

The Emulsifying Properties of *Terminalia Randii* Baker F. Gum in Castor Oil and Liquid Paraffin Emulsions

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A – research concept and design; B – collection and/or assembly of data; C – data analysis and interpretation; D – writing the article; E – critical revision of the article; F – final approval of article.

Abstract

Background: Emulsifying agents are added to preparations to facilitate the process of emulsification during manufacture and also to assure the stability of emulsion during the shelf life of the product. Natural gums have been used as emulsifying agents because they are readily available, non-toxic and cheap.

Objectives: In this study, the emulsifying properties of *Terminalia randii* gum were assessed and compared with a standard emulsifier (Tragacanth gum) using castor oil and liquid paraffin .

Method: Different concentrations (1-10% w/v) of the mucilages of *Terminalia randii* gum and Tragacanth were prepared. Using wet gum method, castor oil (fixed oil) was mixed with the mucilage and methyl paraben was added. Distilled water was used to make up to volume. The procedure was repeated for liquid paraffin (mineral oil). The emulsions were assessed using creaming rate, globule size and viscosity.

Results: Castor oil and liquid paraffin emulsions containing *Terminalia* gum did not show any form of creaming at high concentration (10% w/v). At low concentration (1-2% w/v), the emulsions cracked within 24 hours of preparation. There was no significance ($p > 0.05$) difference in the globule sizes of emulsions containing *Terminalia* or tragacanth gum. The viscosity of emulsions containing *Terminalia* gum increased with increase in concentration of emulsifier and time. It was observed that after one week (168 hours) the viscosity of emulsions containing tragacanth started decreasing and this coincided with the time the emulsions started to cream or eventually cracked.

Conclusion: *Terminalia randii* gum produced more stable emulsions and thus could be used as alternative emulsifying agent.

Keywords: *Terminalia randii* gum, Emulsifying agent, Castor oil, Liquid Paraffin, Emulsions

INTRODUCTION

Pharmaceutical excipients are substances other than the Emulsions are thermodynamically unstable system consisting of two immiscible liquids that can be stabilized by the addition of an emulsifying agent (Anukam *et al.*, 2015). Instability in emulsions can be physical: such as creaming (reversible), cracking (irreversible), phase inversion (irreversible) and flocculation (reversible). Emulsifying agents stabilise emulsions by forming protective sheaths around emulsion droplets or by imparting a charge on the dispersed phase. Some of these emulsifying agents are also known to increase viscosity of the emulsion. Examples of natural emulsifying agents that has been used in emulsion formulations are gums, such as angum gum (Golkar *et al.*, 2015), gum Arabic (Ma *et al.*, 2015).

Natural gums are polysaccharide hydrocolloids, exudates or slims that are pathological products but do not form part of cell wall (Prajapati *et al.*, 2013). They have been used in pharmaceutical formulations as emulsifying agents (Amalesh *et al.*, 2010; Golkar *et al.*, 2015), suspending agents (Rishabha *et al.*, 2010; Sumedha *et al.*,

2015; Wang 2016), binders in tablet formulations (Olayemi and Jacob, 2011; Mistry *et al.*, 2014), novel drug delivery systems (Ganie *et al.*, 2015; Sharma *et al.*, 2016) and they are also used in the food industries as stabilizers and food thickeners. These natural gums are being exploited for use in the pharmaceutical industry because they are non-toxic, not expensive and biodegradable.

The search for new pharmaceutical excipient led to this study, in which *Terminalia randii* gum which has been evaluated as a binder (Bamiro *et al.*, 2010), a directly compressible excipient for controlled release (Bamiro *et al.*, 2012), a suspending agent (Bamiro *et al.*, 2014) will be evaluated for its suitability as an emulsifying agent in castor oil and liquid paraffin emulsions.

MATERIALS AND METHODS

Castor oil and Liquid paraffin oil were purchased from Tunnex Laboratory Engineering, Nigeria Ltd. Tragacanth powder (BDH Chemicals Ltd, England), *Terminalia randii* gum was collected from Olabisi Onabanjo

University Ago-Iwoye, Nigeria and processed in the Pharmaceutics & Pharmaceutical Technology laboratory. All other materials used were analytical grade.

Extraction of *Terminalia* Gum

The trunk of the tree *Terminalia randii* was incised and the gum exudates were allowed to dry and then hand-picked from the trees. The dried gum was washed and dried in hot air oven at 40°C for 48 h. The gum was then crushed to break up the gum and then hydrated in double strength chloroform water for 5 days with intermittent stirring. The mucilage obtained was strained through a clean calico cloth and the gum was precipitated with 95% w/v ethanol. The precipitated gum was filtered, washed with diethyl ether and then dried in hot air oven. The dried gum was pulverized and passed through sieve size No 60 (250 µm) (Bamiro *et al.*, 2010).

Preparation of Emulsions

Emulsions were prepared using wet gum method. The method of Srinu and Kishore (2012) was used, though this was slightly modified. 1 part of the gum (*Terminalia* or tragacanth) at the different concentrations (1 – 10% w/v) was mixed with 2 parts of the water to form the mucilage. 4 parts of castor oil (40% w/v) was added gradually to the mucilage by mixing in a mortar. Methyl paraben (0.13% w/v) was added. The primary emulsion formed was made up to volume with water. Liquid paraffin emulsion was also prepared using the same method but 3 parts of oil was used.

Evaluation of Emulsion

Determination of Emulsion Type

Dye staining test was used to determine the emulsion type (Samanta *et al.*, 2010). About 5 mL of emulsion was mixed with a drop of methylene blue, a drop of the emulsion was now placed on a slide and viewed under the microscope.

Determination of globule size

The globule sizes were determined by measuring about 100 globules under a calibrated microscope. The sizes were determined at different time intervals (0, 2, 3, 4, 24, 72, 168 and 336 hours).

Determination of creaming

30 mL of each emulsion were poured in bottles and kept without disturbance for 5 weeks. The separated phase was measured at different time intervals (0, 2, 3, 4, 24, 72, 168, 336, 504 and 672 hours). This was done in triplicate.

Determination of viscosity

The viscosities of the emulsions were determined with a dial Brookfield's Viscometer, Model LV. Spindle size 4 was used at a speed of 30 rpm. 100 mL of emulsion was poured into a beaker and the viscosities were determined at different time intervals (0, 2, 3, 4, 24, 72, 168, 336, 504 and 672 hours).

Statistical Analysis

Statistical analysis was done to compare the effect of the different binders on the tablet properties using ANOVA (GraphPad Software Incorporation, San Diego, USA). At 95% confidence interval, p values of less than 0.05 were considered significant.

RESULTS AND DISCUSSION

Emulsion Type

Different types of tests such as dye staining, miscibility, conductivity can be used for determining emulsion type (Samanta *et al.*, 2010). In this study, the staining test was used by adding Methylene blue to the emulsion which gave a blue continuous phase with colourless globules in the dispersed phase when viewed under the microscope. Methylene blue being water soluble, dissolved in the aqueous phase of the emulsion, which is the continuous phase thereby indicating that the emulsion is oil-in-water (o/w).

Globule Size

Average globule size of emulsions containing 10% w/v emulsifying agents are presented in Table 1. In this experiment, the average globule size at different time interval was calculated from the globule size distribution obtained. The globule size of emulsions formed was between 9.01-21.63 µm. In most pharmaceutical emulsions, the globule size of dispersed phase is usually between 0.1 -10 µm, though it can be as high as 100 µm (Mahato and Narang, 2011). The globule sizes of emulsions prepared were within the size limits specified for pharmaceutical emulsions. The globule sizes were observed to increase with time. This could be due to coalition of the oil globules. Changes in globule size is one of the key indicators of loss of stability in emulsions (Silva *et al.*, 2010; Khan *et al.*, 2011). There was no significance (p>0.05) difference in the globule sizes of emulsions containing *Terminalia* and tragacanth gums. The globule size distribution of an emulsion gives a statistical inventory of the dispersed phase fragmentation. This is very valuable in practice since the stability and viscosity of emulsions depends on the globule size distribution (Jean-Louis, 2000). Stability of an emulsion can be enhanced by reducing globule size (McClements, 2005).

Creaming rate

In creaming, the dispersed phase separates out and stays at the top of the emulsion if it is oil in water emulsion (Khan et al., 2011) and the emulsion can still be re-dispersed by shaking. When emulsion cream upwards, as was the case with these emulsions, the oil globules which are the internal phase, tends to stay at the top of the emulsion because they are denser. The cream height was observed to increase with time and there was significant ($p < 0.05$) decrease with increase in concentration of emulsifying agents (Figs. 1-4). This decrease could be due to increase in concentration of emulsifying agent, which invariably increased the viscosity of the emulsions. There was no significance ($p > 0.05$) difference in the creaming rate of castor oil emulsion emulsions containing either *Terminalia* or tragacanth gum at high concentrations (5-10% w/v) but at low concentrations, there was significant ($p < 0.05$) difference. There was significance ($p < 0.05$) difference in the creaming rate of liquid paraffin emulsions containing either *Terminalia* or tragacanth gums. This difference in creaming rate could also be due to difference in the densities of the aqueous and oil phase, the globule size and viscosity of the emulsions

Viscosity

Viscosity of an emulsion is affected by the amount of emulsifying agent present in the emulsion. The viscosity of the emulsions was between 5670 – 13000 cP. Emulsions containing *Terminalia* gum has higher viscosities than those containing tragacanth gum. Increase in concentration of emulsifying agents led to increase in viscosities of the emulsions which caused a reduction in the rate at which globules coalesce and also reduction in creaming. The viscosities of emulsions containing *Terminalia* gum were observed to increase with storage time irrespective of the oil type, while the viscosities of emulsions containing Tragacanth gum increased initially before it started to decrease (Figs 5-6). This increase in viscosity could have been responsible for the stability of emulsions containing *Terminalia* gum. Ye et al (2004) also found that xanthan gum increased the viscosity of emulsions thereby reducing the mobility of the globules and their coalescence.

Table 1: Globule sizes of emulsions containing 10% w/v gum

| Globule size (μm) | | | | |
|--------------------------------|-----------------------------------|--|----------------------------|---------------------------------|
| Time (Hours) | Castor oil+ <i>Terminalia</i> gum | Liquid paraffin+ <i>Terminalia</i> gum | Castor oil+ Tragacanth gum | Liquid paraffin+ Tragacanth gum |
| 0 | 9.03 | 10.54 | 10.85 | 11.05 |
| 2 | 10.16 | 12.58 | 12.04 | 11.79 |
| 3 | 11.64 | 13.43 | 12.68 | 12.41 |
| 4 | 13.31 | 13.44 | 13.79 | 14.42 |
| 24 | 16.25 | 13.45 | 15.99 | 15.04 |
| 72 | 16.55 | 15.20 | 17.60 | 16.99 |
| 168 | 18.00 | 15.36 | 17.66 | 18.25 |
| 336 | 19.21 | 15.83 | 21.63 | 21.04 |

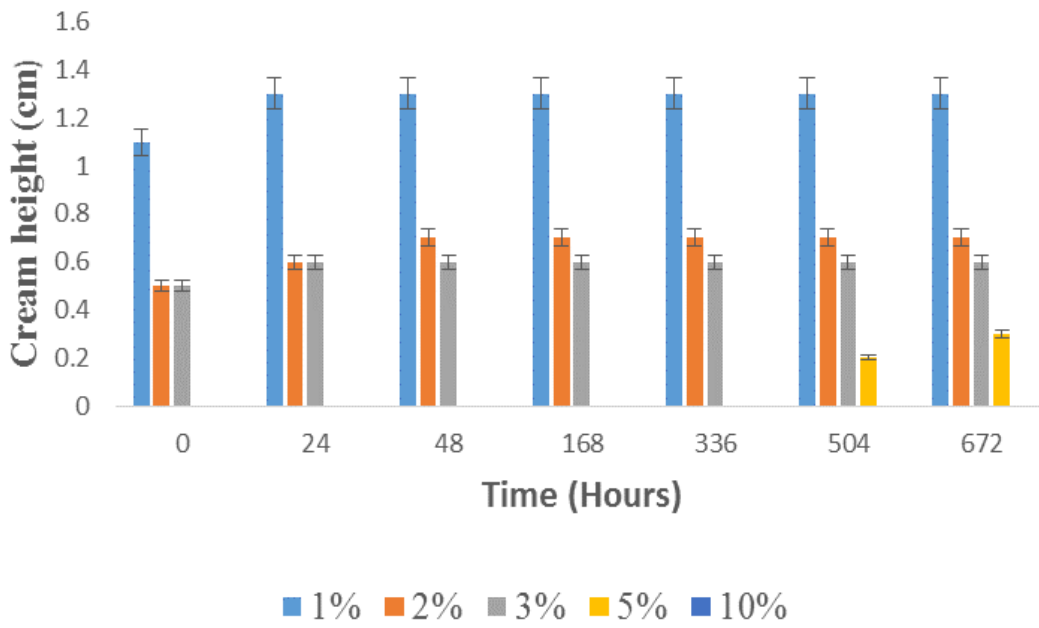


Figure 1: Plot of creaming height against time for castor oil emulsions containing *Terminalia* gum

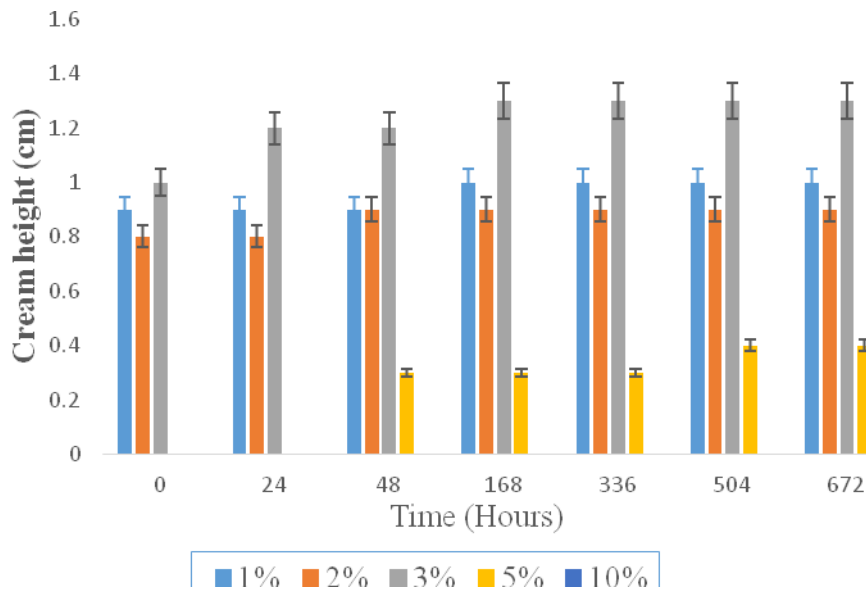


Figure 2: Plot of creaming height against time for liquid paraffin emulsions containing *Terminalia* gum

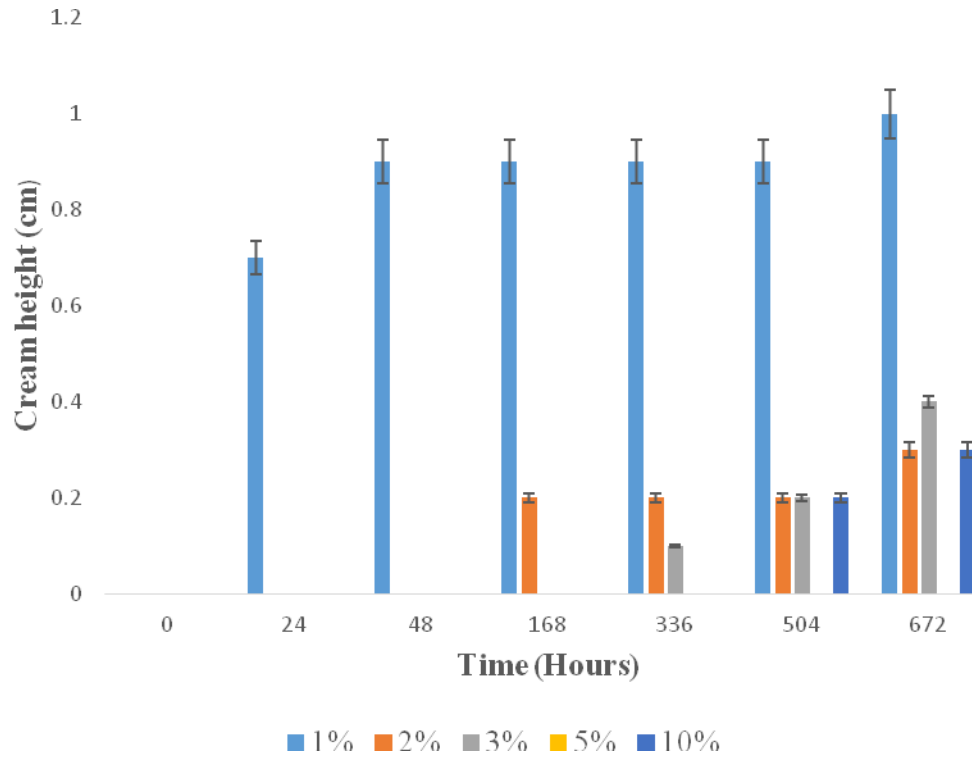


Figure 3: Plot of creaming height against time for castor oil emulsions containing tragacanth gum

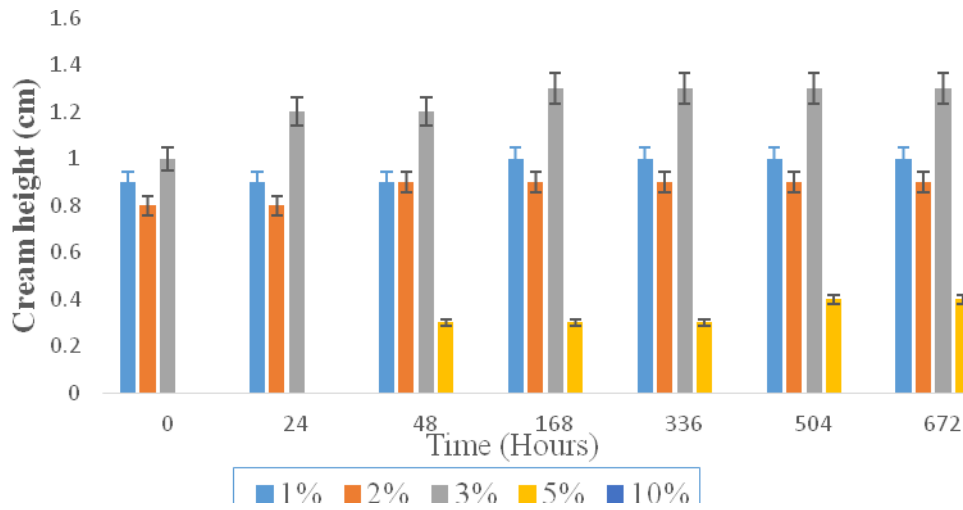


Figure 4: Plot of creaming height against time for liquid paraffin emulsions containing tragacanth gum

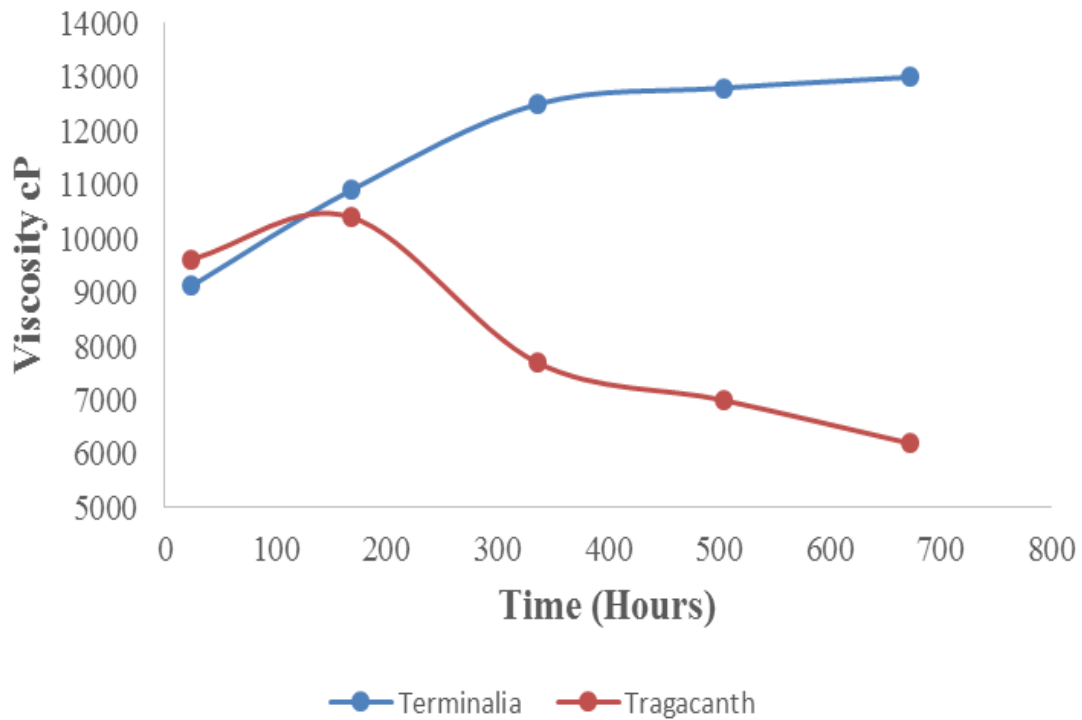


Figure 5: Effect of storage time on the viscosity of emulsion prepared with castor oil containing 10% w/v gum

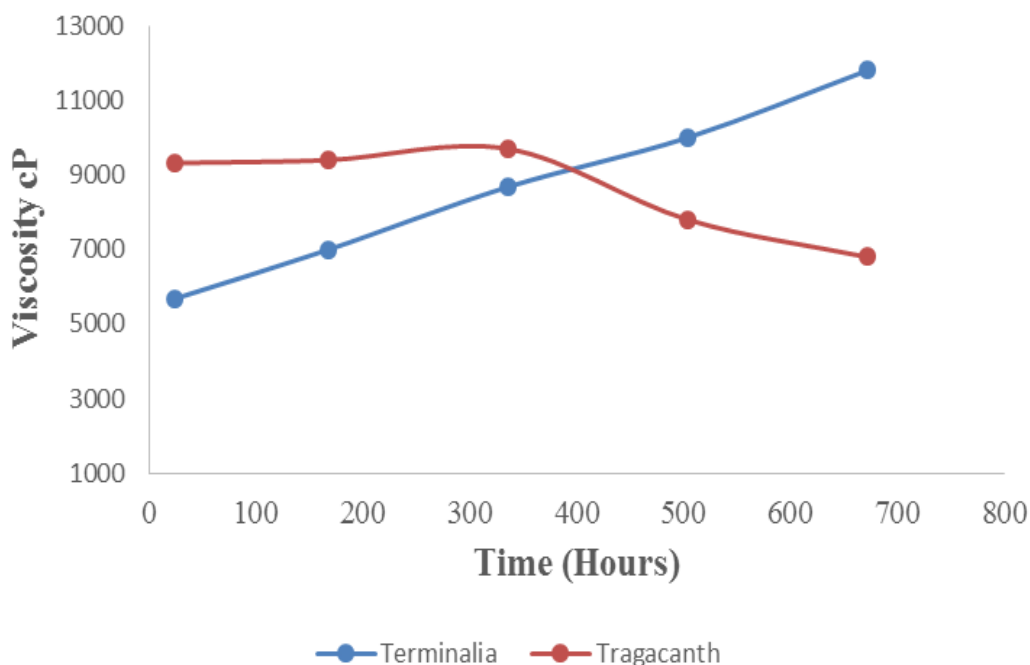


Figure 6: Effect of storage time on the viscosity of emulsion prepared with liquid paraffin oil containing 10% w/v gum

CONCLUSION

This study has shown that to prepare a stable emulsion, the type and concentration of emulsifying agent must be carefully chosen. At high concentrations (5-10% w/v),

emulsions containing *Terminalia* gum did not cream. *Terminalia* gum has good emulsifying properties irrespective of the oil type.

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