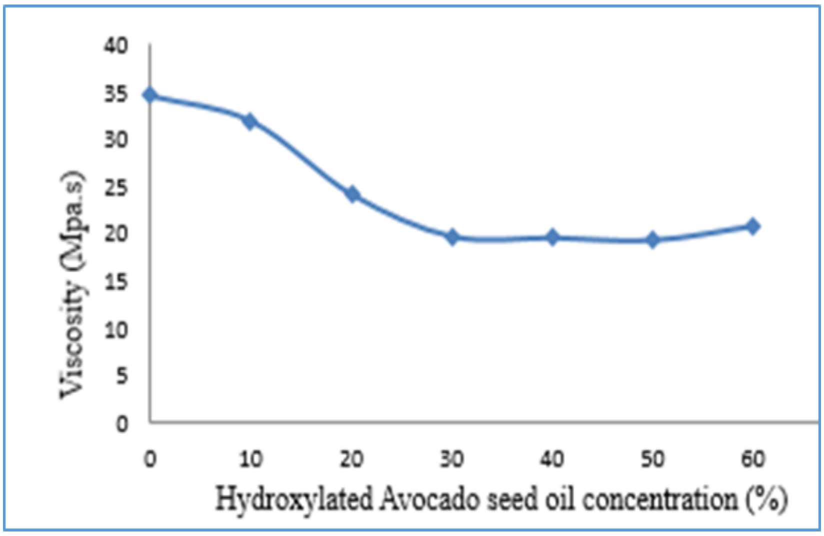

Figure 5. Effect of HASO concentration on the viscosity of HASO/PVAc resin

G) Effect of HASO on the Gel time of HASO/PVAc resin

Figure 6 shows the effect of HASO concentration on the HASO/PVAc copolymer composite. It is observed that as the concentration of HASO increases, the corresponding gel time also increases. Gel time of HASO is relatively longer compared to the gel time of PVAc due to their molecular weight.

Increase in the concentration of HASO leads to gradual and steady increase in gel time of the HASO/PVAc composite. The result showed that, the resin gelled and dried up within two weeks of casting. Therefore it is advisable that, the binder should be used within this period. This behaviour may be attributed to decrease in cross-linking density of the copolymer which gave rise to decrease in viscosity (Yelwa *et al*, 2017).

Gelation is the change in macromolecular structure of resin, which is usually expressed as an increased of viscosity and leads finally to the loss of flow-ability. Gelation has two processes: the initial period of slow viscosity growth and the period of fast structuration (suurpere *et al.*, 2006).

The dry time of any paint is a function of its binders gel time among other factors. On the technical front, gel time enable paint formulator to ascertain the optimum storage period of a binder before its utilization for paint formulation and is important in determination of adhesion (Desai *et al.*, 2003).

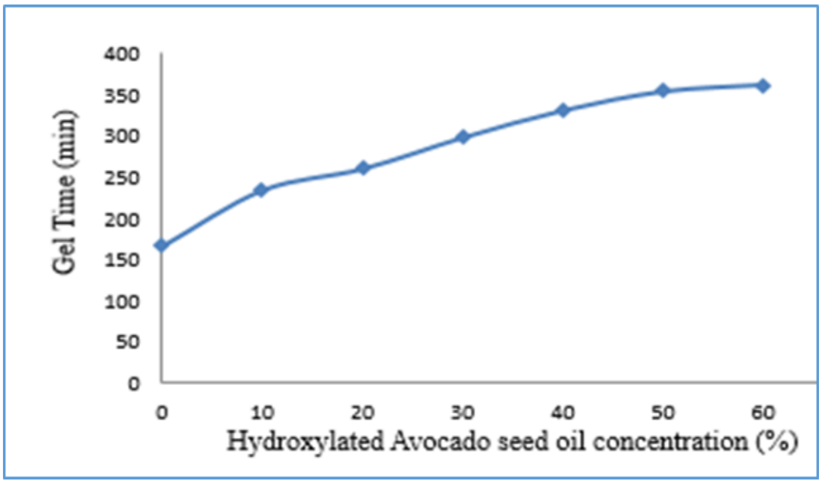

Figure 6. Effect of HASO concentration on the Gel time of HASO/PVAc resin

H) Effect of HASO on the moisture uptake of HASO/PVAc resin

The moisture uptake decreased with increase in the concentration of HASO as seen in figure 7. This is as a result of the hydro-phobicity of HASO. Increase in HASO concentration into the copolymer give rise to corresponding increase in hydrophobicity of the polymer matrix (Naghash *et al.*, 2007).

This result demonstrated that the HASO/PVAc copolymer composite will only hold a very low amount of water, and this will also address one of the shortcomings of pure PVAc, which is high moisture absorption.

Hydrophobic components in the binder give the best combination of improving water resistance of water-borne coating (Emile, 2003). Waterborne coatings are susceptible to durability issues pertaining poor water resistance.

The functional groups on polymers or copolymer resins that can undergo hydrogen or ionic bonding, unless the hydrophilic character is balanced with that of the hydrophobic, the coating will either be water sensitive or the formulation will not have colloidal stability.

One of the main drawbacks of polyvinyl acetate is its high water uptake and this has to be improved in order for the resin to satisfy the coating industry (Yelwa *et al.*, 2017). Water has a negative effect on the thermo-mechanical; chemical and adhesion properties of paint.

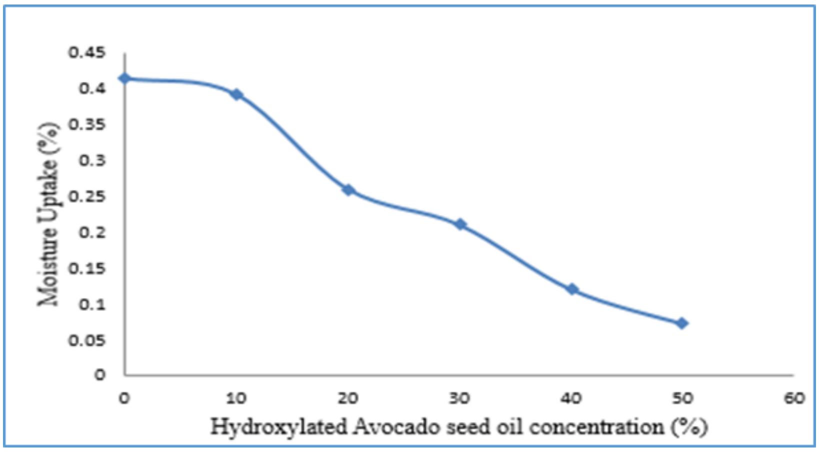

Figure 7. Effect of HASO concentration on the moisture uptake of HASO/PVAc resin

I) Effect of HASO on the Elongation at break of HASO/PVAc resin

Figure 8 shows the effect of HASO on the elongation at break (%) of HASO/PVAc copolymer. At 10% of HASO, there is a notable increase, this may be due to increase in concentration of soft segment (HASO) that impacts flexibility. From 20%, the elongation at break of the copolymer begin to decrease as the concentration of HASO increases. This non-linear behaviour in terms of the mechanical property is caused by the difference in intimate physical interlocking, and the extent of phase separation (Hwang *et al.*, 2012). Elongation at break (%), also known as fracture strain, or tensile elongation at break, is the ratio between changed length and initial length after breakage of the test specimen. It expresses the capability of a natural plant fibre or polymeric materials to resist changes of shape without cracking (Petroudy, 2017). In coating industry, knowing the elongation at break of the resin, will help to streamline the formulation process and provide insight needed to formulate a successful product.

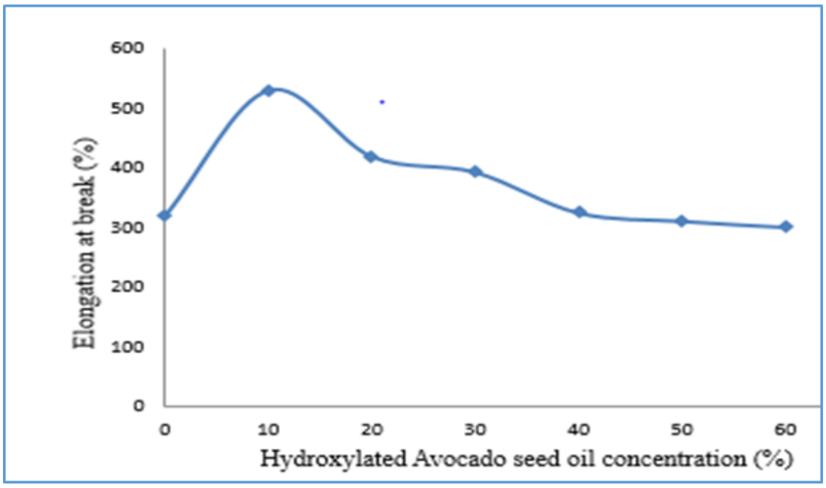

Figure 8. Effect of HASO concentration on the elongation at break (%) of HASO/PVAc resin

J) Effect of HASO on the melting point of HASO/PVAc resin

Figure 9 shows the effect of HASO concentration on the melting point of HASO/PVAc resin, where the melting point decreased steadily as HASO concentration increases. This can be explained on the basis of increase in flexibility as a result of increase in the concentration of HASO which gave rise to increase in molecular mobility (Afsoon *et al.*, 2011). However, at 40% 0f HASO, a slight increase was observed, and later decrease as concentration of HASO increases. This may be due to contributing factors arises from differences in melting points of the two polymer. The polymer blend showed lesser melting temperature as HASO concentration increases. Hence the rigidity of PVAc may be reduced via blending with HASO (Yelwa *et al.*, 2017). This again addresses one of the drawbacks of PVAc which is rigidity. The melting point of a polymer has a direct bearing on its thermal property. Melting point of polymer varies depending on molar mass, intermolecular van der Waal interactions and intrinsic structures that affect the rigidity. In the case of coating industries, the melting point of a binder is related to its thermal resistance as well as to the brittlity (Afsoon *et al.*, 2011).

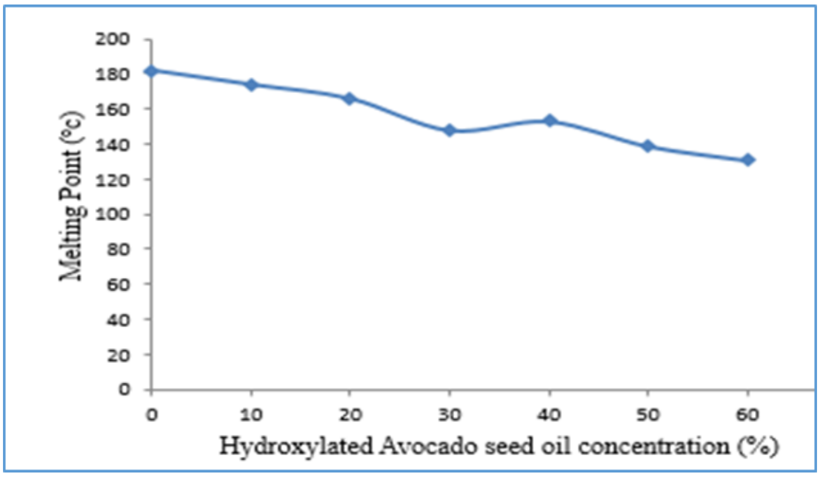

Figure 9. Effect of HASO concentration on the melting point of HASO/PVAc resin

K) Comparison of some physical properties of PVAc, HASO/PVAc Resin and the acceptable level in the coating industry

A comparison properties of PVAc, and HASO/PVAc resins, with the acceptable level in the coating industry is presented in in Table 2. A decrease was observed in density, viscosity, turbidity, moisture uptake and melting point between the PVAc and HASO/PVAc; this shows improvement in terms of film adhesion, flexibility, better optical property, water resistance and brittlity respectively. The refractive index, gel time and elongation at break (%) increased as seen from Table 2, this indicates better gloss, consistency and high ability to withstand weather or service condition (Yelwa *et al.*, 2017).

Table 2. Comparison of some physical properties of PVAc and HASO/PVAc resins

Parameters	**PVA**	**HASO/PVAc**	**Acceptable Level in the coating industry (Osemeahon *et al.*, 2013)**
Density (g/cm^3)	1.212	1.098	1.07 (min.)
Refractive index	1.412	1.428	1.4000 (min.)
Moisture uptake (%)	0.413	0.137	3.10 (max.)
Viscosity (Mpa.s)	34.66	24.11	3.11-38.00
Melting point (°C)	184	169	200 (max.)
Elongation at break (%)	320	418	125 (min.)
Turbidity (NTU)	1652	1647	-
Gel time (Min)	92	256	-
Solubility	Soluble	Soluble	-

Table 3. Comparison of some physical properties of HASO/PVAc film with films from other paint binders

	Physical property							
Type of resin	**Visco-sity (Mpa.s.)**	**Refract-ive index**	**Density (g/cm^3)**	**Melting point (°C)**	**Moisture uptake (%)**	**Elongation at break (%)**	**Formalde-hyde emission (ppm)**	**Literature**
HASO/ PVAc	24.11	1.428	1.098	169	0.137	418	ND	This study
TMU/PS	19.70	1.425	1.0990	262	1.01	425	0.0233	Osemeahon and Dimas (2013)
Polyvinyl acetate	0.40	1.40	1.25	ND	ND	ND	ND	Habibu (2011)
SSO/ PVA	159.19	1.44	1.009	171	0.1381	ND	ND	Yelwa *et al.*, (2017)
Commer-cial UF	451	ND	1278	ND	0.2	ND	ND	Suurpere *et al.*, (2006)
UF/Starch	11.100	1.417	1.166	130	0.6	ND	0.004	Osemeahon *et al.*, (2013)
MU/NR	248	1.3411	0.641	255	1.341	350.43	0.058	Kazys and Rekuviene (2011)
UF/PE	32.60	1.432	1.3362	130	0.0080	250.0	0.0142	Osemeahon and Arachibong (2011).

VI. Conclusion

The polyvinyl acetate was successfully modified with hydroxylated avocado seed oil to form HASO/PVAc copolymer binder. The FT-IR analysis confirmed chemical interaction between HASO and PVAc chemical structure. The physical parameter of the new copolymer binder were analysed. The results shows a good solubility interaction between Hydroxylated avocado seed oil and polyvinyl acetate at 10-40% of the hydroxylated oil.

The physical properties were in conformity with literatures and within the acceptable level in coating industry. The copolymer HASO/PVAc resin shows to be of low moisture uptake and better flexibility against conventional poor water resistance and brittleness associated with polyvinyl acetate. Increase in elongation, and reduction in melting point is also a positive development to the coating industry. Therefore, the analysed result shows that, the HASO/PVAc copolymer could be used as a binder for emulsion paint formulation.

VII. Acknowledgement

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Conflicts of interest: There is no conflict of interest of any kind.

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