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The effect of some physicochemical factors on the stability of arachis oil emulsion formulated with Nigerian type gum arabic

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ABSTRACT

This study aimed at evaluating effects of some physicochemical factors on stability of arachis oil emulsions formulated with gum arabic from Nigeria. Grade One Nigerian type gum arabic was processed from two States in northern Nigeria. Arachis oil emulsions were prepared using these gums as emulsifying agents. Another batch of emulsion formulated using imported gum arabic served for comparison. Globule size, number and effect of ageing were determined. Effect of pH, electrolyte, temperature, anti-oxidants, blending, and atmospheric conditions on the viscosity of the emulsion was determined. The physical appearance of the emulsions did not change after 6 months storage but there was decrease in viscosity. The globule size of emulsions containing the locally sourced gum was smaller ($p<0.05$) than that of the emulsion formulated with the imported gum after 24 h. After 6 months, the globule sizes of both emulsions did not differ markedly from each other ($p>0.05$) and a comparable rate of coalescence ($p>0.05$) between both emulsions was observed. The effect of pH, electrolyte, temperature, anti-oxidants, blending at high speed and atmospheric conditions on the viscosity of both batches was comparable. On the basis of these, the locally sourced gum arabic can be used to formulate arachis oil emulsions.

Key Words: Gum Arabic, emulsions, physicochemical factors, Nigeria

INTRODUCTION

Gum arabic, also known as acacia gum is a natural gum obtained from the hardened sap which flows naturally from or obtained by incision of the stems and branches of two species of the acacia tree; *Acacia senegal* and *Acacia seyal.* The gum is harvested commercially from acacia trees found throughout the Sahel from Senegal and Sudan to Somalia [1, 2]. The gum is odourless and tasteless and adheres to the tongue when placed in the mouth. The unprocessed gum is completely but very slowly soluble in cold water. Aqueous solutions of the gum are viscous and Newtonian in character. This rheological behaviour of the gum makes it a good emulsifying agent [3, 4].

Emulsions are biphasic systems of two immiscible liquids one of which is uniformly dispersed through the other as droplets of diameter greater than 0.1µm. Emulsions contain both a dispersed and a continuous phase, with the boundary between the phases called the interface. They tend to have a cloudy appearance due to the scattering of light which passes through the emulsion by the interfaces and are generally unstable. The particle diameter of the disperse phase of a macroemulsion extends from 0.1 to 100 µm [5]. Due to the thermodynamic instability of emulsions, when left to stand for a long period of time, they tend to revert back to the stable state of the phases which made up the emulsion [6]. The difference in the stability profiles of pharmaceutical emulsions is related to the globule sizes. The higher the mean droplet diameter (MDD) of the dispersed phase, the greater the tendency to experience instability. It has indeed been demonstrated that when the diameter of the oil globules within an emulsion was doubled, the rate of creaming increased by a factor of four. The increase in globule size has also been associated with a decrease in viscosity [7]. Emulsions with large globule sizes are usually those formed with low shear conditions [8]. The system which is thermodynamically unstable is stabilized by the presence of an emulsifying agent. The mechanism of action of emulsifying agents depends upon the formation of film at the interface of two phases. Emulsion stability is a very important attribute of any given formulation [9, 10]. It refers to the ability of an emulsion to resist

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changes in its properties over time. There should be no appreciable change in the mean particle size or the size distribution of the droplets of the dispersed phase. Secondly, droplets of the dispersed phase should remain uniformly distributed throughout the system. Pharmaceutical emulsion stability is characterized by the absence of coalescence of the dispersed phase, absence of creaming and the ability to retain the physical characters such as elegance, odour, colour and appearance [11]. Some factors which may affect the stability of emulsions include addition of an incompatible emulsifying agent, chemical or microbial decomposition of emulsifying agent, addition of electrolytes, exposure to increased or reduced temperature and change in pH.

Gum arabic acts as an emulsifier that stabilizes oil-in-water emulsions and the Nigeria type gum has been investigated for its emulsifying properties [1, 4]. The aim of this study was to evaluate the effects of some these factors on the stability of arachis oil emulsions stabilized with gum arabic grown and processed in Nigeria.

MATERIALS AND METHODS

Materials: The following materials were used as procured from their manufacturers; gum arabic (Sigma-Aldrich, Germany), Two samples of Grade 1 gum arabic obtained from Kazaure in Jigawa State and Bukar abba Ibrahim Farms near Damaturu in Yobe State of Nigeria. All other reagents were of analytical grade.

Methods

Purification of Crude Gums: The crude gum arabic from Jigawa and Yobe States of Nigeria were purified according to the method reported by Anukam and Okafor [4].

Preparation of emulsion: The formula for preparing batches of emulsions using the dry gum method is presented in Table 1. A 30 ml quantity of arachis oil was transferred into a porcelain mortar. Then 7.5 g of gum arabic was dispersed properly in the oil using a pestle. A 15 ml quantity of distilled water was poured into the mortar all at once followed by vigorous trituration in only one direction. Upon formation of the primary emulsion, the emulsion was diluted with 10 ml of distilled water. A 0.18 g and 0.09 g quantity of methyl and propyl parahydroxybenzoate were respectively added in 30 ml of distilled water and added to the dilute emulsion in the mortar. The content of the mortar was poured into a 100 ml measuring cylinder and the final volume made up with distilled water.

Evaluation of the emulsion

Physical appearance: The colour, odour and texture of the emulsions were observed immediately after preparation. This was repeated after 6 months.

Measurement of viscosity: A 30 ml quantity of the emulsion was transferred into a 50 ml Pyrex beaker. The viscometer spindle (size- 62) (DV-I Prime Viscometer, Brookfield Engineering Laboratories Inc. U.S.A) was used. The spindle was rotated at 100 rpm for 5 min at a torque >10 % and temperature of 26±1 ^{0}C and its viscosity was measured. This measurement was taken 48 hours after formulation and after 6 months of storage.

Table 1: Formula for preparing arachis oil emulsion containing gum arabic as an emulsifier

Batches (g or ml) **Ingredients**	**I**	**II**	**III**	**IV**
Arachis oil	30	30	30	30
Gum arabic JG	7.5	-	-	-
Gum arabic YB	-	7.5	-	-
Gum arabic ST	-	-	-	7.5
Methyl parahydroxybenzoate	0.18	0.18	0.18	0.18
Propyl parahyroxybenzoate	0.09	0.09	0.09	0.09
Distilled water to	100	100	100	100

Key: JG = Jigawa State, YB= Yobe State, ST= Standard

Determination of globule size and number: The globule size and number of the emulsion were determined according to the method reported previously [4]. The number of globules was estimated according to the equation [12]:

$$N = \frac{6\varphi \times 10^{12}}{\Pi (Dm)^3} \quad \text{.............................. Eq 1}$$

where:
N = no of globules per ml
Dm = mean diameter of globules
φ = volume fraction of the dispersed phase (quantity of oil in 0.1 ml of emulsion)

Determination of rate of globule coalescence: The rate of globule coalescence of the emulsion was calculated using the Sherman equation [12];

$\ln D_t = \ln D_O + Kt/3$ .... Eq 2

Where:

D_t= the mean diameter of globules at time t

D_o= the mean diameter of globules at time 0

K= rate of coalescence of globules

Determination of pH of the emulsion: The pH of the emulsion was determined using a pH meter (Model 3310, Jenway Ltd Uk). The electrode arm of the pH meter was first standardized in a 7.4 buffer solution. It was dipped into 100 ml of the emulsion and the pH was read off after 2 min. The average of three readings was recorded.

Effect of pH on the viscosity of emulsions: To each batch of emulsion a 1.0 ml of 1 M hydrochloric acid or sodium hydroxide was added before the final volume was made up. The emulsions were kept for 24 h. The pH and viscosity were determined as described earlier. The average of three determinations was recorded in each case.

Effect of electrolyte on emulsion viscosity: A 1.0 ml quantity of 0.5 %w/v sodium chloride, barium chloride or aluminium chloride solution was separately added to each batch of emulsion before the final volume was made up. The viscosity was determined 24 h after the emulsions were made. The mean of three determinations was recorded.

Effect of temperature on the viscosity of the emulsion: Four batches of emulsions were formulated with each of the three gums. A refrigerator fitted with a thermometer was used to maintain the temperature of the first batch of emulsions at 12 $\pm 1^0$C. The other three batches were placed in water baths maintained at 30, 50 and 70 $\pm$ 1^0C. A thermometer was inserted into each of the emulsions to ensure the products attained the temperatures. The viscosity of the emulsions was determined at the stated temperatures after 1 h. The average of three determinations was taken.

Effect of antioxidants: To each batch of emulsion, a 1ml quantity of 0.1 $\%^w/v$ sodium metabisulphite was added before making up the 100 ml volume. The viscosity was measured after 6 months of storage on a vibration free rack. The average of three determinations was recorded

Effect of blending: The formulated emulsion was blended for 1 min in a laboratory blender (Model 38 BL 40 Warling Products). The viscosity was determined immediately after blending and subsequently after six months of storage. The average of three determinations was recorded

Effect of atmospheric conditions: A 100 ml quantity of the freshly prepared emulsion from each batch was placed in an open beaker of known dimensions and left on a vibration free rack for one week. Water was added to make up to 100 ml (due to loss of moisture by evaporation). The emulsion was poured back into an amber coloured bottle and kept on a rack for 6 months after which the viscosity was determined. The average of three determinations was recorded

RESULTS AND DISCUSSION

The raw gum exudates were tasteless and odourless nodules which adhere to the tongue and are difficult to crush. The colour of the raw gum was pale yellow. After purification, clarification and size reduction, the gum arabic obtained from Jigawa (JG) and Yobe (YB) States of Nigeria were off-white powders. The physical appearance of the gums is similar to the imported gum arabic (ST) which is used as the basis for comparison.

The physical properties of arachis oil emulsion: Table 2 shows the physical properties of the emulsions. The emulsions formulated with the various gum arabic were off-white and creamy. The viscosities were 27.9, 27.6 and 22.5 mPas for emulsions containing JG, YB and ST gums respectively. No visible change in colour was detected after six months of storage. The emulsions however became less creamy by the end of the six month storage period. The viscosities after six months storage were 14.7, 14.4 and 13.7 mPas for emulsions containing JG, YB and ST gums respectively. Gum arabic like many other gums consists of polysaccharides which form viscous solutions when brought into contact with water. When kept in storage however, the viscosities of such solutions tend to reduce with time [13].

Globule size and number determination and rate of globule coalescence: Table 2 shows the globule size, number and rate of globule coalescence of the emulsions formulated with various gums 24 h and 6 months after storage. The globule size of the emulsions containing the different gums did not differ markedly from each other after 24 h of preparation but the globule size of the emulsion containing the ST gum was significantly ($p<0.001$) larger. The mean globule size of the emulsions formulated with JG, YB and ST gums are 3.65, 3.51 and 5.59 µm respectively after 24 h. The corresponding mean globule size values of the emulsions after 6 months of storage are 6.90, 6.66

and 8.05 µm. The globule size of emulsions containing JG and YB gums increased in size to a less extent than that containing the ST gum after 6 months of storage, but the difference in their globule sizes after 6 months was not significant ($p>0.05$). The globule numbers of the emulsions after 24 h are 1.1, 1.3 and 0.3 X 10^{10} globules per millilitre of emulsions containing JG, YB and ST gums respectively. The corresponding globule numbers of emulsions containing JG, YB and ST gums after 6 months of storage are 1.7, 1.9 and 1.0 X 10^{9} globules per millilitre. A general reduction in the number of globules occurred in all the emulsions after 6 months. The magnitude of reduction appeared to be the same for all the emulsions. The rate of coalescence followed a similar pattern as the globule number. The rates of coalescence of the globules of emulsion containing JG, YB and ST gums are 0.74, 0.74 and 0.76 per month respectively. The stability of an emulsion is highly dependent on the size of globule achieved. The smaller the size of oil globule, the more stable the emulsion. The globule size increased on storage as a result of coalescence, which is the attempt of the disperse phase in the emulsion to achieve stability (Sherman, 1968; Khan *et al.*, 2011).The increase in globule size has been associated with a decrease in globule number as well as a corresponding decrease in viscosity [8].

Table 2: Effect of storage on emulsion properties

Property	**Batches**					
	JG		**YB**		**ST**	
	24h	**6mths**	**24h**	**6mths**	**24h**	**6mths**
Appearance:						
Colour	Off-white	Off-white	Off-white	Off-white	Off-white	Off-white
Odour	Non rancid	Non rancid	Non rancid	Non rancid	Non rancid	Non rancid
Texture	Creamy	Less Creamy	Creamy	Less Creamy	Creamy	Less Creamy
pH +Hydrochloric	4.26	-	4.30	-	4.09	-
acid	3.33	-	3.35	-	3.28	-
+Sodium hydroxide	4.41	-	4.45	-	4.41	-
Viscosity (mPas) at 100rpm shear rate	27.9	14.7	27.6	14.4	22.5	13.7
Globule size (µm)	3.65±0.191	6.90±0.615	3.51±0.155	6.65±0.547	5.59± 0.512	8.04±0.694
Globule number/ml	1.1 x 10^{10}	1.7 x 10^{9}	1.3 x 10^{10}	1.9 x 10^{9}	0.3 x 10^{10}	1.0 x 10^{9}
Rate of Coalescence/mth	-	0.74	-	0.74	-	0.76

Table 3: Effect of some factors on the viscosity of the emulsion

	Viscosity (mPas) at 100rpm shear rate		
	JG	**YB**	**ST**
Effect of pH			
+ HCl	24.75	25.45	19.35
+ NaOh	30.60	27.90	23.32
Effect of electrolyte			
+NaCl	25.95	26.55	21.75
+BaCl$_2$	24.60	23.85	18.38
+AlCl$_3$	24.45	26.25	18.15
Effect of antioxidant			
24 h	27.9	27.6	22.5
6 mths	24.5	26.0	25.9
Effect of blending at high speed			
24 h			
6 mths	31.05	33.15	24.45
	22.40	25.00	20.20
Effect of atmospheric conditions			
24 h	27.9	27.6	22.5
6 mths	14.7	16.6	15.1

Effect of pH on the viscosity of emulsion: The values of the pH of the emulsions containing JG, YB and ST gum arabic without addition of acid or alkali were 4.26, 4.30 and 4.09 respectively. The viscosities of emulsions containing the respective gums were 27.90, 27.60 and 22.50 mPas without the addition of acid or alkali. In the presence of hydrochloric acid, the pH of the emulsions fell to 3.33, 3.35, and 3.28 for emulsions containing JG, YB and ST gums respectively. The viscosities reduced to 24.75, 25.45 and 19.35 mPas for the respective gums. In the presence of sodium hydroxide, both the pH and the viscosities of the different emulsions increased in all the batches. All the formulated emulsions were slightly acidic. This is attributable to the presence of uronic acids within the structure of gum arabic. The high viscosity values observed on addition of sodium hydroxide may be due to the fact that the optimum pH of gum arabic lies between 4 and 6 [14] and the addition of sodium hydroxide brought the emulsion within this pH range.

Effect of electrolytes on the viscosity of emulsion: The viscosity of the emulsion containing JG, YB and ST gum upon addition of sodium chloride had respective mean values of 25.95, 26.55 and 21.75 mPas. These values are lower than the viscosity values obtained without the addition of any electrolyte to the formulated emulsion. A similar decrease in viscosity was observed across the different batches of formulated emulsions as barium chloride or aluminium chloride was added. The addition of electrolytes of different valencies causes general decrease in viscosity. The decrease in viscosity is proportional to the valency of the cation [15]

Effect of temperature on the viscosity of emulsion: Figure 1 shows the effect of temperature on the viscosity of emulsions containing JG, YB or ST gum arabic. It is seen that the viscosity decreased as the temperature increased in all the batches. This agrees with the observation made by [16], which stated that increase in temperature decreases both density and viscosity of a polymer solution. The emulsion containing JG and YB gums maintained a higher viscosity than the emulsion containing ST gum at all temperatures.

Effect of addition of anti-oxidant on the viscosity of ageing emulsion: The viscosity of all the emulsion batches formulated with JG, YB or ST gums had a higher value when an anti-oxidant was included in the formulation than those in which no anti-oxidant was added even after 6 months of storage. The emulsions containing JG and YB gums had mean viscosity values of 24.5 and 26.0 mPas respectively, while the emulsion containing the ST gum had a mean value of 25.9 mPas. Oxidation is a common chemical instability experienced by emulsions. Many fats and oils used for emulsions are susceptible to oxidation. This includes emulsified lipids which are particularly sensitive to oxidation. Many drugs incorporated into emulsions also have this challenge which results in decomposition of the drug. Antioxidants are therefore a very important additive in oil-in-water emulsions [5, 14]

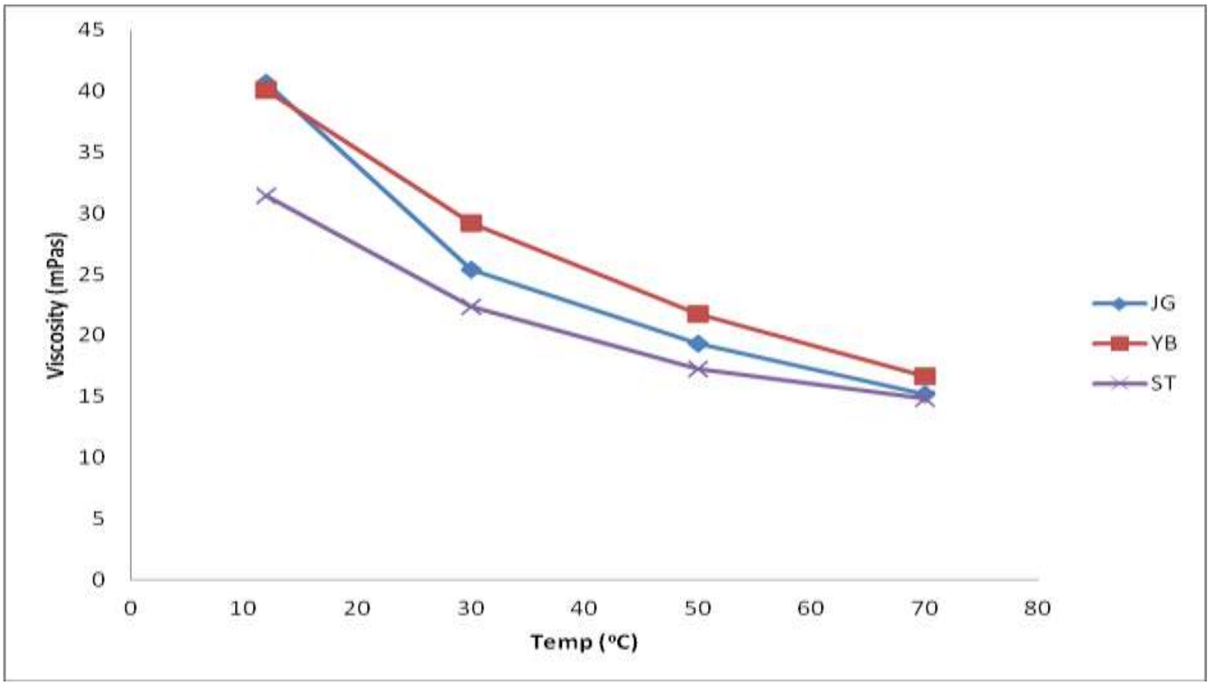

Figure 1: The effect of temperature on the viscosity of formulated arachis oil emulsions

The addition of anti-oxidant deterred oxidation of the arachis oil which is evident in the comparatively high viscosity observed at the end of the 6-month storage period The prevention of this kind of instability is therefore a useful way of preserving emulsions

Effect of blending at high speed on the viscosity of emulsion: Blending of the different batches of emulsions containing JG, YB or ST gum resulted in higher viscosity values after 24 h or 6 months than the emulsions which were not blended. The viscosity of the blended emulsions formulated with JG, YB or ST after 24 h and 6 months are 31.05 and 22.40, 33.15 and 25.00, and 24.45 and 20.20 respectively. The effect of blending the emulsion at a high speed brings to light the effect of different process variables on the emulsion stability. Stirring intensity and diameter of dispersed particles affect the eventual stability of the formulated emulsion [11]. Blending at high speed causes a decrease in the mean droplet diameter (MDD) which has a corresponding increase in the viscosity. Additionally, it has been reported that emulsions with large globule sizes are those usually formed with low shear conditions [8].

Effect of atmospheric conditions on viscosity of emulsion: The effect of atmospheric conditions on the viscosity of the formulated emulsions when compared with the different gums showed a general reduction in the viscosity of the emulsions. However, no marked difference was observed in the viscosity of the emulsions when compared with that of the other batches of emulsions that were not subjected to this test but kept for the same total storage period of 6 months. It can be inferred that exposing the emulsion to the atmospheric oxygen had no further noticeable effect on the viscosity of the emulsion formulated with the JG, YB and ST gum apart from the effect of ageing on viscosity.

CONCLUSION

The Nigeria type gum arabic has a great potential as an emulsifying agent in the formulation of oil-in-water emulsions. Both locally sourced gums formed emulsions with good properties comparable to the imported gum arabic. Emulsions containing the gums showed ability to stabilize an arachis oil emulsion against changes in pH, salt effect, temperature and other changes. Further reports will include application of the Nigerian type gum arabic as a tablet binder.

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