

Full Length Research Paper

Influence of starch addition on properties of urea formaldehyde/starch copolymer blends for application as a binder in the coating industry

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Urea formaldehyde resin was reactively blended with various concentrations (10 to 70%) of cassava starch in order to develop a paint binder for emulsion paint formulation. Some physical properties and formaldehyde emission of the blended resin were investigated. Viscosity initially decreased before a gradual increase was noted with increase in starch concentration. Refractive index and elongation at break increased initially but gradually decreased with cassava starch content in the blend. Gel time, density, melting point, moisture uptake and formaldehyde emission decreased with increase in starch inclusion. The interaction between the two different polymers shows that 50% starch was the optimal loading inclusion. This new system has advantages of low brittleness, low formaldehyde emission and water reduction characteristics. Therefore the polymer blend can be recommended as binder for coating industry.

Key words: Urea formaldehyde, starch, copolymer, binder.

INTRODUCTION

The development of reliable high performance coating materials with excellent thermal and mechanical properties is the focus of modern technology. Water-borne coatings are finding more application due to increased legislative restrictions on the emission of volatile organic materials to the atmosphere (Motawie et al., 2010). Depending on the type of binder, paints are classified into 2 main categories; that is, oil paint which is oil based, and emulsion paint which has synthetic resin as the binder, and its water soluble as against oil paint which is solvent (organic solvent) soluble. Although oil-based paints display a lot of advantages such as water resistance, durability and flexibility, its major drawback is its use of organic compound as solvent, which is threatened by growing proliferation of Volatile organic

compounds (VOC) regulations imposed worldwide.

Water-borne resins are polymeric materials whose composition enables them either to dissolve or to swell in water. Most buildings are protected and decorated using water-borne paints due to their ease of applications, fast drying, non-odour, good wash-ability and finish. However, although most household paints are water based, this is not true of industrial paints because of the special requirement of the industrial coatings. Hence, satisfactory water based polymers with the required properties have not yet been developed thus leading to emphasis on sustainable commercial production of water-borne paints (Hasmukh and Sumeet, 2010; Motawie et al., 2010; Osemeahon, 2011). Developing nations however, will need to put more effort into the development of local

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technologies for the purpose of achieving the above target. Polymer blends has grown tremendously in leaps and bounds over the past few decades (Oluranti et al., 2011). Blending could be a simple process for developing new composites for coating systems. Polymer blend is one of the most useful approaches to prepare new materials with specially tailored and improved properties that are often absent in a single polymer. The performance of a polymeric material can be improved by selection of suitable ingredients and their ratios, leading to the formation of a new material with enhanced physical, chemical and mechanical properties (Kaniappan and Latha, 2011; Hwang et al., 2012). The synthesis of a new class of urea formaldehyde (UF) resin through a one step process was reported by Osemeahon and Barminas (2007). Although this new class of resin showed much improvement in terms of formaldehyde emission and moisture uptake (water resistant), compared with the traditional urea formaldehyde (UF) resin, its brittleness and hardness remains a source of concern (Osemeahon and Archibong, 2011). Hence the need to modify this new class of urea formaldehyde (UF) resin in order to address the problem of brittleness and further reduce both formaldehyde emission and moisture uptake.

This work seeks solution to the problem, by the copolymerization reaction between urea formaldehyde/cassava starch (UF/CS). Starch is one of the most abundant natural polymers, used in a wide range of products including binders, sizing materials, glues and pastes. Inexpensive materials such as starch, are biodegradable additives, which are appropriate for blending with synthetic polymers (Amine et al., 2010; Borghei et al., 2010). The cassava starch has a high purity level, excellent thickening characteristics, a neutral (bland) taste, desirable textural characteristics, relative cheapness and contains a high concentration of starch (dry-matter basis) which is an advantage over other grains or root crop. Its high paste viscosity, clarity, freeze-thaw stability and renewability are advantageous to many industries.

The Nigeria government cassava project is aimed at increasing the utilization of cassava for industrial purposes, which is why recently, many research has focus on conversion of cassava into industrial products and raw materials (Akpa, 2012).

MATERIALS AND METHODS

Urea formaldehyde, sodium di-hydrogen phosphate, sulphuric acid, sodium hydroxide pellets, sucrose, were reagent grade products from the British Drug House (BDH). The materials were used as received. Cassava tubers were collected from a farm in Yola, Nigeria.

Treatment/preparation of starch

The cassava tubers were washed, dried and grounded into

powder. It was treated by dispensing in cold water and filtering. The filtrate was heated using hot plate with occasional stirring of 5 min intervals, until colloidal suspension was observed which formed a gel on cooling. The method used for the resin synthesis, blend/film preparation and determination of the film properties was according to Osemeahon and Archibong (2011).

Resin synthesis

Urea formaldehyde (UF) resin was prepared by reacting one mole (6.0 g) of urea with three moles (24.3 ml) of 37% (w/v) formaldehyde using 0.2 g of sodium dihydrogen phosphate as catalyst. The pH of the solution was adjusted to 6, using 0.5 m H₂SO₄ and 1.0 m NaOH solution. The reaction was then allowed to proceed for 2 h in a thermostatically control water bath at 70°C, after which the sample was removed and kept at room temperature (30°C).

Blend/film preparation

Blending of urea formaldehyde resin with cassava starch was carried out by preparing 10% cassava starch in urea formaldehyde at room temperature (30°C). The solution was mixed thoroughly using a magnetic stirrer. The above procedure was repeated at different cassava starch concentrations (10, 20, 30, 40, 50 and 70%) respectively and the resulting blends analyzed. Copolymer of the different resins obtained with various cassava starch concentrations were introduced into a glass Petri dish for casting. The resins were then allowed to cure and set for seven days at 30°C and the physical properties of these copolymers were carefully investigated.

Determination of viscosity and gel time

A 100 ml Phywe made graduated glass macro-syringe (Phywem, Gottingen, Germany) was utilized for the measurement. The apparatus was standardized with a 20% (w/v) sucrose solution whose viscosity is 2.0 mPa.s at 30°C. The viscosity of the resin was evaluated in relation to that of the standard sucrose solution at 30°C. Five different readings were taken for each sample and the average value calculated. The gel point of the resin was determined by monitoring the viscosity of the resin with time until a constant viscosity profile was obtained.

Determination of density, melting point and refractive index

The density of the different resins was determined by taking the weight of a known volume of resin inside a density bottle using Mettler Model, AT400 (GmbH, Greifensee, Switzerland) weighing balance. Five readings were taken for each sample and average value calculated. The turbidity of the resin samples was determined using Hanna microprocessor turbidity meter Model, H193703 (Villafraanca Padovana, Italy). The melting points of the different film samples were determined using Galenkamp Melting apparatus Model, MFB600-010F (Loughborough, UK). Different copolymer samples were grounded into powder and some quantity of each sample was introduced into different capillary tubes. The melting point was then taken one after the other for all samples. The refractive indices of the resin samples were determined with Abbe refractometer (Bellinglam and Stanley, Tunbridge Wells, Kent, UK). Five readings were taken for each sample and the average value calculated for each of the aforementioned parameters. The properties of the blends were also determined according to standard methods (AOAC, 2000).

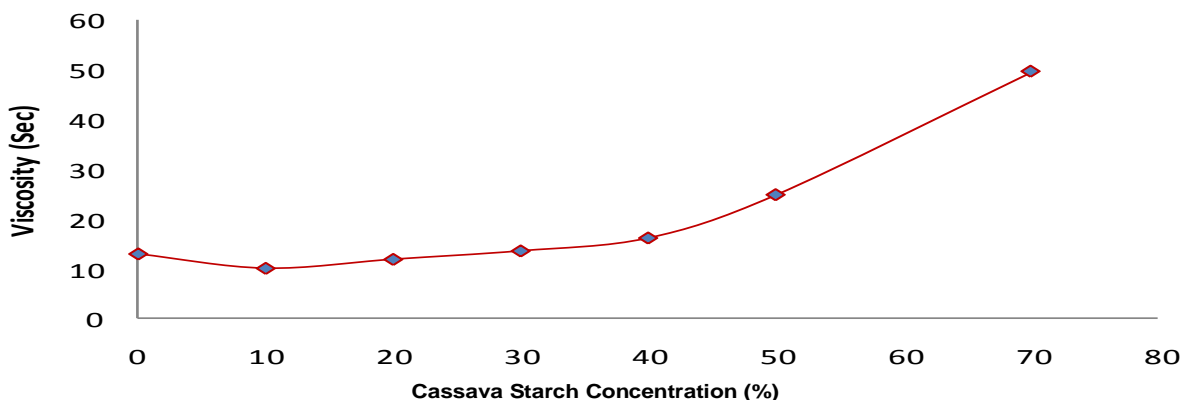


Figure 1. Effect of cassava starch concentration on viscosity of methylol urea resin.

Determination of moisture uptake

The moisture uptake of the resin film was determined gravimetrically as known weights of the sample were introduced into desiccators containing a saturated solution of sodium chloride. The increase in weight (wet weight) of each sample was monitored until a constant weight was obtained. The difference between the wet weight and dry weight of each sample was then recorded as the moisture intake by resin. Triplicate determinations were made for each sample and the average value recorded.

Determination of formaldehyde emission

Formaldehyde emission test was performed by using the standard 2 h desiccators' method. The evaluation of the absorbed formaldehyde by the 25.0 ml water was obtained from standard calibration curves derived from refractometric technique using Abbe refractometer. In brief, the prepared resin was aged for 2 days. At the end of this period, the resin was poured into a mold made from aluminium foil with a dimension of 69.6 × 126.5 mm and thickness of 1.2 mm. The mold and its content was then allowed to equilibrate for 24 h in the laboratory after which it was then placed inside a desiccator along with 25 ml of water, which absorbed the formaldehyde emitted. The set up was allowed to stay for 2 h after which the 25 ml water was removed and analyzed for formaldehyde content. Triplicate determinations were made for each sample and mean value recorded.

Elongation at break

Elongation at break was measured, using Inston testing machine (model 1026). Resin films of known dimension 50 mm long, 10 mm wide and 0.15 mm thick were brought to rupture at a clamp rate of 20 mm/min and a full load of 20 kg. A number of five runs were done for the sample and the average elongation evaluated and expressed as the percentage increase in length.

RESULTS AND DISCUSSION

Viscosity and gel point

The viscosity of a substance (liquid, gas or fluid) is its

resistance to flow. 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; Akpa, 2012).

Because of the presence of functional groups in the polymeric backbone, inter-polymeric specific interactions have long been known to result in unusual behavior and material properties that are dramatically different from those of the nonfunctional polymers. These interactions include ion-ion Coulombic interaction, hydrogen bonding and transition metal complexation of the component polymer chains, resulting in solution viscosity variation. Rheological properties such as the viscosity can be directly correlated to the evolving physical and mechanical properties during resin cure (Derkyi et al., 2008; Osemeahon, 2011). The effect of cassava starch on the viscosity of urea formaldehyde resin is observed (Figure 1). At low concentration of 10% cassava, the viscosity decreased slightly and then increased with increase in cassava starch concentration. This phenomenon can be explained in terms of specific interactions between urea formaldehyde and cassava starch.

In a dilute system, there are strong specific interactions and the complexes are isolated from each other with the formation of compact structure, which reduced the viscosity of the blend solution. However, as the blend concentration increases, the isolated complexes combined, lead to the formation of a gel-like intermolecular complex structure leading to increase in viscosity of the copolymer blend system, since the variation is linear, it indicates the miscibility of polymer blend (Osemeahon and Barminas, 2007; Reddy et al., 2008).

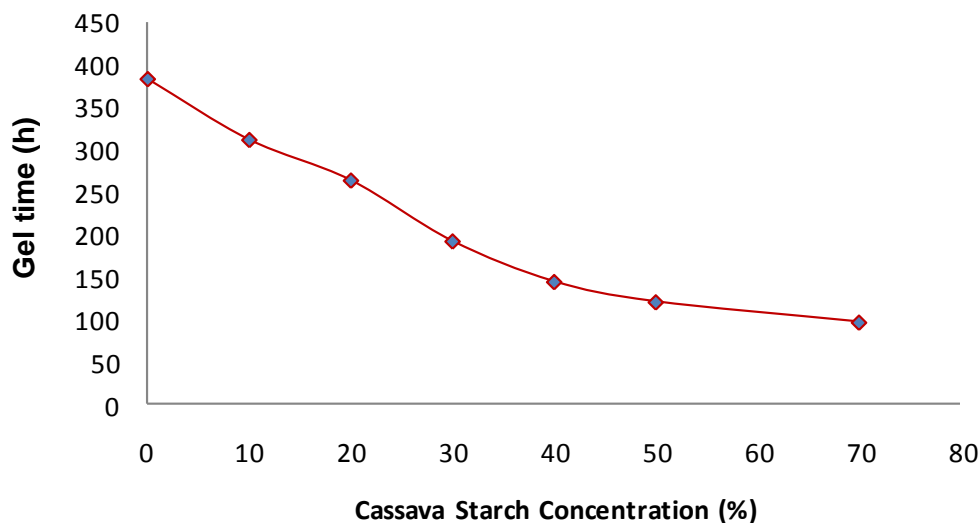


Figure 2. Effect of cassava starch concentration on gel time of methylol urea resin.

Viscosity also increases with increase in solid content as a consequence of higher surface interactions among the particles. Concentrated polymer shows a great deal of interaction between the macromolecule. The higher the concentration, the higher the viscosity observed (Palma, 2007; Taghizadeh and Toroutan, 2005). The gel time or pot life is the maximum length of time the system remains in sufficiently fluid condition to be applied to a substrate. 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. The gel time is an important kinetic characteristic of curing because it describes the attainment of certain critical conversion responsible for the transition from liquid to solid state of the curing process. The gel point is characterized by the appearance in the reactive system of macromolecule with an infinitely large molecular weight (Desai et al., 2003; Derkyi et al., 2008). Figure 2 shows the effect of cassava starch concentration on gel time. It can be observed that the gel time decreases with increase in cassava starch concentration. The reaction between monomers leads to the formation of network, hence gelation. Both molecular weight and poly-dispersity increase until one single macromolecule is formed. At this point, the behavior of the system changes from liquid-like to rubber-like thus the reactive system becomes a gel (Gonzalez et al., 2012).

Density

Density is a physical property of matter that expresses a ratio of mass to volume. It is very useful for identification and characterization of substances. The density of a

paint binder in the coating industry has a profound influence on factors such as pigment dispersion, brushability of paint, flow, leveling and sagging (Kazys and Rekuviene, 2011; Osemeahon and Archibong, 2011). Figure 3 shows the effect of cassava starch on the density of urea formaldehyde resin. The gradual decrease observed in density with increase in cassava starch concentration can be as a result of differences in the molecular features and morphology which influenced the packing nature of resin molecules as the concentration of starch increases (Osemeahon and Barminas, 2007). This result is similar to the findings of Barminas and Osemeahon, 2007 when natural rubber was blended with methylol urea resin.

Melting point

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 brittleness. Urea formaldehyde resin is known to compose of molecules that cross-link into clear hard plastics (Afsoon et al., 2011; Osemeahon et al., 2010). Figure 4 shows the effect of cassava starch on the melting point of methylol urea resin where the melting point decreased slightly at the beginning up to 10% cassava starch. Thereafter, a sharp decrease in melting point was observed. The melting points of the two monomers are very different which probably result from different contributing factors.

At a certain domain of concentrations of the two copolymers, their melting point decreased even below the melting point of each monomer. The melting point of the

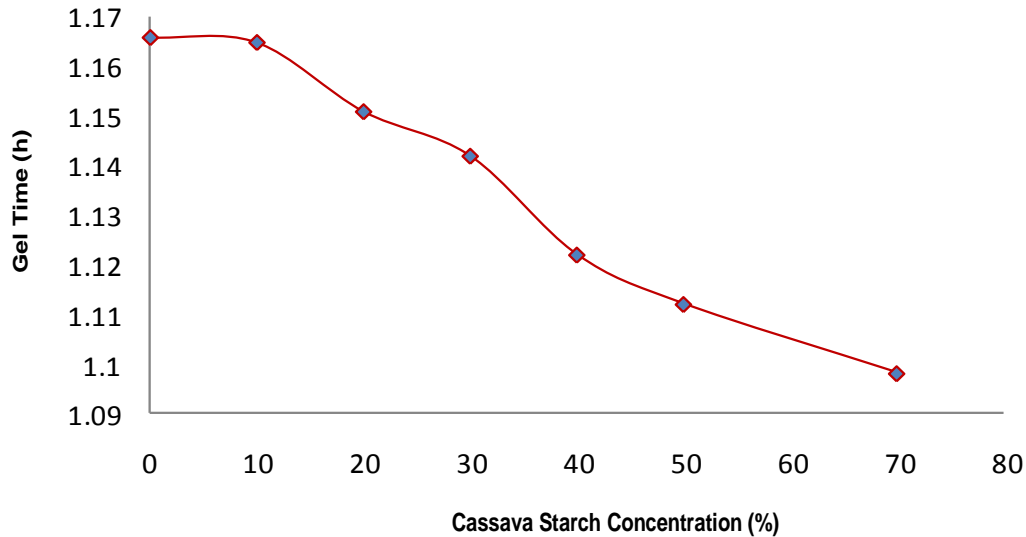


Figure 3. Effect of cassava starch concentration on the density of methylol urea resin.

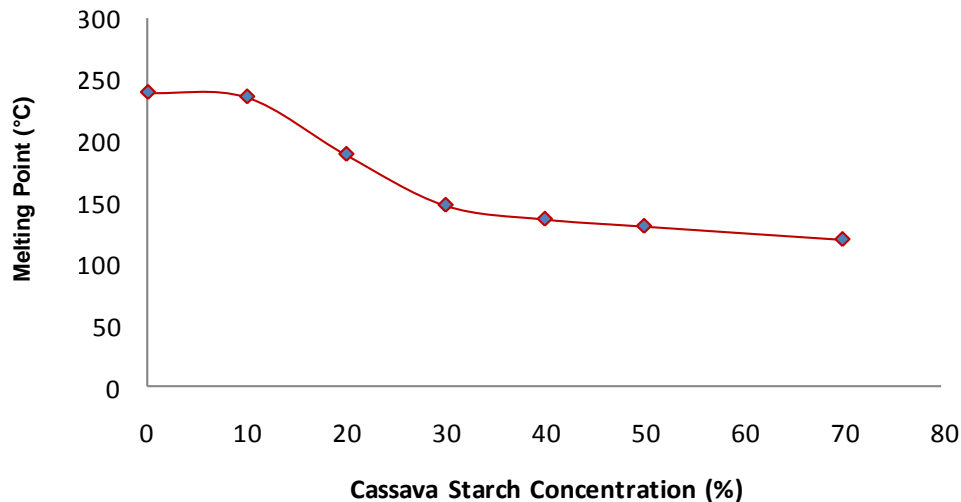


Figure 4. Effect of cassava starch on the melting point of methylol urea resin.

blended films shifted due to interaction of the two biopolymers as influenced by the lower melting temperature of cassava starch (Afsoon et al., 2011; Wirongong et al., 2011). This decrease in melting point is due to increase in molecular mobility which is a positive result.

Refractive index

Gloss is a measure of the ability of the coated surface to reflect light. Reflection of light from surfaces can be classified according to the diffuse component or the specular component, which is expressed as a function of

the incidence angle and refractive index of the material, the surface roughness and a geometrical shadowing function. Gloss is a necessary coating property when the purpose is for surface aesthetics or decoration (Kaygin and Akgun, 2009; Yumiko et al., 2010). Figure 5 presents the effect of cassava starch on the refractive index of urea formaldehyde resin. Initially, the refractive index increased from 0 to 20% cassava starch inclusion after which a gradual decrease in refractive was observed with increase in cassava starch concentration. This result is due to differences in the level of specific interaction between the two polymers resulting in molecular weight, molecular features and molecular orientations depending on morphology and crosslink density (Qi et al., 2002).

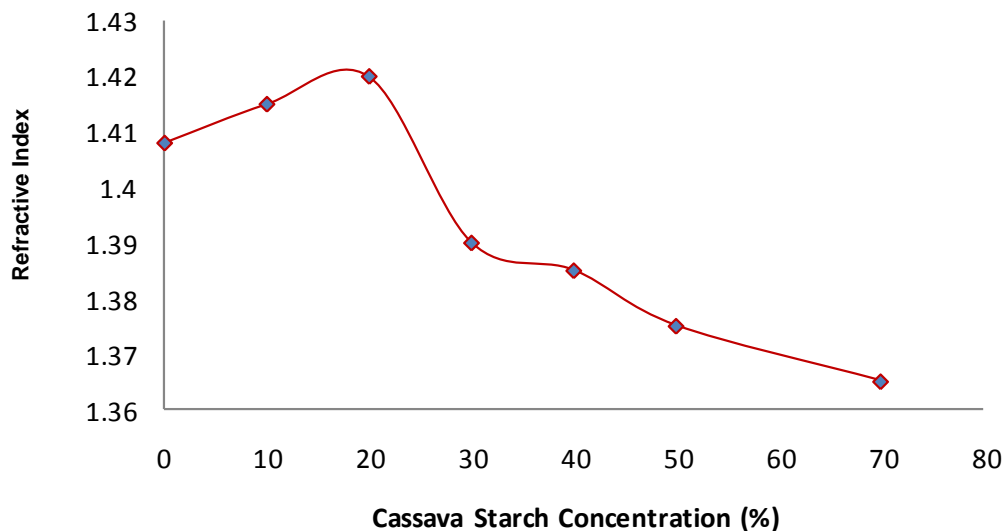


Figure 5. Effect on cassava starch concentration on the refractive index of methylol urea.

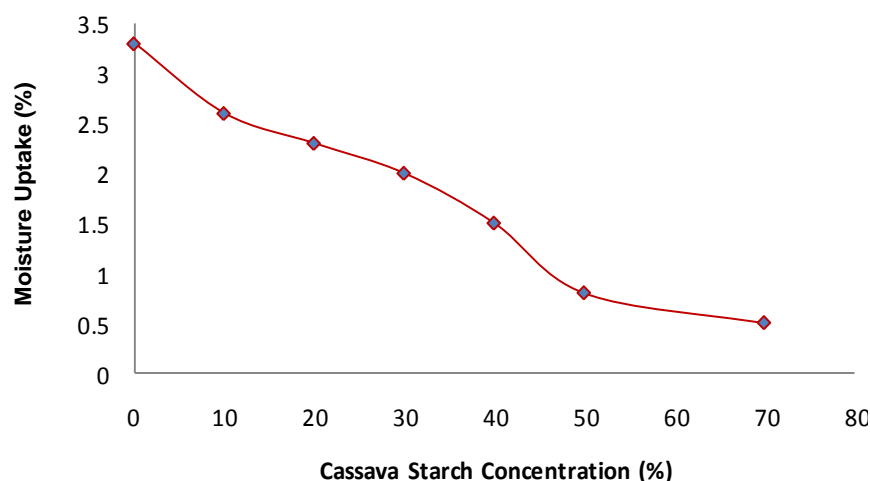


Figure 6. Effect of cassava starch on the moisture uptake of methylol urea resin.

This suggests that from 0 to 20% cassava starch inclusion in the gloss property especially on a smooth surface increases after which a decrease is observed.

Moisture uptake

Polymeric binders play a major role in moisture transport properties of paint because it is one of the major constituent. Susceptibility to durability issues pertaining poor water resistance is one of the draw-backs of water-borne resin. The functional groups on polymers that are used can undergo hydrogen bonding. A detailed knowledge of moisture transport is essential for understanding the resistance of a material against

attacks from its environment (Emile, 2003; Bharath and Swamy, 2009).

Figure 6 shows the effect of cassava starch on the moisture uptake of methylol urea resin. It can be observed that moisture uptake drastically dropped at the beginning and decreased steadily after 10% blend until after 40% blend. The different levels of interactions gave rise to polymers with different morphology and crosslink density. From 0 to 10% blend, the molecular size holes in the copolymer structure were rapidly reduced. After this period, the size of the molecular size holes might have slowly decreased with increase in cassava starch loading; hence the steady reduction in moisture uptake. After the steady decrease in moisture uptake between 10 to 40% blend, a sharp decrease in moisture uptake was

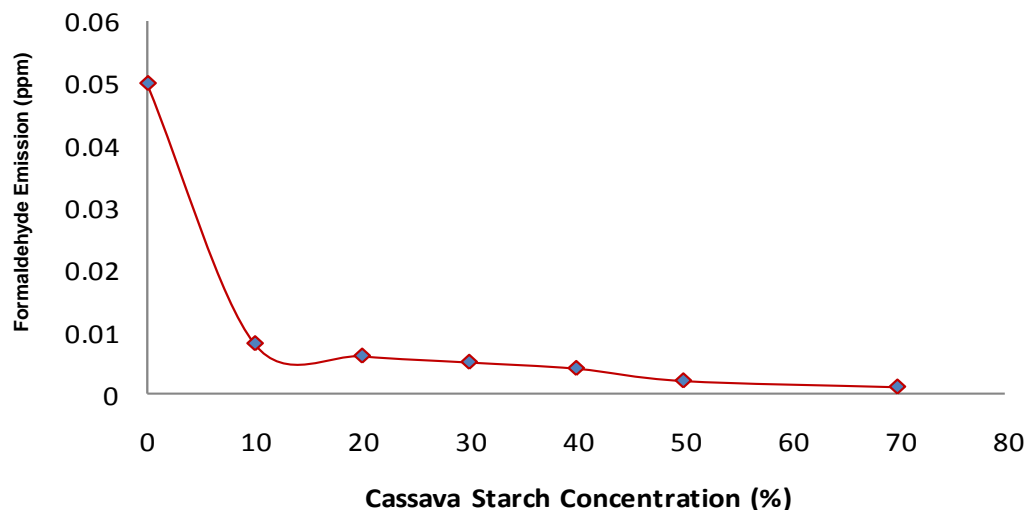


Figure 7. Effect of cassava starch on the formaldehyde emission of methylol urea resin.

noted. This might be as a result of the drastic decrease in molecular size hole with increase in cassava starch inclusion. Blending improves the resistance to moisture susceptibility of mixtures as also seen in waste plastic coating of aggregates (Bindu and Beena, 2010).

Formaldehyde emission

A serious drawback of urea formaldehyde resin is the emission of the hazardous formaldehyde during cure. The issue of formaldehyde exposure in homes is long-standing and has been studied overtime. Hydrolysis of cured urea resins has been known to be responsible for formaldehyde emission leading to sick building syndrome. It is therefore important to determine the formaldehyde emission from synthesized urea-formaldehyde resin (Derkyi et al., 2008; Park et al., 2010). It is observed in Figure 7 that the level of emission decreased with increase in the concentration of cassava starch in the blend. This important promising trend is due to the gradual decrease in the methylol urea content with increase in the cassava starch content in the blend. The blending process has succeeded in reducing the emission lower than the permissible level of 0.1 ppm as stated by Pizza et al. (2002). This agrees with the findings of the copolymerization of urea formaldehyde with acrylamide (Abdullah et al., 2010).

Elongation at break

The structure of thermosetting resins also leads to some unwanted mechanical properties that limit their structural applications. They are relatively brittle. Elongation at break can be a propel tool to determine the adhesion

between phases because of its sensitivity for load transfer between phases (Cakir et al., 2012; Hwang et al., 2012).

Figure 8 shows the effect of cassava starch on the elongation at break of methylol urea resin. A steady increase in elongation at break was observed from 0 to 40% cassava starch and an abrupt increase at 50% due to compatibilization effect and improved interfacial adhesion, hence flexibility is improved and brittleness reduced. Also, a decrease was observed after 50%. This non-linear behavior 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). The result is in consonance with the report of Cardona and Moscou (2010) where resole phenolic resins were modified by forming copolymer with cardanol (main component of cashew nut shell liquid), the presence of cardinol decreases the crosslink density and makes the resole phenolic resins less brittle. Table 1 shows the comparison of some physical properties of UF and UF/CS resins.

Conclusion

The analyzed results obtained from the sample analysis showed that the interaction between methylol urea and cassava starch has remarkable synergistic properties with 50% of cassava starch being the optimal inclusion value. The values of moisture uptakes, formaldehyde emission and melting point obtained from this experiment are within the acceptable levels required in the coating industry as shown in Table 1.

This results present cassava starch modified methylol urea as a resin with relatively very low moisture uptake and formaldehyde emission against the traditional hard

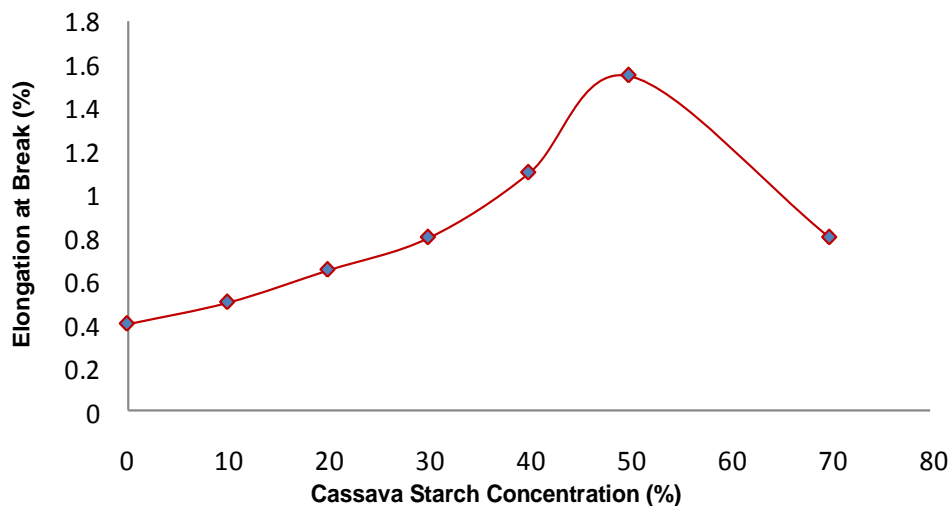


Figure 8. Effect of cassava starch on the elongation at break of methylol urea resin.

Table 1. Comparison of some physical properties of UF and UF/CS resins.

Parameter	UF	UF/CS	Acceptable Level in the coating industry (Osemeahon et al., 2009)
Density (g/cm)	1.168	1.166 ± 0.001	1.07 (min.)
Refractive index	1.410	1.417 ± 0.1000	1.4000 (min.)
Formaldehyde emission (ppm)	0.0500	0.0040 ± 0.002	0.1 (max.)
Moisture uptake (%)	3.3000	0.6000 ± 0.001	3.10 (max.)
Viscosity (mpa.s)	3.1111	11.1000 ± 0.1100	3.11- 38.00
Elongation at break (%)	40	160.00 ± 0.01	125(min.)
Melting point (°C)	250.00	130.00 ± 0.10	200(max.)

and brittle resin. The increase in elongation at break and reduction in density is also a positive value to the coating industry. Therefore, this work has shown that biodegradable starch films could be used to produce copolymer composite binder for the coating industry especially in the formulation of emulsion paints.

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