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Acacia caven Gum Studies of Hydrodynamic Parameters

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Abstract

The *acacia caven* gum (ACG) is an exudate from *Acacia caven* tree, this gum is dissolved in water and purified by precipitation with ethanol. In this paper the intrinsic viscosity, [η], is calculated by dissimilar equations, as Huggins, Kramer, Arrhenius-Rother-Hoffmann, Staudinger & Heuer, Square (S), Square root (SR), S-SR average plot, average values of S-SR, and being Kraemer is taken as standard. The different methods are evaluated and compared. From [η] measurements are determined the molecular weight (M_V) and hydrodynamic radius (RH), with values of 1060 Kg/mol and 24 nm, respectively. This polysaccharide acquires a random coil conformation with Mark-Houwink parameters of a=0.5507 and k=0.0225 cm^3/g. This polysaccharide is applied as thickener, coemulsifier and gelling in different industry.

Keywords: *Acacia caven* gum; Intrinsic viscosity; Molecular weight

Abbreviations: ACG: *Acacia Caven* Gum; MG: Mesquite Gum; BG: Brea Gum; AG: Arabic Gum.

Introduction

Acacia caven, also known as serrano shrub, espinillo or aromo, is a plant of the family *fabaceae*, of the subfamily *Mimosoideae.* In this case, the *Acacia caven* belongs to the acacieae tribe and belongs to the acacia genus. Tree or small tree between 1.5 and 5m tall with hemispherical and extended crown, trunk up to 25 cm in diameter, persistent bark, longitudinally fissured, dark brown or grayish color, straight spines geminated, whitish, between 5 and 25 mm in length, very sharp, without stings. Bipinnate compound leaves with 3-15 pairs of pineapples and numerous leaflets, pubescent petiole. Hermaphrodite flowers, sessile, yellow-orange (golden), fragrant, arranged in globose, lateral heads. Fruit, legume thick, cylindrical, indehiscent, pericarp of lustrous black color at maturity, numerous seeds, biseriates. The espinillo (A caven) is a species typical of the province of Chaco and the province of the spinal, which is why it is abundant in the N and E of San Luis. Ancestral Uses: healing (leaves), sneezing (seeds), digestive (seeds), as a wound disinfectant (ashes and exudate); firewood and fences [1-13].

Roasted seeds are used for the preparation of coffee, and your flowers they are used for the preparation of tea. The flowers present intense aromas which are used for the preparation of aromatic products, essential oils and its essence is used in perfumery [14,15]. All Acacia gums are nonstarch, highly branched polysaccharides, either neutral or slightly acidic. The structures of natural gums vary considerably but many have a 1→3 linked D-galactopyranose backbone consisting of four sugars, L-arabinose, L-rhamnose, D-galactose, and D-glucuronic acid [16]; although for *Acacia caven* there is no reliably determined structure, but it is supposed to be similar to the Arabic gum with which it is botanically related. In addition, gums also contain amino acids, e.g., hydroxyproline, serine, proline, threonine, leucine, glycine, and histidine, along with trace amounts of lipids. Polysaccharides form complexes with calcium, magnesium, iron, zinc, and potassium for better stability [17].

The Arabic gum from *Acacia senegal/seyal* trees is now used in lithography, flocculating agent, binder of color pigments, for corrosion inhibition, in the cosmetic industry as an adhesive, and the stabilization of carbon nanotubes [18]. Arabic gum is obtained by removing the bark of trees such as *Acacia senegal*; it is a heteropolysaccharide hiperbranched consisting of a main chain of β-galactopyranose units to which L-ramnopiranose with residues of glucuronic acid and L-arabinofuranoses [19]. Similar structure is determinate for Acacia glomerosa [20], similar studies were released for Acacia drepanolobium and Acacia. kirkii ssp. kirkii var. kirkii [21]. The mesquite gum (MG) is a highly branched polysaccharide. The primary structure of the MG component has been described in detail: a central backbone comprised of β(1→3)-linked D-galactose residues, to which side oligosaccharide chains of varying size are attached at O(6); these branches contain L-arabinose and D-galactose and minor amounts of L-rhamnose, D-glucoronate and 4-O-methyl-D-glucuronate [22].

Mesquite gum is effective in the preparation of oil-in-water emulsions, over a wide range of pH values, encapsulant, foaming agent, colloid protector, baking [23], and etnopharmacy [24]. The different gums obtained from the Acacia species, including arabic gum, have very small differences with respect to each other. There are different papers in which Mark-Houwink parameters have been determined which can be seen in Table 1. In many works these gums are compared with mesquite gum and arabic gum that has similar use qualities [25,26]; although it can also be compared with the exudate of chañar brea or Brea gum (BG) from *Cercidium praecox*) [27-29].

Gum	Plant	[η] cm^3/g	k (cm^3/g)	*a*	MW (Kg/mol)	Reference
AG	*Acacia senegal*	12.5-25.4	0.0130	0.54	320-580	Anderson & Rahman (1967) [30]
AG	*Acacia senegal*	15.5-40	0.0160	0.53	400-2200	Vandevelde & Fenyo (1985) [31]
AG	*Acacia Senegal*	10.4-19.8	0.0300	0.47	240-780	Idris et al. (1998) [32]
AG	*Acacia senegal*	23	-	-	268-2340	Sanchez et al. (2001) [33]
AG	*Acacia senegal*	23	0.0130	0.54	108-1340	Al-Asaf et al. (2004 a-b) [34,35]
AG	*Acacia senegal*	16.2-80.2	-	-	286-2670	Renard et al. (2006) [36]
AG	*Acacia senegal*	18.2-21.6	-	-	622-2540	Wang et el. (2007) [37]
AG	*Acacia senegal*	1-73	-	-	250-1000	Yebeyen et al. (2009) [38]
AG	*Acacia Senegal* *Acacia seyal*	6-8 6	-	-	525-400 770	Cozic et al. (2009) [39]
AG	*Acacia senegal*	17.7	0.0151	0.52	797	Rinaudo et al. (2008) [40], Lopez-Franco et al. (2012) [41]
AG	*Acacia spp.*	19.81	0.01311	0.5406	760	Masuelli (2013) [42]
AG	*Acacia Senegal* *Acacia seyal*	22.8 16.5	-	-	680 820	Lopez-Torres et al. (2015) [43]
AG	*Acacia Senegal var. senegal,* *Acacia mellifera,* *Acacia seyal var. seyal,* *and Acacia tortilis var. raddiana*	-	-	-	Mn 240 2010 2950 2060	Daoub et al. (2018) [44]
MG	*Prosopis alba,* *P. glandulosa,* *P. glandulosa* var.	11 10 10-12			460 250 180-680	Anderson & Farquhar (1982) [45]

	glandulosa, *P. glandulosa* var. *torreyana,* *P. juliflora,* *P. laevigata* and *P. velutina*	10 14 12.6 4.3			450 310 800 83	
MG	*Prosopis laevigata*	-	-	-	3.5-940	Orozco-Villafuerte (2003) [46]
MG	*Prosopis spp.*	8.3-9.9	0.0147	0.50	318-453	Rinaudo et al. (2008) [40], Lopez-Franco et al. (2012) [41]
BG	*Cercidium Precox*	52.91	0.1347	0.4133	1890	Masuelli et al. (2019) [29]

Table 1: Mark-Houwink Parameters of Arabic gum (AG) and mesquite gum (MG).

In this work we will study the *Acacia caven* gum obtained from exudate of *Acacia caven* tree. At ACG we will perform physicochemical studies in aqueous solution using density and viscosity measures. The [η] is calculated by diverses equations using Kraemer as standard. Also, we will obtain Mark-Houwink parameters (M-H) from intrinsic viscosity measurements, using dextrans as reference for calibration [47]. From M-H parameters we will determine the molecular weight and the hydrodynamic parameters of ACG.

Intrinsic Viscosity and Hydrodynamic Parameters

Using capillary viscometer, the viscosity can be calculated from equation:

$$\eta = A\rho t \qquad (1)$$

Where η is viscosity (poise), A is viscometer constant (cm^2/s^2), ρ is solution density (g/cm^3), and t is drainage time (s).

The relative viscosity (η_r), is:

$$\eta_r = \frac{\eta_s}{\eta_0} = \frac{\rho_s t_s}{\rho_0 t_0} \qquad (2)$$

Where the subindex "*s*" indicates "solution" and "*0*" indicates "solvent" viscosity.

The "specific viscosity" is calculated from:

$$\eta_{sp} = \eta_r - 1 \qquad (3)$$

In Huggins' method [48], intrinsic viscosity [η] is defined as the ratio of the increase in relative viscosity (η_{sp}) to concentration (c in g/cm^3) when the latter tends towards zero.

$$\frac{\eta_{sp}}{c} = [\eta] + K_H [\eta]^2 c \qquad (4)$$

Where K_H is Huggins constant.

The Kraemer [49] propose the equation:

$$\frac{\ln \eta_r}{c} = [\eta] + K_K [\eta]^2 c \qquad (5)$$

Where K_K is Kraemer's constant.

Arrhenius-Rother-Hoffmann [50-51] proposes this equation:

$$\frac{\ln \eta_r}{c} = [\eta] + K_A \ln \eta_r \qquad (6)$$

Where K_A is Arrhenius-Rother-Hoffmann constant.

Staudinger & Heuer [52] propose the following equation:

$$\ln \frac{\eta_{sp}}{c} = \ln[\eta] + K_{S-H} [\eta] c \qquad (7)$$

Where K_{S-H} is Staudinger & Heuer constant.

In this work four alternatives are proposed to calculate the intrinsic viscosity. First a quadratic method on both sides of equality; the second is a method that proposes the square root on both sides of equality; the third is a graphical average of an empirical combination of quadratic and square root. Finally, the fourth method proposes an average between the two values obtained from the intrinsic viscosity by the quadratic and the square root [53].

The first method is a square plot where the figures of the equations 8 are made (S), from the ordinate to the origin, the [η] is obtained from them.

$$\left(\frac{\eta_{sp}}{c}\right)^2 = [\eta]^2 + k_{p2} c^2 \qquad (8)$$

The second is a plot method where the figure of the equation 9 is made (Square Root method or SR). The intrinsic viscosity is obtained from the ordinate to the origin.

$$\left(\frac{\eta_{sp}}{c}\right)^{1/2} = [\eta]^{1/2} + k_{p1/2}c^{1/2} \qquad (9)$$

From average numeric value from equation 8 and 9 is obtained (Average Value S-SR).

Average Plot S-SR equation 10 is an empirical equation and is proposed, from the plot realization; from the ordinate to the origin the intrinsic viscosity is obtained.

$$\frac{1}{2}\left\{\left(\frac{\eta_{sp}}{c}\right)^2 + \left(\frac{\eta_{sp}}{c}\right)^{1/2}\right\} = 0.8[\eta]^2 + \frac{1}{2}k_p\left(c^2 + c^{1/2}\right) \qquad (10)$$

Where k_{p2}, $k_{p1/2}$ and k_p are constants which contain the intrinsic viscosity function.

Therefore, an average of both plot solutions gives a value of less than 5% regarding the Kraemer method.

The Mark-Houwink [54,55] equation 11 describes the relationship between intrinsic viscosity and molecular weight. Since molecular weight is related to the size of the polymer chain [56]. The calculation of Mark-Houwink (M-H) parameters is carried out by the plot representation of the following equation:

$$\ln[\eta] = \ln k + a \ln M_w \qquad (11)$$

Where k and a are M-H constants, depending upon the type of polymer, solvent and temperature. The exponent a is a function of polymer geometry and varies from 0.5 to 2.0 [57].

Using the polymer standards, as dextrans [47], a plot of ln [η] versus ln M_w usually gives a straight line. The slope of this line is the value of a and its intercept is equal to ln k. The a values of 0.5 reflect a sphere, from 0.5-0.8 a random coil, and from 0.8-2.0 a rod like conformation [58,59].

The Einstein relation [60] is used for determinate hydrodynamic radius (R_H),

$$M[\eta] = \nu_{a/b} N_A \frac{3}{4}\pi (R_H)^3 \qquad (12)$$

The relation of intrinsic viscosity and specific volume is determinate $\nu_{(a/b)}$, called Einstein viscosity increment (Simha number or shape factor) [61-66], as:

$$\nu_{a/b} = \frac{[\eta]}{V_s} \qquad (13)$$

And V_s is specific volume (cm^3/g).

Experimental

***Acacia caven* gum (ACG):** The exudate from *Acacia caven* once collected was dissolved at 25ºC in distilled water, microfilter with Durapore® filter of 0.45 µm. Once filtered, it was precipitated with ethanol several times, then dried at 50°C and ground (separating with 200 mesh). Finally, it was redissolved in distilled water at a concentration of 0.1% wt.

Viscosity and density: Measurements were taken from fresh ACG in aqueous solutions of 0.1-1 % wt. Solutions and dissolutions were prepared with deionised water. The different temperatures were maintained using a HAAKE C thermostatic bath (±0.1°C). Determinations were done using an Ubbelohde viscometer (IVA 1), with a water draining time of 36.07s. The density of each solution was measured using an Anton Paar DMA35N densimeter.

Results and Discussion

Dilutions were prepared from *Acacia caven* gum solutions; the intrinsic viscosity was determined by several methods. In this paper, the M-H parameters were used to calculate the M_v, using K=0.0225 cm^3/g and a=0.5507, respectively. The [η] by Huggins method was of 46.0520 cm^3/g; by Kraemer method the [η] was of 46.7780 cm^3/g. From Kraemer method the viscometric molecular weight was calculated with a value of 1060 kg/mol. This polysaccharide acquires is random coil conformation in aqueous solution with much branched characteristic.

Figure 1 shows the Huggins and Kraemer methods; in this figure we can see the difficulty that both methods converge for the calculation of the intrinsic viscosity. For this work the Kraemer method is taken as standard and with which the rest of the methods are compared. One of the problems that normally involve the Huggins or Kraemer method is its slope and also its R^2 approximately equal to zero. In this case Huggins acquires an R^2 approximately equal to zero, not so Kraemer.

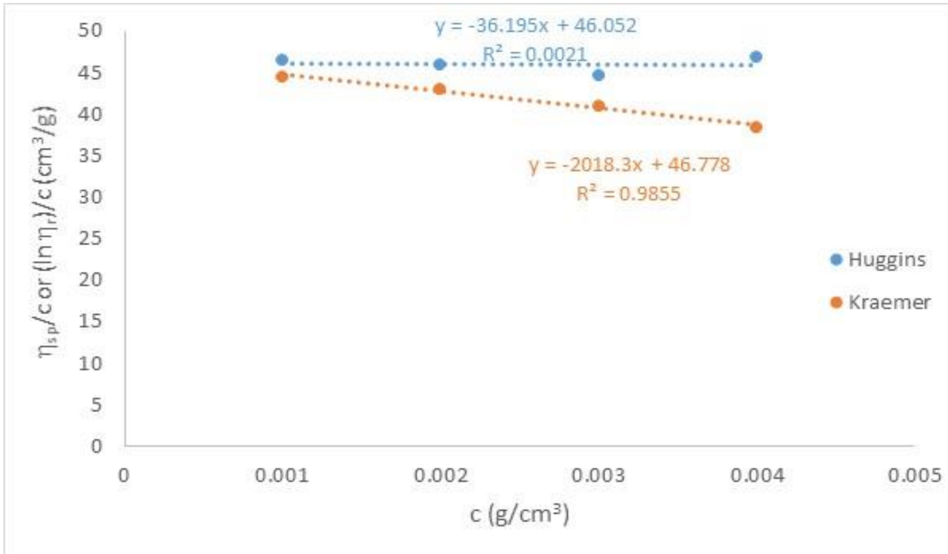

Figure 1: Huggins and Kraemer methods.

The Arrhenius-Rother-Hoffmann equation, observed in Figure 2, is one of the oldest and most accurate methods of plot calculation of the intrinsic viscosity with RE% less than 2.18%. This classic method is universal and applicable to macromolecules of all types, although it is not widely used. The Staudinger-Heuer methods with a RE% less than 3.06%, as can be seen in Figure 3.

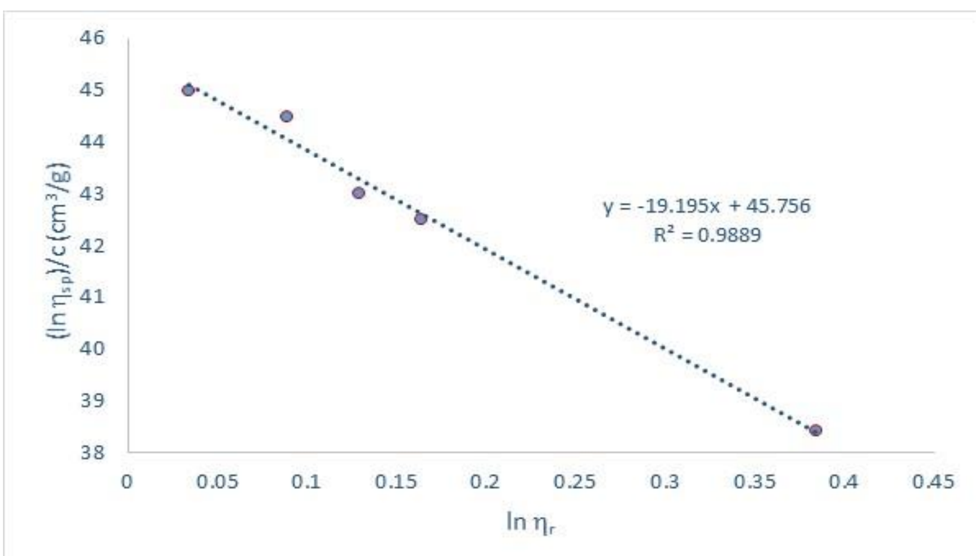

Figure 2: Arrhenius-Rother-Hoffmann Equation.

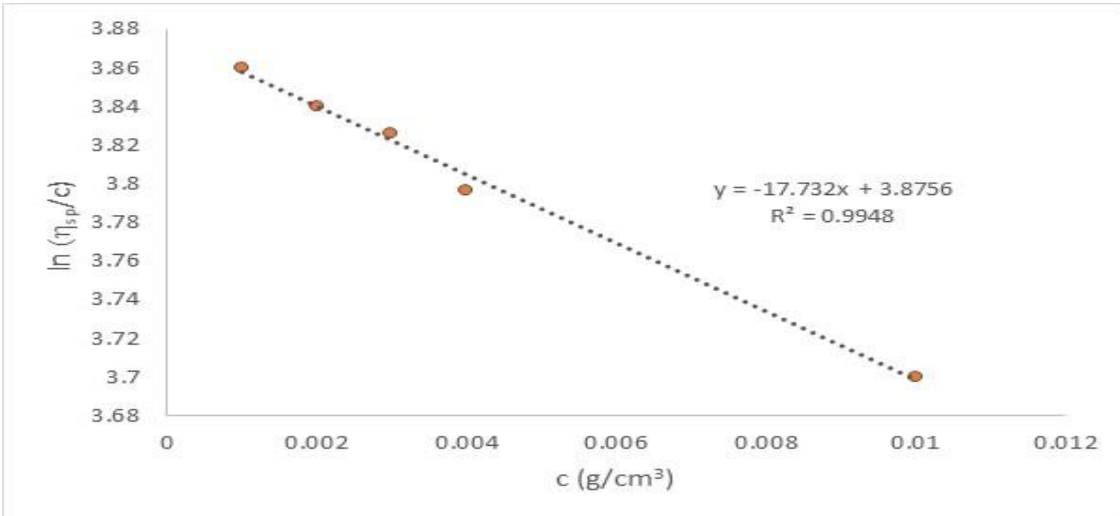

Figure 3: Staudinger-Heuer equation.

In this paper two ways to calculate are proposed that serve to determine the intrinsic viscosity and are plot methods that take into account the average between two values obtained by different methods that we will detail next.

The plot method is based the equations 8 and 9, plotted Figure 4, 5 and 6. Finally, from the ordinate to the origin, the intrinsic viscosity is obtained and the average is obtained from them (Table 2). The relative percentage error with respect to Kraemer of equation 8 is 0.23%; and of equation 9 obtained error respect to Kraemer is 8.31%. Therefore, an average of both plot solutions gives a value of 4.27% regarding the Kraemer method (Table 2).

Methods	**Huggins**	**Kraemer**	**Arrhenius-Rother-Hoffmann**	**Staudinger-Heuer**
[η] (cm³/g)	46.0520	46.7780	45.7560	48.2116
R^2	-	0.9855	0.9889	0.9948
RE%	1.55	-	2.18	3.06
Methods	Square	Square Root	Average Value S-SR	Average Plot S-SR
[η] (cm³/g)	46.8860	50.6688	48.1096	48.7774
R^2	0.9981	0.9921	-	0.9794
RE%	0.23	8.31	4.27	2.84

Table 2: Intrinsic viscosity by different methods.

In the second case, an empirical equation 10 is proposed, which is a combination of the previous plot method (an empirical combination of equations 8 and 9). Figure 6 is the realization of the plot of this equation, from the intercept, the intrinsic viscosity is obtained. The intrinsic viscosity obtained is with a value of 48.11 cm³/g and RE%=2.84%.

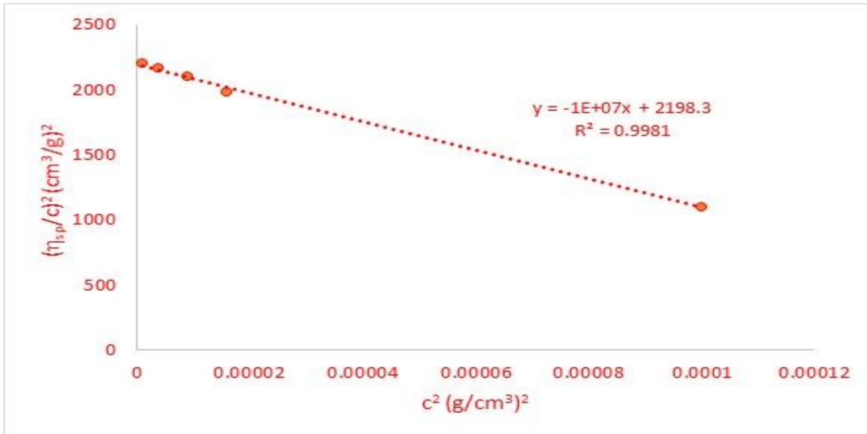

Figure 4: Square methods.

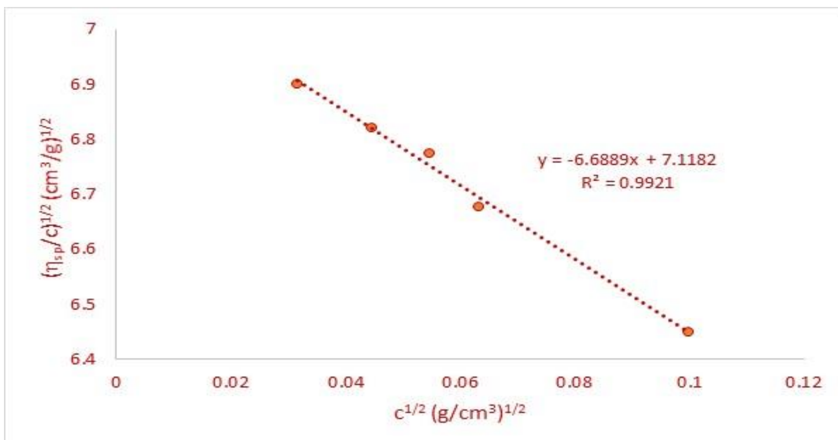

Figure 5: Square root method.

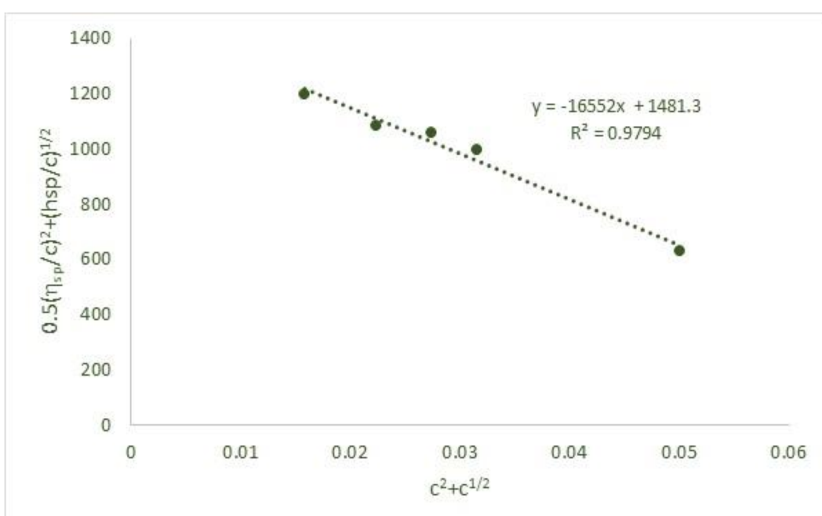

Figure 6: Average Plot S-SR method.

The Mark-Houwink parameters observed in Table 3. The R_H of the polymers change with the type solution and with temperature via changes in their chain flexibility. The molecular weight determined for this work is 1060 kg/mol, with an intrinsic viscosity by Kraemer method of 46.7780 cm^3/g. What is very clear is that it is a molecule that acquires a random coil conformation (quasispherical shape), that is very branched and that water is a little ideal solvent; clarifying that it is only valid for the treatment performed in this work. M-H value of "*a*" confirm that for these conditions. Empiric functions can be used to facilitate the calculation of these parameters in an acceptable way, as proposed in this work.

R_H (nm)	M (kg/mol)	$\nu_{a/b}$	k (cm^3/g)	*a*
24	1060	2.52	0.0225	0.5507

Table 3: Mark-Houwink and hydrodynamic Parameters.

Comparing the aforementioned AG with ACG, this gum is in the range of molecular weights and intrinsic viscosities observed, its Mark-Houwink parameters show the similarity of both gums, regardless of their origin. As for comparing this gum with MG and BG, it should be taken into account that they have similar physicochemical characteristics, but it must be borne in mind that structurally they are different in the composition and bonding forms of the monosaccharaides in the polysaccharide.

Conclusion

The *acacia caven* gum in aqueous solution acquires a random coil conformation (ellipsoid shape) which was confirmed by the parameter "*a*" of Mark-Houwink. Regarding the intrinsic viscosity measurement Arrhenius-Rother-Hoffmann, Staudinger-Heuer, Square and Average Value S-SR methods are good and comparable with Kraemer that was taken as standard. Also, the methods proposed in this paper were suitable for the calculation of intrinsic viscosity. Branching characteristic was observed in *acacia caven* gum.

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