

Physicochemical Properties of Gum Arabic Blended with Cassava Starch and Carboxymethyl Cellulose

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Abstract: In this study the physicochemical properties of gum arabic blended with cassava starch and carboxymethyl cellulose was analyzed. The results obtained showed that blending of gum arabic with cassava starch and carboxymethyl cellulose led to increase in moisture content, pH and ash content, with moisture content and some ash content values lying within WHO/FAO standards of not more than 15% and 2-4% respectively for gums. The results also revealed that blending of gum arabic with carboxymethyl cellulose led to increase in viscosity and swelling index while blending with cassava starch led to decrease in viscosity and swelling index. These results suggest that blending of gum arabic with carboxymethyl cellulose and/or cassava starch can be used as modification method to tailor the properties of gum Arabic to specific applications, and as cost cutting measure, as carboxymethyl cellulose and cassava starch are cheaper than gum arabic.

Keywords: Cassava starch, Carboxymethyl cellulose, Food industry, Gum Arabic, Pharmaceutical industry, Modification,

I. INTRODUCTION

Gum Arabic (GA) is a pale to orange brown colour natural organic adhesive (gum) that exudes from the exterior (stems and branches) of *Acacia Senegal* or *Acacia Seyal*, one of the closely related species of *Acacia*, in the form of dry hard nodules. GA is a neutral or slightly acidic complex compound of high molecular weight glycoproteins and polysaccharides and their metal (Ca, K and Mg) salts [1], [2]. Due to the complex composition of GA numerous papers have been published on this subject [3]. The Major polysaccharide is Arabic acid, a polysaccharide that links D-galactose with branches containing L-rhamnose, D-glucuronic acids and L-arabinose. The proteins are essentially classified as arabinogalactanes, rich in hydroxyproline. The chemical composition of GA vary slightly with factors as source, climate, season, age of the tree, etc. [2]. Unlike other vegetable gums, GA dissolves very well in water (up to 50%), resulting to a colorless, tasteless solution which does not interact easily with other chemical compounds [4]. GA is used as a thickener, suspender, emulsifier, stabilizer, flavor carrier, binder and encapsulating material in textile, food, lithography, beverage, dairy and ice cream, plastic, cosmetic, confectionery, pharmaceutical and miscellaneous industries [1], [5], [6].

The many uses of GA highlight the fact that it is a very important industrial raw material, particularly in the food and pharmaceutical industries. The resultant high demand for GA and the dwindling supply occasioned by draught and crisis in the major producing countries—Sudan and Nigeria have led to hike in the price of GA, and thus increase in production cost of products that use it. This increase in price have forced businesses that use GA to look for cheap and sustainable alternatives. These alternatives are either used in their original form or are modified. In this regard, many studies have been conducted. Example is the modification of the functionality of starch through physical, chemical or biotechnological means [7], [8]. Modification alters the properties, shelf life stability in final products, and also stabilizes starch granules during processing making starch suitable for many food and industrial applications [8]. Starch is a natural polymer made of two major components: amylose (structurally linear polymer) and amylopectin (highly branched polymer), extracted from several sources as semi-crystalline granules with different shapes and diameters. Each structure plays a vital role in the ultimate functionality of native starch and its derivatives. Properties such as Viscosity, shear resistance, gelatinization, texture, and so on are functions of native starch's amylose/amylopectin ratio [9]. Starches are of great value for the food industry but have some limitations, like insolubility in cold water, low stability to freeze-thawing, syneresis, in the native form, that in some cases make difficult their utilization [10], [11], thus their modification. Carboxymethyl cellulose (CMC) or cellulose gum [12] is an alternative to GA derivative, synthesized by alkali-catalyzed reaction of cellulose with chloroacetic acid, often used as its sodium salt—sodium carboxymethyl cellulose. CMC is used primarily because it is a nontoxic hypoallergenic with high viscosity. It is used in food as modifier or thickener, as a stabilizer to stabilize emulsions in products like ice cream and bakery products [13], [14]. CMC is also a constituent of several non-food products, such as toothpaste, laxatives, diet pills, water-based paints, detergents, textile sizing, reusable heat packs, paper products, artificial tears, drilling mud, cation-exchange resin [13]; and electrodes [14]

Despite the advantage of low cost that the alternatives to GA have, GA is still preferred because the quality of GA-based products is superior. Considerable attention has been given, presently, to the study of various hydrocolloids and their blends because their rheological and functional properties are complementary. And recent applications have proven that such blends can produce new food formulations and ingredients [15]. There has been substantial developments recently concerning the elucidation of the structure and functional characteristics of GA [3], and considerable number of studies involving its use to modify the functional properties of important industrial raw materials and products have been carried out. However, a review of the literature revealed that only few studies aimed at modifying the functional properties of GA have been reported.

This study aims to investigate the potential of blending as a cost-cutting measure and a modification technique that could be used to tailor the properties of GA to specific products.

TABLE I. Labels of blends and their corresponding blending ratios

GA ₁ /CS ₁		GA ₁ /CS ₂		GA ₁ /CMC ₁		GA ₁ /CMC ₂		GA ₁ /CC ₁ /CS ₁			GA ₁ /CS ₁ /CMC ₁		
GA ₁	CS ₁	GA ₁	CS ₂	GA ₁	CC ₁	GA ₁	CC ₂	GA ₁	CC ₁	CS ₁	GA ₁	CS ₁	CC ₁
2.0	0.0	2.0	0.0	2.0	0.0	2.0	0.0	2.0	0.0	0.0	2.0	0.0	0.0
1.8	0.2	1.8	0.2	1.8	0.2	1.8	0.2	1.8	0.2	0.0	1.8	0.2	0.0
1.6	0.4	1.6	0.4	1.6	0.4	1.6	0.4	1.6	0.3	0.1	1.6	0.3	0.1
1.4	0.6	1.4	0.6	1.4	0.6	1.4	0.6	1.4	0.4	0.2	1.4	0.4	0.2
1.2	0.8	1.2	0.8	1.2	0.8	1.2	0.8	1.2	0.5	0.3	1.2	0.5	0.3
1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	1.0	0.6	0.4	1.0	0.6	0.4

The subscripts 1 and 2 stand for 0.5µm and 0.25µm particle sizes respectively

C. *Determination of Moisture Content*

Pulverized GA (2g) was accurately weighed into a crucible, oven-dried at 105°C for 5 hours, allowed to cool in a desiccator and the weight taken. The experiment was done in triplicate and an average of the results taken. Moisture content was calculated as a percentage of the weight loss from the original weight using equation 1. The procedure was repeated for CMC, CS and all the blends.

$$\text{Moisture content} = \frac{W_1 - W_2}{W_1} \times 100 \quad (1)$$

Where W₁ = Weight of sample before heating and W₂=Weight of sample after heating

D. *Determination of Ash Content*

2g of pulverized GA sample was accurately weighed into a crucible, heated on a burner in air to remove its smoke then burned in a furnace at 650-700°C for 8 hours and the weight noted after allowing it to cool in a desiccator. The ash content was expressed as a percentage ratio of the weight of

II. MATERIALS AND METHODS

A. *Plant materials*

Gum Arabic (GA) sample, Carboxyl Methyl Cellulose (CMC; EASTCHEM brand), and Cassava tubers were obtained at Arafat market, a chemical store, and Gangare market respectively in Jos North Local government area of Plateau State, Nigeria. The dried need quality gum was separated from the Gum sample which consisted of large and small nodules, tree bark and organic debris by hand-picked select gum method, pulverized using a blender, sieved and stored in a bottle until use. The cassava tubers were processed to Cassava Starch (CS). The tubers were first washed with tap water, peeled, washed again with distilled water then graded and dried under room temperature. The dried cassava was then crushed with mortar and pestle, and sieved into different particle sizes.

B. *Blending of GA with CS and CMC*

several blends were prepared by mixing 0.5µm GA with 0.5µm and 0.25µm CS and CMC at 35°C. The blends were prepared and labeled as shown in table I below.

the ash to the weight of the oven-dried sample. The ash content was calculated using equation 2 shown below. The same procedure was used to determine the ash content of CMC, CS and all the prepared blends.

$$\text{Ash content} = \frac{Z - X}{Y - X} \times 100 \quad (2)$$

Where X= Weight of empty crucible, Y = Weight of sample + crucible and Z = Weight of Ash + crucible

E. *Determination of pH*

2 grams of each of GA, CS, CMC and their various blends were weighed separately and dissolved in 10ml of distilled water, made up to 100ml and the pH of the solutions taken using a calibrated pH meter [16].

F. *Determination of Relative Density*

The densities of GA, CS and CMC were measured at warm temperature (30°C) using 100cm³ density bottle. The weight of the clean dry density bottle was measured as (M₁) in grams then filled with the sample and weighed as (M₂) in

grams. The density was then calculated using the following formula:

$$\text{Density} = \frac{M_2 - M_1}{100} \quad (3)$$

G. Determination of Relative Viscosity

2g of each of GA, CS, CMC and their various blends were weighed separately, dissolved in 10mls of distilled water and the solutions made up to 100ml and the viscosities measured at 25°C using Viscometer (Brookfield RV, DV Viscometer; AMETEK Brookfield, USA).

H. Determination of Swelling Index

The swelling index of GA, CS, CMC and their various blends were determined as follows: 2g of each sample was placed in a white cloth of known weight, tied with a rope, immersed in 100ml of distilled water for 1 hour and allowed to stand (for 10 - 12 minutes) until the last drop of water was observed, and the weight recorded. The swelling index was calculated using the following formula:

$$\text{Swelling index} = \frac{W_2 - W_1}{W_1} \times 100 \quad (4)$$

Where W_1 = Initial weight of sample and W_2 = final weight of sample

III. RESULT AND DISCUSSION

A. pH Value

Table II shows that the pH of GA (4.62±0.001) is slightly more acidic than that of CS (6.45±0.02), while that of CMC (7.01±0.01) is neutral. As shown in table III, Increase in pH was observed with increasing level of CS (4.87-5.99) and CMC (5.25-6.24) in the various blends. The table also shows that the particle size of CS and CMC has little effect on the pH of the blends. Knowledge of pH of a substance is an important parameter in determining its suitability in formulations since the stability and physiological activity of most preparations depends on pH [17]. The results of this study showed that the solutions of GA/CS, GA/CMC and GA/CS/CMC blends are acidic. The pH values are in good agreement with reported pH values for Gums by several authors [17]-[20]

TABLE II. Physicochemical Parameters of GA₁, CS₁ and CMC₁

Parameter	GA ₁	CS ₁	CMC ₁
% Moisture Content	12.12±0.025	13.40±0.05	13.70±0.06
Viscosity/cp	400±0.6	230±0.62	21440±1.0
pH	4.62±0.01	6.45±0.02	7.01±0.01
% Swelling Index	47.30±0.2	25.90±0.25	81.0±1.8
Density/ gcm ⁻³	0.64±0.01	0.765±0.01	0.55±0.01
% Ash Content	2.70± 0.01	4.90± 0.2	8.95±0.02
Solubility in cold water	Soluble	Insoluble	Soluble
Solubility in warm water	Soluble	Soluble	Soluble

The subscript 1 stands for 0.5µm particle size

TABLE III. pH values of blends

GA ₁ /CS ₁	GA ₁ /CS ₂	GA ₁ /CMC ₁	GA ₁ /CMC ₂	GA ₁ /CMC ₁ /CS ₁	GA ₁ /CS ₁ /CMC ₁
4.62	4.62	4.62	4.62	4.62	4.62
4.87	4.89	5.25	5.26	5.25	4.87
4.98	5.00	5.54	5.56	5.48	5.05
5.22	5.24	5.92	5.93	5.75	5.11
5.44	5.46	6.09	6.10	5.94	5.67
5.99	6.10	6.24	6.25	6.16	5.87

The subscripts 1 and 2 stand for 0.5µm and 0.25µm particle sizes respectively

B. Moisture content

The moisture contents of GA (12.12%), CS (13.40%) and CMC (13.70%) are within FOA international specification of ≤15% for gums [21]. As shown in table IV, as the quantity of CS and CC increased, there is an increase in moisture content in all the two-component and three-component blends. All the moisture content values are within FAO international specification of ≤15% for gums [21].

It is important to investigate the moisture content of a material as the economic importance of an excipient for industrial application lies not only on the cheap and availability of the material but optimization of production processes such as drying, packaging and storage [22]. Moisture content is a good parameter for detecting the quality of crude drugs. Low or high moisture content compromise the quality of drug and affects its efficacy.

TABLE IV. % Moisture contents of blends

GA ₁ /C ₁	GA ₁ /CS ₂	GA ₁ /CC ₁	GA ₁ /CMC ₂	GA ₁ /CMC ₁ /CS ₁	GA ₁ /CS ₁ /CMC ₁
12.12	12.12	12.12	12.12	12.12	12.12
12.95	12.85	13.00	12.98	13.13	12.95
13.05	13.00	13.10	13.14	8.25	7.55
13.55	13.25	13.15	13.45	9.05	8.50
13.60	13.40	13.40	13.60	9.55	9.50
13.66	13.56	14.00	13.97	10.95	9.60

The subscripts 1 and 2 stand for 0.5µm and 0.25µm particle sizes respectively

Ash Content The ash content of GA is 2.70%, CS is 4.90% and CMC is 8.95% (shown in Table II). Table V shows that there is an increase in ash content as the quantity of CMC and CS increases, with CMC having more effect than CS. The ash content of some of the GA/CS, GA/CMC and GA/CS/CMC blends fall within the range (of 2-4%) of international specification [21], while others were not. Ash is a non-volatile inorganic residue that remains after burning off the organic material [23] Ash content is used to determine the critical

levels of foreign matter, acid insoluble matter, salts of calcium, potassium and magnesium [2], [24]. Ash content is also an important property considered as purity parameter in gums. Low value of ash is an indication of low level of contamination.

TABLE V. % Ash Contents of Blends

GA ₁ /CS ₁	GA ₁ /CS ₂	GA ₁ /CM ₁	GA ₁ /CMC ₂	GA ₁ /CMC ₁ /CS ₁	GA ₁ /CS ₁ /CMC
2.70	2.70	2.70	2.70	2.70	2.70
2.60	2.59	3.05	3.04	3.05	2.20
2.65	2.61	3.60	3.59	3.10	1.70
3.00	2.98	4.15	4.10	3.45	2.65
3.50	2.38	5.35	5.30	4.05	3.00
4.50	4.46	6.10	6.00	4.10	3.10

The subscripts 1 and 2 stand for 0.5 μ m and 0.25 μ m particle sizes respectively

C. Viscosity Table II

shows that CMC has the highest viscosity (21440cp) while CS has the least (230cp). The solubility and viscosity of GA are in agreement with literature, which showed that GA has high water solubility and a relatively low viscosity compared to other gums. As shown in table VI, the viscosity of GA increased from 400 to 7040cp as the quantity of CMC was increased and decreased from 400cp to 240cp as the quantity of CS was increased. Table VI also shows that particle size has little effect on the viscosity of the blends.

High relative viscosity of a gum solution suggest the presence of high molecular weight compounds in the gum's constitution [25]. Viscosity plays an important role in food and pharmaceutical processing. Gums with high viscosity are food additives that can influence processing condition such as water retention, reduction of evaporation roles, alteration of freezing rate and modification of ice crystal formation as in the manufacture of ice-cream [26].

TABLE VI. Viscosity/cp of Blends

GA ₁ /CS ₁	GA ₁ /CS ₂	GA ₁ /CMC ₁	GA ₁ /CMC ₂	GA ₁ /CMC ₁ /CS ₁	GA ₁ /CS ₁ /CMC ₁
400	400	400	400	400	400
320	320	720	710	320	720
310	300	1840	1800	160	1700
280	278	3040	3000	320	2800
260	259	5040	5000	420	4940
240	237	7040	7010	880	6740

The subscripts 1 and 2 stand for 0.5 μ m and 0.25 μ m particle sizes respectively

D. Swelling Index

Swelling index is a measure of the water holding capacity of a gum. Table II shows the swelling capacity, which signifies the hydrophilic nature of GA, CMC and CS. CMC has the highest swelling index (81%) followed by GA (47.30%) and CS (25.90%). Table VII shows the swelling index of GA/CS, GA/CMC and GA/CS/CMC blends. Increase in the quantity of CMC increased the swelling index of GA while decrease in swelling index of GA was observed with an increase in quantity of CS.

This result implies that in applications where swelling is a desired quality, such as in the production of drugs where the capacity of a gum to swell into a gelatinous material from which embedded drug could be released [18], CMC can be used to improve GA. Table vi also shows that particle size has very little effect on GA/CMC and GA/CS blends.

TABLE VII. % Swelling index of Blends

GA ₁ /CS ₁	GA ₁ /CS ₂	GA ₁ /CMC ₁	GA ₁ /CMC ₂	GA ₁ /CMC ₁ /CS ₁	GA ₁ /CS ₁ /CMC ₁
47.30	47.30	47.30	47.30	47.30	47.30
45.10	45.00	50.70	50.65	45.10	50.70
40.90	40.70	55.60	55.55	44.30	53.45
38.06	38.00	63.10	63.00	46.15	55.90
33.50	33.40	69.00	68.95	48.90	57.60
30.60	30.45	76.00	75.96	52.45	60.30

The subscripts 1 and 2 stand for 0.5 μ m and 0.25 μ m particle sizes respectively

IV. CONCLUSION AND RRECOMMENDATION

This study has shown that some of the blends of GA/CS and GA/CMC have good physicochemical properties that are within the range of WHO/FAO standards for GA, thus can be used in food, pharmaceutical and other industries where as emulsifiers, binders and thickeners. For instance, that CMC can be used to improve the binding capacity of GA for usage in pharmaceutical industries, where swelling is a primary mechanism in diffusion controlled release dosage form, as tablet binders [18], [30].

The findings of this study have also shown that blending (of GA with CMC and CS) can be employed to tailor the properties of GA to specific applications, and to reduce production cost as CMC and CS are cheaper than GA.

The morphological properties of hydrocolloids are of importance when considering applications based on surface characteristics. It has been reported that particle size and specific surface area influence the hydration behaviour of gums, which in turn influence their intrinsic viscosity and molecular mass [17], [27]–[29]. Thus, we recommend the determination of the morphological properties of CS, GA, CMC and their various blends and the exploration of the

qualities of products, particularly food and pharmaceutical products, in which GA blends, with CS and CMC, are used.

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