

*Full Length Research Paper*

# Effect of urea formaldehyde viscosity on some physical properties of a composite from reactive blending of urea formaldehyde with natural rubber

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**In this work, the effect of urea formaldehyde (UF) viscosity on a composite derived from reactive blending of UF with natural rubber (NR) was investigated. Some physical properties of the urea formaldehyde-natural rubber (UF-NR) obtained at different UF viscosities (2.1-256.3mPa·s) were evaluated. The melting point, refractive index, density and formaldehyde emission were found to increase with increase in UF viscosity while the dry time, moisture uptake and elongation at break were found to decrease with increase in viscosity. UF viscosity below 30.55mPa.s was found to produce UF-NR blend soluble in water and beyond this value the composite became insoluble in water. Thus processing of UF-NR blend for emulsion paint formulation could be suggested below this viscosity level. The result obtained from this experiment will offer formulators different options and to control formulation processes towards developing a paint binder for emulsion paint formulation from UF-NR blend, which may be acceptable to the coating industry.**

**Key words:** Paint binder, amino resins, urea formaldehyde, natural rubber, viscosity.

## INTRODUCTION

The acceptance of urea formaldehyde resin as a universal material in many engineering areas such as in the coating industry is impeded by some of its inherent qualities such as brittleness, poor water resistance and formaldehyde emission (Barminas and Osemeahon, 2006; Osemeahon and Barminas, 2007). These disadvantages limit its uses. However, UF resins offer a wide range of conditions that make synthesis of these resins with important properties such as gel time, tack and spreadability of the uncured resin possible. Also, formaldehyde emissions and the durability of the cured resin can be controlled and specifically tailored for the final end use of the resins (Osemeahon and Barminas, 2006b).

Park et al. (2002) reported that the ultimate performance of a fully cured amino resin largely depends on its synthesizing parameters, such as the ingredient mole ratio, catalyst, viscosity, reactivity and so on. These parameters are frequently adjusted empirically to tailor the resin properties to specific production requirements such as

the resin reactivity, formaldehyde emissions, and water resistance etc.

In the coating industry, an understanding of the viscosity of the paint binder is very important because it controls factors such as flow rates, leveling and sagging, thermal and mechanical properties, dry rate of paint film and adhesion of the coating to substrate (Osemeahon and Barminas, 2007). Kim (2001) reported that the polymerization reaction in urea formaldehyde resin synthesis is normally ended when the viscosity is optimal. Thus in the coating industry a knowledge of the viscosity of the binder is of considerable importance both from the manufacturing processes, pot stability and rate of cure of the paint film (Achi, 2003).

In our previous experiments (Osemeahon and Barminas, 2007; Barminas and Osemeahon, 2007), we reported both the synthesis of UF through a new synthetic route and the reactive blending of natural rubber with this new class of UF resin as a way of developing a paint binder for emulsion paint formulation from amino resin. In order to optimize the blending reaction between UF and NR, this study was set out to investigate the effect of UF viscosity on the UF/NR blend. This will offer formulators

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with varied options to tailor performance.

## MATERIALS AND METHODS

### Materials

Urea, formaldehyde, sodium dihydrogen phosphate, sulphuric acid, sodium hydroxide pellets and sucrose were reagent grade products from British Drug House (BDH). Natural rubber from *Hevea brasiliensis* (NG805) was obtained as exudates from Rubber Research Institute Benin City, Nigeria. All materials were used as received.

### Resin synthesis

The one-step- process as described by Barminas and Osemeahon (2007) was used in the preparation of trimethylol urea 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 (Chen et al., 2001). The pH of the solution was adjusted to 6 using 0.5 M H<sub>2</sub>SO<sub>4</sub> and 1.0 M NaOH solutions. The solution was then heated in thermostatically controlled water bath at 70°C. The reaction was allowed for 2 h after which the sample was removed and kept at 30°C. The samples with different viscosities used in this experiment were obtained by removing 100 ml of resin from the synthesized resin at 24 h intervals for the period of 144 h and their viscosities determined.

### Preparation of UF-NR composite films

Blend of UF and NR film was obtained as described earlier (Osemeahon and Barminas, 2007). In brief, 25 ml of natural rubber was added to 75 ml of UF (2.1 mPa.s) to form UF-NR composite blend. The moisture was stirred and left for 24 h at 30°C and then poured into a glass Petri dish for casting (Mirmohseni and Hassazeideh, 2000). The blend was also allowed to cure and set for seven days at 30°C. The above procedure was repeated at different UF viscosities (2.1 - 256.3 mPa.s). The physical properties of these films were then investigated.

### Determination of viscosity

The method reported by Barminas and Osemeahon, (2007) was adopted for the determination of the viscosity of UF resin. Five different readings were taken for each sample and the average value calculated.

### Determination of density, turbidity, melting point and refractive index

The above properties were determined according to standard methods from Official Method of Analysis International (AOAC, 2000). The density of the different resins was determined by taking the weight of a known volume of resin inside a density bottle using metler (Model, AT400) weighing balance. Five readings were made for each sample and average value calculated. The turbidity of the resin samples was determined by using Hanna microprocessor turbidity meter (Model, H193703) (Barminas and Osemeahon, 2006). The melting points of the different film samples were determined by using Galenkamp melting apparatus (Model, MFB600-010F). The refractive indices of the resin samples were determined with Abbe refractometer (Barminas and Osemeahon, 2006).

### Determination of moisture uptake

The moisture uptakes of the different resin film were determined gravimetrically (Osemeahon and Barminas, 2007). Known weight of the sample was introduced into a desiccator containing a saturated solution of sodium chloride. The increase in weight (wet weight) of the sample was monitored until a constant weight was obtained. The differences between the wet weight and dry weight of each sample were 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 desiccator test as reported by Kim (2001). The mold used was made from aluminium foil with a dimension of 69.6 x 126.5 mm (Kim, 2001) and thickness of 12.0 mm (Wang and Chen, 2002). The emitted formaldehyde was absorbed in 25.0 ml water and analyzed by a refractometric technique using Abbe refractometer. Triplicate samples were used and average value taken.

### Tensile test

Tensile properties (tensile strength and elongation at break) were measured as described by Wang and Chen (2002) using Instron Testing Machine (Model 1026). Resin films of dimension 50 long, 10 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 each sample and the average elongation evaluated and expressed as the percentage increase in length.

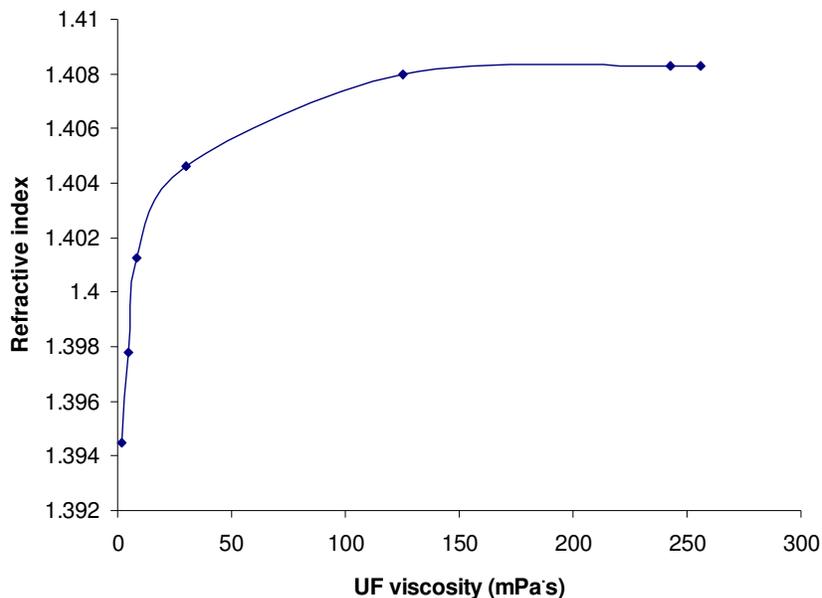
### Dry time and water solubility

The relative degree of cure (Reaction time) was expressed in the form of dry time (dry to touch). This was measured by the qualitative finger-making test (Ali et al., 2001). The solubility of methylol urea in water was obtained by mixing 1 ml of the resin with 5 ml of distilled water at 30°C (Osemeahon and Barminas, 2006).

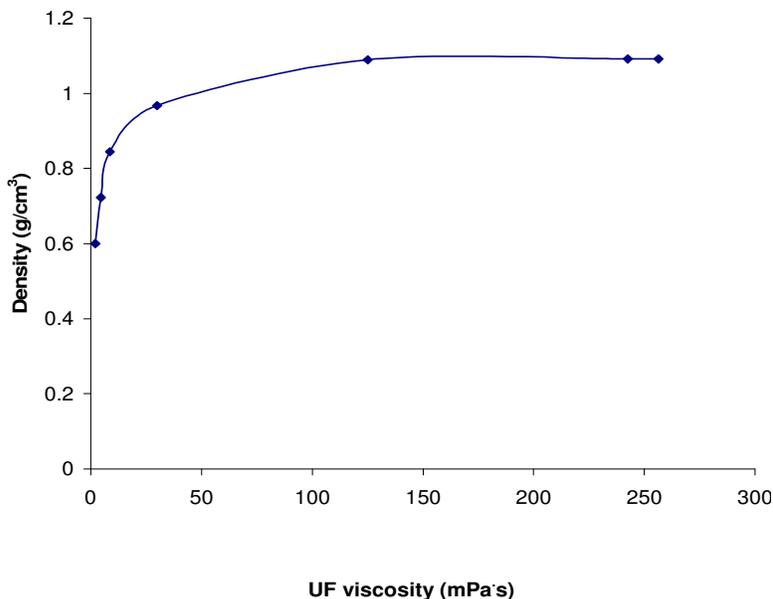
## RESULT AND DISCUSSION

### Refractive Index

Gloss is an important factor of many coating products (Osemeahon and Barminas, 2006b). The gloss of a paint coating with or without pigments is a function of the refractive index of the surface, the angle of incidence of the beam of light and the nature of the material (Trezza and Krochta, 2001). Figure 1 shows the effect of UF viscosity on the refractive index of UF-NR blend. It can be observed that the refractive index increased rapidly from 2 - 15 mPa.s viscosity levels. After this, little or no change in refractive index is observed with further increase in UF viscosity. This is attributed to the differences in molecular weight and cross link density of the different viscosities (Trezza and Krochta, 2001; Johnson and Wilkes, 2001). At the beginning, the molecular weight increased with increased in viscosity until an optimum growth was observed. At the gel point, the resin may be characterized by



**Figure 1.** Effect of UF viscosity on the refractive index of UF – NR blend



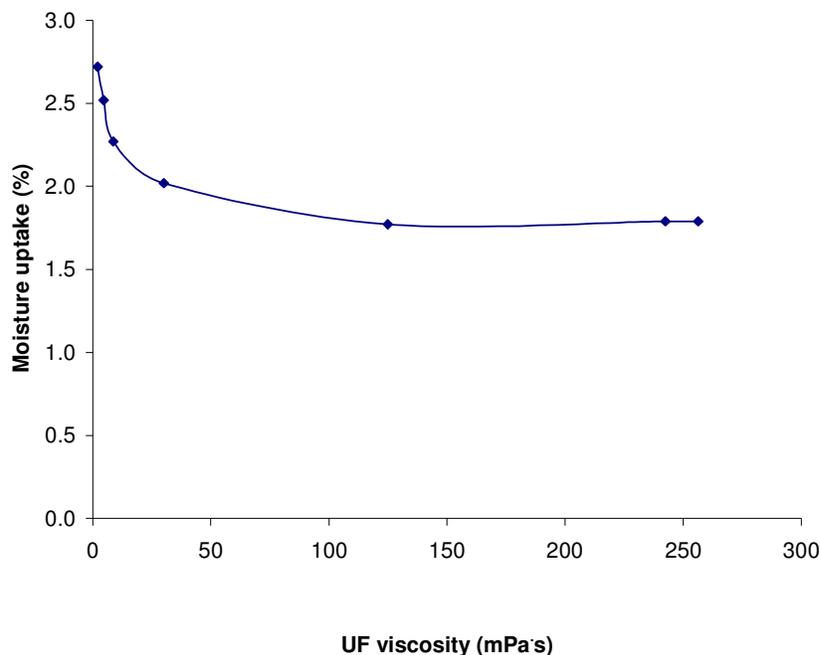
**Figure 2.** Effect of UF viscosity on the density of UF- NR blend.

molecular rearrangement and crosslinking of molecules. This gives an account of the plateau regime observed in Figure 1 (Nakason et al., 2001; Osemeahon and Barminas, 2006b)

### Density

In the coating industry, the density of the paint binder has profound influence on factors such as pigment disper-

sion, brushability of paint, flow, leveling and sagging (Lowel, 1990; Osemeahon and Barminas, 2006b). The effect of UF viscosity on the density of UF-NR blend is shown in Figure 2. The viscosity increased with increase in UF viscosity until the gel point. This was then followed by a constant regime with further increase in viscosity. The increase in density with increase in UF viscosity is due to increase in molecular weight while the constant regime may be attributed to gelation (Sekaran et al., 2001).



**Figure 3.** Effect of UF viscosity on the moisture uptake of UF- NR blend.

### Moisture uptake

The interaction of structural network of polymer resins with water is both of fundamental and technical interest (Osemeahon and Barminas, 2006a). Water uptake affects vital properties of the polymer material such as physical, mechanical, thermal and structural properties (Hu et al., 2001, Noguera et al, 2001). One of the major drawbacks of UF resins is their poor water resistance (Conner, 1996). In the paint making industry, the moisture uptake of the paint binder is very crucial because it is responsible for blistering and broominess of paint film (Barminas and Osemeahon, 2006).

Figure 3 shows the effect of UF viscosity on the moisture uptake of UF-NR blend. It is observed that the moisture uptake decreases with increase in UF viscosity and become constant beyond the gel points. This result can be explained in terms of the differences in crosslink density at different UF viscosity (Noguera et al., 2001; Hu et al., 2001). As the viscosity of UF increases, the molecular weight and hence crosslink density also increases until the gel point is reached after which the crosslink density remain constant. The higher the cross linkages the lower the void spaces available for moisture accommodation (Osemeahon and Barminas, 2006a).

### Melting point

The melting point of a polymer has a direct bearing to its thermal property (Bindu et al., 2001). It is related to its molecular weight, degree of crosslinking and the level of

rigidity of the polymer (Park et al., 2001). Figure 4 shows the effect of UF viscosity on the melting point of UF-NR blend. The melting point initially increased rapidly with UF viscosity and then plateau out at the gel point. This type of behaviour agrees with the report of Ma et al. (2002) and Markovic et al. (2001) which was attributed to differences in molecular weight and crosslink density. At the beginning, the molecular weight increased with increases in UF viscosity until optimum growth was obtained. Molecular rearrangement and crosslinking of resin molecules may account for the plateau regime (Osemeahon and Barminas, 2006a).

### Formaldehyde emission

One of the major disadvantages of urea formaldehyde resin is the emission of the hazardous formaldehyde during cure (Kim, 2001; El-Naggar et al., 2001; Pizzi et al., 2001). In the development of paint binder from urea formaldehyde resin, serious effort must be made to reduce formaldehyde levels to acceptable one (Barminas and Osemeahon, 2006a,b).

Figure 5 shows the effect of UF viscosity on formaldehyde emission of UF-NR blend. It can be observed that the formaldehyde emission increased with increase in UF viscosity. This trend can be ascribed to two reasons; firstly, it may be due to increase in the rate of condensation reactions with increase in UF viscosity thereby increasing the rate of emission of formaldehyde in the process (Nakason et al., 2001; Trumbo et al., 2001); Liang and Wang, 2001). Secondly, it may be due to increase in stress during resin cure with increase in UF resin viscosi-

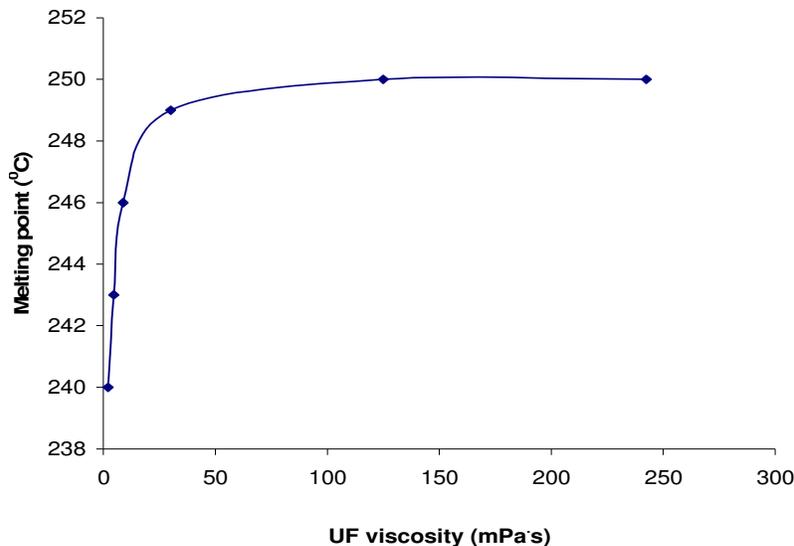


Figure 4. Effect of UF viscosity on the melting point of UF- NR blend.

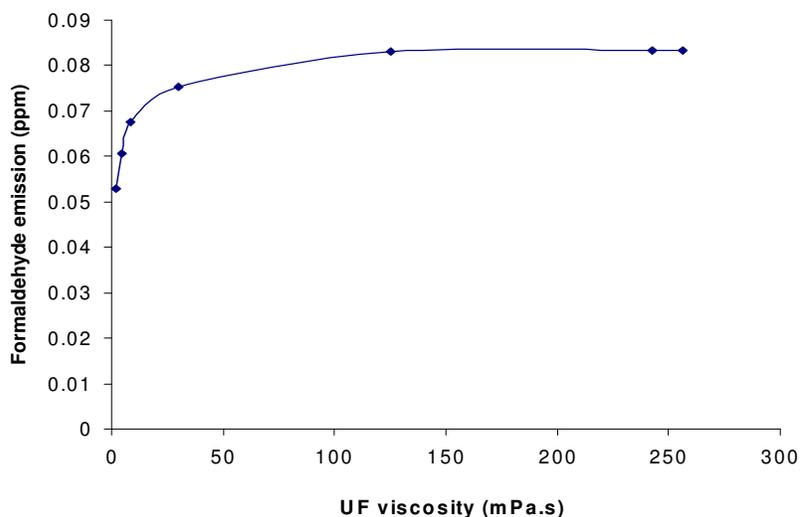


Figure 5. Effect of UF viscosity on the formaldehyde emission of UF- NR blend.

sity. Reduction in stress during cure reduces emission (Osemeahon and Barminas, 2006a). Low UF viscosity gives rise to low molecular weight which favours molecular chain mobility that enhances flexibility of polymer network; flexibility reduces stress during cure and reduction of stress reduces emission (Osemeahon and Barminas, 2006a; Chian and Yi, 2001).

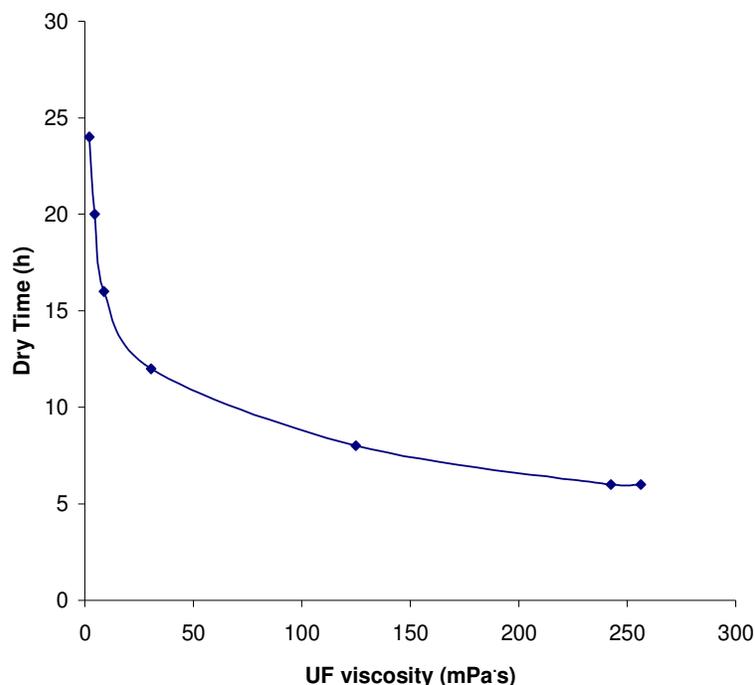
### Dry time

The time it takes for a paint to dry (reaction time) after application is an important factor for the paint formulator (Osemeahon and Barminas, 2006a). This is because if

the paint dries too fast, it will be prone to brittleness and if it dries too slowly, the paint film may be subjected to pick up dirt (Trumbo et al., 2001). Figure 6 shows the effect of UF viscosity on the dry time of UF-NR blend. The result showed that the dry time decreased with increase in UF viscosity in the blend. This can be explained in terms of increase in molecular weight and crosslink density with increase in UF viscosity (Lowel, 1990).

### Tensile test

Elongation at break determines to what extend a material stretches before breaking and hence the ductility of flexi-



**Figure 6.** Effect of UF viscosity on dry time of UF- NR blend.

**Table 1.** Effect of UF viscosity on the tensile strength and elongation at break of UF-NR blend.

Viscosity (mPa·s)	Tensile strength (kg/cm <sup>3</sup> )	Elongation at break (%)
2.10	0.021	440
4.57	0.041	400
8.73	0.061	365
30.55	0.078	320
125.00	0.101	280
142.50	0.106	285
256.3	0.106	285

bility or the material. One of the shortcomings of UF resin is that it is too hard and brittle and poor resistance to crack propagation (Lowel, 1990). In the coating industry, a paint binder must be able to withstand stress emanating from variation in environmental factors. Therefore, in developing paint binder from amino resin, tensile property such as elongation at break must be taken into consideration (Osemeahon and Barminas, 2006a).

The effect of UF viscosity on the tensile strength and elongation at break are shown in Table 1. It is observed that the tensile strength decreases while the elongation at break increases with increase in UF viscosity. This trend can be explained in terms of the increase in molecular weight and hence crosslink density of the UF-NR blend with increase in UF viscosity (Tai and Li, 2001; Ma et al., 2002). Differences in crystallinity or crystalline orientation of resin molecules with increase in UF viscosity may also

be responsible for this result (Xie et al., 2001; Chain and Yi, 2001).

### Solubility in water

In the development of amino resin for emulsion paint formulation, the solubility of the resin in water is paramount (Osemeahon and Barminas, 2006a). It is important both from the technical and processing point of view. This is more so because the solubility of urea formaldehyde resin decreases with increase in viscosity (Lowel, 1990; Park et al., 2001). Table 2 shows the effect of UF viscosity on solubility of UF-NR blend in water. Below a viscosity of 30.55mPa.s, the UF-NR blend is soluble and beyond this point the resin is insoluble in water. This result is attributed to differences in molecular weight and

**Table 2.** Effect of UF viscosity on the water solubility of UF-NR blend.

Viscosity (mPa.s)	Solubility properties in water
2.10	Soluble
4.57	Soluble
8.73	Soluble
30.55	Slightly soluble
125.00	Insoluble
242.50	Insoluble
256.3	Insoluble

crosslink density (Lowel, 1990). Perhaps, the viscosity of 30.55 mPa.s seems to represent the gel point of the resin. Thus processing of UF-NR for emulsion paint formulation could be suggested below this viscosity value.

## Conclusion

This study examined the effect of UF viscosity on some physical properties of UF-NR blend. The results obtained shows that UF viscosity has a significant influence on the properties of UF-NR blend. A viscosity below 30.55 mPa.s is soluble in water and beyond this value the composite became insoluble in water. Thus processing of UF-NR blend for emulsion paint formulation could be suggested below this viscosity level. While the level of formaldehyde emission was found to increase with increase in UF viscosity why that of moisture uptake on the other hand decreased with increase in UF viscosity. Thus this study will contribute immensely toward the optimization of the reactive blending of UF with NR.

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