

Hydrodynamic Properties of Whole Arabic Gum

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Abstract The most economically important of the hydrodynamic properties of a material are viscosity and density, which allow determining the intrinsic viscosity of raw materials used in the food industry. They serve as an indirect measure of molecular weight (M), hydrodynamic radius (R_H), number of Simha, ($v_{(P)}$), Perrin parameter (P); hydration value (δ), Scheraga-Mandelkern parameter (β), and Flory parameters (ϕ_0 and P_0). Normally, these parameters are reported at a temperature of 25°C, which limits their use at different temperatures. This work studies the temperature-dependence of whole arabic gum (WAG) in aqueous solution, finding that in aqueous solution, this biopolymer presents a random coil shape with $v_{(P)} \approx 2.55$. The behavior of WAG in this system indicates that it behaves as a colloidal particle that tends to compact as temperature increases (R_H decrease). The M of WAG calculated here are 760000 g/mol. The Mark-Houwink-Sakurada equation constants, a and k , for WAG in water solvent-temperature systems have been reported already, where the value of a ranges from 0.5496 to 0.5085 within a temperature range of 20 to 50°C.

Keywords: whole arabic gum, hydrodynamic parameters, Mark-Houwink-Sakurada parameters, temperature

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1. Introduction

Arabic gum is a polysaccharide of vegetable origin with a high molecular weight [1,2] (mainly obtained from *Acacia senegal*, *Acacia seyal*, *Acacia karoo*, *Acacia polyacantha*, *Acacia sieberana*, etc.). The different arabic gums can act as thickeners and gelling agents [3], and they show some functional properties such as emulsification [4,5], stabilization [6] and microencapsulation [7,8]. Arabic gum used in food emulsions has highly hydrophilic and anionic properties [9]. This polysaccharide is used in the food, pharmaceutical [10,11], biotechnology [12,13], adhesive [14] and mining industries [15,16]; it has the capacity to stabilize oil-in-water emulsions in low-viscosity concentrated solutions [17].

Arabic gum is a branched, neutral or slightly acidic, complex polysaccharide obtained as a mixed calcium, magnesium, and potassium salt. Its backbone consists of 1,3-linked β -D-galactopyranosyl units. The side chains are composed of two to five 1,3-linked β -D-galactopyranosyl units, joined to the main chain by 1,6-linkages. Both the main and the side chains contain units of β -L-arabinofuranosyl, β -L-rhamnopyranosyl, β -D-glucuronopyranosyl, and 4-O-methyl- β -D-glucuronopyranosyl, the latter two mostly as end units. The characteristics of arabic gum may vary significantly, depending on the geographical origin and age of the trees, climatic conditions, soil environment, and even the place of exudation on the tree. Several studies have been undertaken to obtain identifiable methods and parameters that allow to characterize commercial gum

arabic and to detect adulterants. Because of its heterogeneous nature, arabic gum has been described as heteropolymolecular, having, on the one hand, variation in monomer composition and/or in the linking and branching of the monomer units and, on the other hand, and a wide molecular weight distribution. Its heterogeneous nature has been studied extensively using different techniques: hydrophobic affinity chromatography, anion exchange chromatography, gel permeation chromatography, high performance size exclusion chromatography, flow field-flow fractionation, enzymatic degradation, and sequential Smith-degradation. Using hydrophobic affinity chromatography, arabic gum was separated into three fractions. Most of the gum had very low protein content (0.35%) and was referred to as an arabinogalactan (AG). It represented 88.4% of the total gum and was found to have a molecular mass of 380000 g/mol. The second fraction represented 10.4% of the total gum and was referred to as an AG-protein complex (AGP) with a molecular mass of 1450000 g/mol. The protein content of the AGP was 11.8%. The smallest fraction (1.2% of the total gum) was referred to as a low molecular-weight glycoprotein (GP) with a molecular mass of 250000 g/mol and a protein content of 47.3% [18].

As with most polymers, the functional properties of gums as thickener, gelling agent, etc., depend on several factors:

- The intrinsic characteristics of the molecule, such as molecular weight, degrees of ionization and branching, etc.
- The extrinsic characteristics of the system, such as pH, ionic strength, temperature, concentration of the other components.

Each gum has certain physical and chemical characteristics that cannot easily be substituted with the use of another polysaccharide. The combination of two or more of these compounds generates new functional properties that they do not have individually; this is the case of the emulsification of oil-water systems, which is achieved with a mixture of gums.

In the food industry, arabic gum is used for ice cream, confectionery, fruit juices, beer, wine, mayonnaise, cheese, jams, dressings, sausages, dietary products, etc.

Arabic gum is obtained by removing the bark of trees such as Acacia Senegal. It is a highly branched heteropolysaccharide consisting of a main chain of β -galactopyranose units to which L-rhamnopyranosyl residues of L-arabinofuranoses and glucuronic acid are joined; molecular weight varies between 250000 and 1000000 g/mol. In its natural state, it is a compact molecule. Two of its main features are its high solubility in water (50%) and the low viscosity that it develops [19,20,21,22].

Many studies have been conducted on the different physical and chemical properties of acacia, particularly on intrinsic viscosity, through analysis with gel permeation chromatography (GPC) coupled to a multi-angle laser light scattering detector, refractive index detector and intrinsic viscosity [23,24], finding small variations from one species to another. In addition, comparative studies were performed between acacia polyacantha and commercial acacia, as in [25].

The structure of acacia gum molecules was studied by SEC-malls, refractometry, and viscometry [26]. It was found that $[\eta]$ of whole gum was 23 cm³/g, R_H was 11.9 nm, and, as the exponent ($n = 0.55$) of the power-law between R_g and M was 530000 g/mol, it was suggested that acacia gum molecules acquire a random coil shape.

Three types of acacia were studied, with molecular weights ranging between 770000 and 400000 g/mol and intrinsic viscosity between 6 and 8 cm³/g, as in [27].

There is a detailed study of Acacia senegal gum and its fractions with a variation in molecular weight of 286000-2690000 g/mol and an intrinsic viscosity of 16.2 to 80.2 cm³/g, as shown in [28].

In a dilute solution, the properties of two specially matured arabic gum samples were compared to the conventional gum using static light scattering; the average molecular weight varied from 622000 to 2540000 g/mol, and intrinsic viscosity from 18.2 to 21.6 cm³/g, as in [29].

The hydrodynamic of κ -carrageenan was studied [30] using a quite different methodology (sedimentation velocity and equilibrium analytical ultracentrifugation and viscometry); the molecular weight and the asymmetric conformation of the κ -carrageenan molecule provide an estimate of its large capacity to imbibe water. According to the data obtained, the hydrodynamic of the polysaccharide in solution was consistent with an extended conformation, while making the crude approximation of a rigid structure, it corresponds to an equivalent hydrodynamic prolate ellipsoid with an aspect ratio of ~15:1 and a frictional ratio f/f_0 of ~7.6, which is consistent with a large hydration (~50 g water per g of dry polysaccharide, corresponding to a molecular expansion of ~100x), which is consistent with one the key functional properties of κ -carrageenans as a high water binder in food.

Studies of the hydrodynamic properties of polysaccharides at different temperatures have been made for dextran [31,32], pectin [33], and chitosan [34,35].

This work presents an experimental study with whole arabic gum in dilute solutions, besides studying the effect of temperature on the hydrodynamic properties of the gum in order to determine their conformational characteristics. The technique used was intrinsic viscosity ($[\eta]$) measurement, which was used to calculate the molecular weight (M) and the rest of the hydrodynamic parameters.

2. Materials and Methods

2.1. Sample Preparation

Five grams of whole arabic gum (STANTON, Argentine) powder were dispersed in pure deionized water and stirred gently stirring at room temperature for 2 h. The dispersions were kept at 3-1°C for 24 h to allow the hydration of the biopolymers. Centrifugation at 9.0g for 60 min allowed the separation of soluble from insoluble fractions. Both fractions were freeze-dried and sealed in zip plastic bags and then kept in desiccators. Finally, whole arabic gum was dissolved in distilled water to prepare a solution of 1% (wt.).

2.2. Density Measurement

The density of solutions and solvents were measured with an Anton Paar densimeter DMA5N.

2.3. Capillary Viscometry

Solutions and reference solvents were analyzed using an Ubbelohde [52] 1C viscometer (IVA) under precise temperature control using a thermostatic bath (Haake 1C).

2.4. Theory

The Hagen-Poiseuille law describes the following equation [36],

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

Where η (poise) is the viscosity, fluid density (ρ g/cm³), A (cm²/s²) is the instrumental constant of the viscometer, and t is the time of draining of liquid (s).

The relative (η_{rel}), specific (η_{sp}), and reduced (η_{red}) viscosities [37] were calculated from

$$\eta_{rel} = \frac{t\rho}{t_0\rho_0} \quad (2)$$

Where t is the flow time of the polysaccharide sample, and t_0 is the flow time of the solvent in seconds.

Where

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

$$\eta_{red} = \frac{\eta_{sp}}{c}$$

A plot of η_{red} vs. concentration yields the intrinsic viscosity, $[\eta]$, at the intercept, while the slope is related to the concentration-dependence [38], k_H ,

$$\eta_{red} = [\eta] + k_H [\eta]^2 c \quad (4)$$

This way of calculating intrinsic viscosity requires several concentrations in order to determine it. The

intrinsic viscosity may be easily calculated by the Solomon-Ciuta single-point equation [39],

$$[\eta] = \frac{1}{c} \sqrt{2\eta_i - 2\ln \eta_r} \quad (5)$$

Solomon-Ciuta, we arrived at the formula that allowed us to calculate the intrinsic viscosity of polymer solutions by a single viscosity determination. The formula was verified for different polymer-solvent systems and the values were consistent with the ones obtained by extrapolation. The double point method could also be used [40].

The relation between M and intrinsic viscosity is given by the Mark-Houwink-Sakurada equation,

$$[\eta] = k (M)^a \quad (6)$$

The calculation of the Mark-Houwink-Sakurada (M-H-S) parameters is carried out by the graphic representation of the following equation:

$$\ln[\eta] = \ln k + a \ln M \quad (7)$$

Where k and a are M-H-S constants; these constants depend on the type of polymer, solvent, and the determinations of the temperature of viscosity. The exponent a is a function of polymer geometry which varies from 0.5 to 2. These constants can be determined experimentally by measuring the intrinsic viscosity of several polymer samples for which the molecular weight has been determined by an independent method (i.e. osmotic pressure or light scattering) [41]. Using the polymer standards, a plot of $\ln[\eta]$ vs $\ln M$ usually gives a straight line. The slope is a value and the intercept is equal to $\ln k$ value [42]. The M-H-S exponent bears the signature of a three-dimensional configuration of a polymer chain in the solvent environment. The a values range from 0 to 0.5 for a rigid sphere in ideal solvent, from 0.5 to 0.8 for a random coil in good solvent, and from 0.8 to 2 for a rigid or rod-like polymer (stiff chain). The fact that the intrinsic viscosity of a given polymer sample varies according to the solvent used provides and insight into the general shape of the polymer molecules in solution. A long-chain polymer molecule in solution takes on a somewhat kinked or curled shape, intermediate between a tightly curled mass (coil) and a rigid linear configuration. All possible degrees of curling may be displayed by any molecule, but there will be an average configuration that will depend on the solvent. In a good solvent that shows a zero or negative heat of mixing with the polymer, the molecule is fairly loosely extended, and the intrinsic viscosity is high. The Mark-Houwink “ a ” constant is close to 0.75 or higher for these “good” solvents. In a “poor” solvent that shows a positive heat of mixing, segments of a polymer molecule attract each other in solution more strongly than they attract the surrounding solvent molecules. The polymer molecule assumes a tighter configuration and the solution has a lower intrinsic viscosity. The M-H-S “ a ” constant is close to 0.5 in “poor” solvents [43]. For a rigid or rod-like polymer molecule that is greatly extended in solution, the M-H-S “ a ” constant approaches a value of 2.0. The hydrodynamic radius (R_H) for a sphere ($v_{(p)} = 2.5000$) is given by the Einstein relation [44],

$$[\eta]M = v_{(p)} N_A \frac{3}{4} \pi (R_H)^3 \quad (8)$$

The viscosity of liquids is highly dependent on temperature and its complex relations [45]. The change of viscosity at different temperatures is commonly calculated with an equation of the Arrhenius form:

$$\eta = A_{vf} \exp\left(\frac{E_{avf}}{RT}\right) \quad (9)$$

Where η is the viscosity (poise); E_{avf} is the viscous flow activation energy (cal/mol); R is the gas constant (1.98cal/mol K), and T is temperature (K). The pre-exponential factor A_{vf} is considered independent or approximately independent of temperature. Equation 9 is convenient for calculating E_{avf} in a discrete range of temperatures.

For simplicity reasons, proteins and macromolecules may be treated as rigid molecules in a hydrodynamic study [46]. It is worth noting that the size of proteins is much bigger than that of solvent (water) molecules [47]. Thus, D from spherical proteins in dilute aqueous solutions can be approximately described by the Stokes-Einstein equation, which assumes a rigid solute sphere diffusing in a solvent continuum. The correlation of this equation with the molecular weight and the viscosity is as follows:

$$D(\text{cm}^2 / \text{s}) = 8.34 \times 10^{-8} \left(\frac{T}{\eta M^{1/3}} \right) \quad (10)$$

The diffusion coefficient is an important physical-chemistry property of biological molecules. In several biological and industrial processes, the diffusivity value is required for designing and analysing the process. For example, the protein diffusion coefficients are crucial for analysis, extraction, and transport in porous media and drying processes. In addition, diffusion coefficients are related to hydrodynamic properties that can provide information about the size and shape of macromolecules and proteins.

Hydrodynamic properties such as η and D , intrinsic viscosity $[\eta]$, as well as equilibrium solution properties such as the hydrodynamic radius R_H , can be combined to construct dimensionless quantities that are universal in the sense of being independent of the size of the macromolecular particle, while depending more or less on its shape or conformation.

Typical examples are the Scheraga-Mandelkern parameter β given by

$$\beta = \frac{\eta_0}{f} \left(\frac{M[\eta]}{100} \right)^{1/3} \quad (11)$$

The friction coefficient f is derived from the measurement of the diffusion coefficient as $f = k_B T/D$ where k_B is the Boltzmann constant and T the absolute temperature. In equation 11, η_0 is the solvent viscosity. Other classical size-independent combinations are the Flory parameters, which combine intrinsic viscosity $[\eta]$ and the radius of gyration R_g :

$$\varphi_0 = \frac{[\eta]M}{6^{3/2} R_g^3} \quad (12)$$

another combines the friction coefficient with the radius of gyration:

$$P_0 = \frac{f}{6\eta R_g} \quad (13)$$

These quantities have been proposed along the years, at different times and by various eminent scientists, after whom they are named. Because of the diversity of their origin, the set of classical universal size-independent quantities suffers some inconveniences. Two of them, unimportant but somehow cumbersome, are related to the diversity not only of the symbols employed to represent them, but mainly of the disparity of their numerical values and of the order of magnitude for typical cases; thus, β takes the values of 2.112×10^6 and about 2.3×10^6 for a sphere and a random coil, respectively, while the values for these two structures in the case of the ϕ_0 are 9.23×10^{23} and $2.60 \times 10^{23} \text{ mol}^{-1}$. Thus, it is accepted that, for every flexible-chain polymer in a θ (ideal) solvent [48], there is a universal value of $\phi_0 = 2.50 \times 10^{23} \text{ mol}^{-1}$. Therefore, the values deviate from the ideal conditions of β , ϕ_0 and P_0 deviate from linearity of the biopolymer, ie is hyperbranched macromolecule.

In the case of rigid macromolecules [49], it is also conventional, particularly for rigid macromolecules, to combine a solution property with the volume of the particle itself, or with a quantity directly derived from it. Thus, it is a common practice to express the frictional coefficient of rigid structures as

$$P \equiv \frac{f}{f_0} = \frac{f}{6\pi\eta_0 (3V/4\pi)^{1/3}} \quad (14)$$

Where (in our notation) f_0 is the frictional coefficient of a sphere with the same hydrodynamic volume V (hydrated or solvated) as the particle.

The term f/f_0 is sometimes denoted as P is called the Perrin number, where f is the friction coefficient expression. A similar combination involves intrinsic viscosity and specific volume:

$$v_{(p)} \equiv \frac{[\eta]}{V_s} \quad (15)$$

$v_{(p)}$ is called Einstein viscosity increment, and V_s is specific volume (cm^3/g). For ellipsoids, as studied by Simha, $v_{(p)}$ is a function of the axial ratio [50].

The corresponding value of 'hydration' of the molecule, δ , is defined by

$$\delta = (V_s - \bar{v})\rho_0 \quad (16)$$

and was found to be ~ 50 g solvent bound per g of solute. Nevertheless, because of the approximations we have made, the actual numerical value must be treated with great caution; this treatment does however suggest that polysaccharides are highly expanded, but perhaps not to the same extent as coil-like polysaccharide structures [51].

3. Results and Discussion

3.1. Arrhenius Plot and Chain Flexibility

The Arrhenius plot, in Figure 1-a, shows the linear relationship between the \ln of viscosity and the inverse temperature, where the value obtained from E_{avf} is 4000

cal/mol, and that of A_{vf} is $1.29 \times 10^{-5} \text{ g/cm s}$, with $\sigma^2 0.9898$. This occurs due to the higher resistance to flow of bigger biopolymers, which, in consequence, require more energy. Figure 1-b shows that the intrinsic viscosity of whole arabic gum is influenced by temperature.

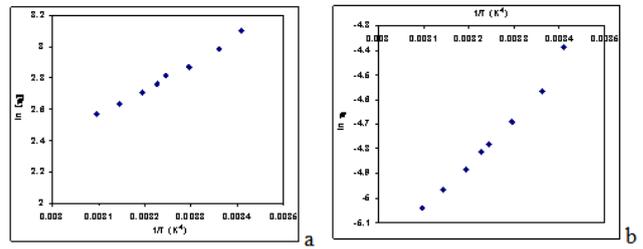


Figure 1. a- Logarithmic plot of the intrinsic viscosity ($[\eta]$, cm^3/g) in function of $1/T$ of WAG. b- Influence of temperature on viscosity (Arrhenius plot)

Within a limited temperature range, the variation of the viscosity of a polymer solution is generally similar to that which is usual in liquids (equation 9). According to the Stokes-Einstein equation, the diffusion coefficient is inversely proportional to the solution viscosity, which increases with temperature. Hence, a lower diffusion coefficient corresponds to a smaller molecule (see Figure 2-a). In the case of dextran, there is a mathematical relationship between D and T and, in turn, between D and M [53].

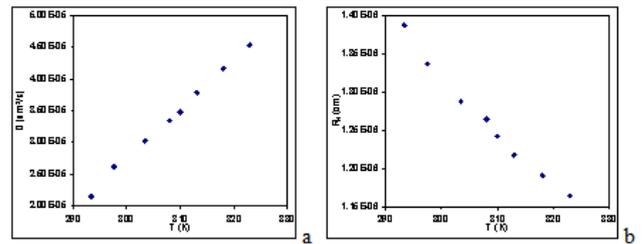


Figure 2. Whole arabic gum data as a function of temperature: a- Diffusion Coefficient, b- Hydrodynamic Radius

Table 1. Dates of intrinsic viscosity, Mark-Houwink-Sakurada parameters as a function of temperature

T (K)	Mark-Houwink-Sakurada Parameters		
	$[\eta]$ (cm^3/g)	k (cm^3/g)	a
293.36	22.20	0.01301	0.5496
298.16	19.81	0.01311	0.5406
303.46	17.71	0.01316	0.5321
308.16	16.77	0.01321	0.5278
309.86	15.90	0.01326	0.5235
313.16	14.98	0.01331	0.5189
318.16	14.01	0.01338	0.5135
322.96	13.12	0.01342	0.5085

The hydrodynamic radius and intrinsic viscosity are higher at high molecular weights and decrease with increasing temperature [54,55] (Figure 2-b). Table 1 shows the values of the parameters of Mark-Houwink-Sakurada of a and k for each temperature. These studies of M-H-S parameters are usually carried out at a given temperature, obtaining a consistent result but in a very limited range of temperatures [26].

The molecular weight of WAG calculated is 760000 g/mol. The values of "a" at different temperatures for WAG-water system, indicating that it has a random-coil conformation with a tendency to acquire spherical shape

to 322.16 K. The “ a ” values given at different temperatures shows that this polysaccharide in aqueous solution behaves in a conformation from random coil or spherical, as observed by other authors as in [26,27,28,29]. However, the values calculated in this work ensure that WAG in this system has the conformation of a random coil with a tendency to an ideal system when T increases.

Furthermore, the influence of temperature on the intrinsic viscosity is given by the chain flexibility parameter ($d\ln[\eta]/dT$), which provides information about the conformation of the macromolecule chain in solution [29]. The chain flexibility parameter ($d\ln[\eta]/dT = 1661.5 \text{ K}^{-1}$) indicates that whole arabic gum has low chain flexibility.

3.2. Hydrodynamic Properties

The analysis of the values of the hydrodynamic properties of WAG in aqueous solution can be seen in Figure 3 and Table 2; all vary with temperature. β values increase from 1.70 to 1.84×10^6 when temperature increases, indicating the non-linearity of WAG. The values of ϕ_0 and P_0 decrease from 4.54 to $2.68 \times 10^{23} \text{ mol}^{-1}$ and from 5.26 to 4.23 respectively, which shows the low flexibility of the particles. The value of P decreases from 3.90 to 3.62 and $\nu_{(P)}$ with 2.55, which confirms that WAG in aqueous solution is a biopolymer with a random coil shape, and with a tendency to compaction when temperature increases (R_H decreases) [56].

Table 2. Hydrodynamic properties of WAG in water solution at different temperatures

T (K)	Hydrodynamic Parameters		
	P	$\beta \times 10^{-6}$	$\phi_0 \times 10^{23}$
293.36	3.90	1.70	4.54
298.16	3.67	1.81	4.05
303.46	3.62	1.84	3.62
308.16	3.67	1.81	3.43
309.86	3.71	1.79	3.25
313.16	3.65	1.82	3.06
318.16	3.68	1.81	2.87
322.96	3.75	1.78	2.68

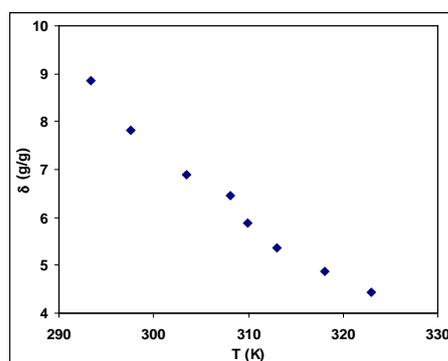


Figure 3. Hydration value of WAG in water solution as a function of temperature

The value of δ decreases from 8.86 to 4.46g/g as expected; this phenomenon occurs because of the loss of water due to the compression of WAG as colloidal particle, by the effect of the increasing temperature [57]; a similar phenomenon is the drying gels to form films, or the compaction of a sponge.

3.3. Mark-Howink-Sakurada Properties

The Mark-Houwink-Sakurada parameters for WAG vary with the dissolvent and with temperature. This is because the hydrodynamic radius of the macromolecules changes with the type solution and with temperature via changes in their chain flexibility. In a good solvent ($a \rightarrow 0.5000$), a temperature increase results in a decrease of intrinsic viscosity and in a less-extended conformation ($D >$ and $R_H <$) because the entropy value increases with an increase in temperature, which is unfavorable for an extended conformation ($E_{avf} >$). In the case of a poor solvent, a temperature increase causes an increase in entropy, which favors a less-extended conformation.

The range of M determined by other authors [22-27] varies from 254000-770000 g / mol, with ranges of $[\eta]$ from 6 to 26.5 cm³ values of “ a ” from 0.4718 to 0.5469. According to studied in this work, all the parameters determined they are situated within the ranges established by other authors mentioned above. The main difference is that the rest of the authors only work at a single temperature of 20 or 25°C, which makes difficult a comparison to other T.

Mark-Houwink-Sakurada values of “ a ” confirm that for these conditions, WAG behaves as a random coil biopolymer. Empiric functions can be used to facilitate the calculation of these parameters in an acceptable way. As the equations that relate the parameters of M-H-S with temperature, they can be related to thermodynamic parameters, as E_{avf} , these parameters depending on the properties of the solute with the solvent.

4. Conclusions

The numerical value of “ a ” indicates that WAG acquires a random coil shape in aqueous solution; it decreases as temperature increases; and k demonstrates that the value of the parameters increases when temperature increases [33,58]. The M-H-S parameters have temperature functionality. Given the values of “ a ” is the random-coil conformation. The values of the M-H-S parameters may be universalized with certain precautions, for calculating of molecular weight in a temperature range of 25-50°C.

Due to the lack of data on the uniformity of intrinsic viscosity measurements in the WAG/water system, M-H-S values facilitate the calculation of molecular weight ($M = 760000 \text{ g/mol}$) in an acceptable range temperatures.

For this system, WAG possesses a random-coil conformation as a nonlinear biopolymer which tends to compact with increasing temperature (where R_H and $[\eta]$ decreases), which require an increase in power consumption due to the difficulty of flow (where D and E_{avf} increase) [59,60]. This phenomenon is observed in the case of ideal solvents. This conclusion is supported by the data obtained from the hydrodynamic properties analyzed.

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