Research Article

Thermodynamic Properties of Gum Arabic and *AcaiaPolyacantha* Gum Solutions

Malik A. Abdelrahman Elsheikh[†,†], Saphwan Al-Assaf[‡], Mohamed E. Osman[†], Elfatih A. Hassan[†], Glyno.Philips[‡,‡]

[†]Chemistry Department, Faculty of Applied Medical Science (Turabah), Taif University, Saudi Arabia
[‡]Glyn O. Phillips Hydrocolloids Research Centre, Glyndwr University, PlasCoch, Mold Road, WrexhamLL11 2AW, UK.
[†]Department of Chemistry, College of Science, Sudan University of Science and Technology, Sudan
[‡]Phillips Hydrocolloid Research Ltd., 45 Old Bond Street, London W1S 4AQ, UK.

Abstract

The thermodynamic properties of A. senegal gum, A. seyal gum (gum arabic) and A. polyacantha gum solutions were measured and calculated. The partial specific volume of gum, partial specific volume of the solvent (water) and the volume fractions were measured and calculated. The results show that the sequences of the specific volumes of the gums under studies are in the order of decreasing molecular weight of these gums. The results of density of solid gums indicate that A. seyal molecule is more compact than A. senegal gum molecule and A. polyacantha gum molecule. The chemical potentials have been calculated from osmotic measurement for different concentrations, and then the second virial coefficients, the free energy of mixing and the gums–water interaction parameter χ were calculated. The obtained results indicate that A. polyacantha gum thermodynamically resembles A. senegal gum and they were interacting with water more than A. seyal gum.

Keywords: *Gum Arabic; Acacia senegal; Acacia seyal; Acacia polyacantha; Thermodynamic properties.*

1. Introduction

1.1 Gum arabic

Gum arabic is dried exudates from the stems and branches of *Acacia senegal* and *Acacia seyal* trees (family *Leguninosae*) [1]. *Acacia polyacantha* (family *Leguninosae*) exudates are closely related, and can be distinguished from the *A. senegal* exudates by differences in physicochemical characteristic [2,3].

Crud gum arabic, (has different uses) is a complex copolymer of polysaccharide with high molecular mass and complex structure [4-7]. It is a branched molecule [8] with protein content of about 2.0–2.5%. Gum is present in a mixed calcium, magnesium and potassium salts of polysaccharidic acid [9,10]. It is composed of six carbohydrate moieties: galactopyranose, arabinopyranose, arabinofuranose, ramnopyranose, glucuropyranosyluronic acid and 4-o-methylglucuropyronosyl uronic acid [11-14]. Polysaccharides that contain arabinose and galactose as their major constituents are called arabinogalactan (AG) [15,16]. So all *A.* gums chemically are Arabinogalactan-proteins (AGP), and they are described as 'heteropolymolecular', i.e. having either a variation in monomer composition and/or a variation in the mode of linking and branching of the monomer units, in addition to a distribution in molecular weight[17]. The weight average molecular weight of *A. senegal*, *A. seyal* and *A. polyacantha* are 8.64×10^5, 1.55×10^6 and 3.18×10^5 respectively, while the number average molecular weight are 2.86×10^5, 5.16×10^5 and 1.43×10^5 respectively [17]. The chemical constituent, the molecular structure and the molecular weight affect the behavior of molecules in solutions. Gum arabic finds a wide application in food and beverages as a natural emulsifier particularly for citrus water and low viscosity [18- 20]. In food industry, gum arabic is primarily used in confectionery, bakery, dairy, beverage, microencapsulating agent and to a lesser extent in textiles, ceramics, lithography, cosmetics and pharmaceutical industry[21].

1.2 Thermodynamics study

To understand the behavior of gum molecule in solutions it is necessary to measure and calculate some thermodynamic parameters and functions. Thermodynamics has an immense predictive power and the thermodynamic laws can be used to predict the direction in which a process would proceed. Thermodynamic parameters and functions include the density of solid gum, the partial specific volume, the volume fraction, the osmotic pressure, the chemical potential of solute and solvent, the second virial coefficient, the gum-solvent interaction parameter χ and the free energy of mixing.

The density and the partial specific volume of the solid gum give a good idea about the distance between the molecules. The density of the gum can be determined by pyknometer using solvent that gum is insoluble in and cannot be affected by it. Gradient tube method also used to determine the density of the polymer [22].

In general the partial molar volume of a substance in a mixture is the change in volume per mole added to a large volume of the mixture. If the molecular masses of the components are not known exactly (gum molecule), then their mole fractions cannot be calculated. It is more convenient to use specific partial functions, i.e., functions referred to one gram rather than to one mole of the component. The partial specific volume of component can be measured by density measurements performed at constant molalities of the components[23-25].

The partial molar (specific) volume of a solution can be calculated from the density and chemical compositions[22,26]. For a binary solution, the partial molar (specific) volume at constant temperature and pressure could be calculated by graphical method. There are two graphical methods for calculating partial molar (specific) volume [22].

One of the most function characteristics in the behavior of each component in a solution is the chemical potential of the component. It is equal the change in internal energy of a solution in addition to an infinitely small number of moles of ith component, referred to that amount of substance at constant volume, entropy, and quantity of each of the other components. Moreover, the chemical potential could be calculated from the partial specific volume and osmotic pressure [22,26 and 27].

Osmosis is the phenomenon of penetration of a solvent into a solution through a semi permeable membrane. The tendency of solvent molecules to pass spontaneously into a solution, is due to the inequality of chemical potential of pure solvent and solution estimated quantitatively by osmotic pressure, which has the dimension of pressure (atm , Nm^{-2}). The osmotic pressure, the density of solvent and the density of solid gum measurements lead to calculate the second virial coefficients (A_2), and then to calculate the parameter χ_1 [26,27], which are the measure of thermodynamic affinity of a solvent for the polymer, or a measure of the quality of a solvent. The smaller χ_1, means thermodynamically is the better solvent. For poor solvents, $A_2 < 0$; $\chi_1 > 0.5$, for good ones, $A_2 > 0$; $\chi_1 < 0.5$, and for ideals ones $A_2 = 0$; $\chi_1 = 0.5$ [22].   The parameter χ_1 can be determined from osmotic pressure [22]. The free energy of mixing (ΔG^m) could be calculated from the chemical potential and the weight fractions of solute and solvent, its value is an indication of the interacting of the solute with solvent[26].

The aim of this work is to investigate the thermodynamic properties of *A. senegal* and *A. seyal* and *A. polyacantha* gum solutions and to determine some important thermodynamic parameters and functions. This includes the density of solid gum, the partial specific volume, the volume fraction, the osmotic pressure, the chemical potential of solute and solvent, the second virial coefficient, the gum-water interaction parameter χ and the free energy of mixing. The other aim is to compare and contrast the thermodynamic properties of *A. senegal, A. seyal* and *A. polyacantha* gum solutions.

2. Experimental

2.1 Materials

Three samples of each type of *Acacia* gum (*A. seyal, A. senegal* and *A. Polyacantha*) were collected, and one sample of each type had been made by coning and quartering [28]. The gum samples used in this study were cleaned by hand to ensure they were relatively free from sand, dust and bark impurities. Then were grounded using pestle and mortar, backed in labeled self sealing polyethylene bags.

2.2 Thermodynamic measurements and calculations

The thermodynamic of polymer solutions can be applicable to gum solutions since gum molecules are classified as biopolymer molecules, the gum arabic solutions were characterized by different thermodynamic parameters.

2.2.1 Partial specific volume of solvent

The tangent method was used [22] by dissolving a constant weight of gum sample   in different weights of water. The densities of solutions were determined by a pyknometer at 294K, and then the total volumes of the solutions were calculated. The volumes of solutions were plotted against weight of solutions. The partial specific volume of water is equal to the   ∂ V / ∂ g which can be calculated from graph [22, 26].

2.2.2 Partial specific volume of the gum

Tangent method was used in which different weights of gum were dissolved in a constant weight of water. The densities of solutions were determined by a pyknometer at 294K and then the total volumes of the solutions were calculated. A graph of the volume of solutions versus the weights of gum was plotted. The partial specific volume of gum sample is equal to the   ∂ V / ∂ g which can be calculated from the graph[22].

2.2.3 Density of solid gum

The density of *A.* gums can be determined at 294K by weighting out a certain weight of the gum sample with a certain weight of acetone (gum insoluble in acetone) in a density bottle. First the density of acetone was determined, second the volume occupied by gum was

calculated, from the volume occupied by the gum and the weight of the gum, the density of the gum was calculated.

2.2.4 Osmotic pressure of gum solutions

Osmotic pressures of gum solutions were measured using osmomat[R]050 colloidal osmometer at 294K by preparing different concentrations of gum solutions.

3. Results and discussion

The partial specific volume of the solvent and solute were calculated by using the tangent method [22]. From the slopes values (Fig. 1-6), the partial specific volumes of solvent (water) and solute (gum) were obtained. The partial specific volume of water and *seyal* gum for a *A. seyal* gum solutions were obtained from the slopes value (Fig.1 and 2), which were found to be 1.0016 $cm^3\ g^{-1}$ and 0.6426 $cm^3\ g^{-1}$ respectively. For *A. senegal* gum solutions the partial specific volume of water and *A. senegal* gum (Fig. 3 and 4) were found to be 1.0018 $cm^3\ g^{-1}$ and 0.6425 $cm^3\ g^{-1}$ respectively, for *A. polyacantha* gum solutions (Fig.5 and 6) were found to be 1.0021 $cm^3\ g^{-1}$ for water and 0.6421 $cm^3\ g^{-1}$ for *A. polyacantha* gum. The results show that the sequences of the partial specific volume of the gums under studies are in the order of decreasing molecular weight of these gums i.e. *A. seyal*, *A. senegal* and *A. polyacantha*. The partial specific volume of *A. senegal* is very close to that of the *A. seyal*, although there is noticeably difference between their molecular masses; This, may be, due to the compactness of *A. seyal* molecules which is greater than the *A. senegal* molecules.The results of the partial specific volume of gums also show that A*. senegal* molecule expands in water more than *A. seyal* molecule.

The volume fractions of water (ϕ_1) and that of gums (ϕ_2) in gums solutions of different concentrations (Table 1) were calculated using equations (1) and (2) [22]. The results show that A*. seyal* has the largest volume fraction followed by A*. senegal* and finally A*. polycacntha*. The sequences of the volumes fraction were related to the sequences of the weight average molecular weight and the partial specific volumes of the exudates samples of the gums.

$$\phi_1 = \bar{V}_1 / \bar{V}_1 + \bar{V}_2 \qquad (1)$$

$$\phi_2 = \bar{V}_2 / \bar{V}_1 + \bar{V}_2 \qquad (2)$$

Where:
$\bar{V}_1$= partial specific volume of the solvent (water)
$\bar{V}_2$ = partial specific volume of the solute (gum)

Table 2 shows the osmotic pressures of different concentrations of *A. seyal*, *A. senegal* and *A. polyacantha*. The results obtained show that at the same concentration of *A. polyacantha* and *A. senegal* have high value of osmotic pressure than the *A. seyal*. This mean that they interact with water more than *A. seyal* and this was due to the structural variations. According to equation (3) [22], it was possible to determine the change in chemical potential of water as a solvent in different gums solutions (Table 3). The results show that the changes in chemical potential of water in *A. polyacantha* gum solutions is greater than the changes in chemical potential of water in *A. senegal* gum solutions which is greater than the changes in chemical potential of *A. seyal* gum solutions. The results also show that the changes in chemical potential of *A. senegal* values are more closed to the values of *A. polyacantha* than *A. seyal* values.

$$\mu_1 - \mu_1^0 = \Delta\mu_1 = -\bar{V}_1 \Pi \qquad (3)$$

Where:
$\Delta\mu_1$ change in chemical potential of water
$\bar{V}_1$ partial specific volume of water
Π osmotic pressure of the solution

The second virial coefficient (A_2) was obtained from the slope of the graph by plotting (Π /c)½ versus concentration (c) equation (4) [22,29,30] of different *A.* gum samples (Fig.7-9).The second virial coefficients were found to be 0.97x10^{-4}, 1.85x10^{-4} and 2.22x10^{-4} for the *A. seyal*, *A. senegal* and *A. polyacantha* respectively (Table 4). The results explained that water is good solvent for the three types of gums ($A_2 > 0$), also the result indicate that *A. polyacantha* and *A. senegal* gum have closer and higher values of second virial coefficient than *A. seyal*, indicating that both of them were interacting with water more than the *A. seyal* gum.

$$(\Pi /c)^{1/2} = (RT/M_n)^{1/2} + (RT/M_n)^{1/2} A_2 M_n/2 \ \mathbf{c}(4)$$

Where:
R is the gas constant
T is the temperature in Kelvin

The number average molecular weights (M_n) of *A. seyal*, *A. senegal* and *A. polyacantha* were calculated from the intercept of graphs of Fig. 7, 8, and 9 using equation (4), they were found to be 4.7x10^5, 2.4x10^5 and 1.9x10^5 respectively. The values of the number average molecular weight obtained from the osmotic pressure measurements were close to the values that were obtained by gel permeation chromatography for the same samples[17] as shown in Table 5. These very close results obtained by the gel permeation chromatography and the osmotic pressure methods improve the accuracy of all results obtained in this study by the osmotic pressure technique.

The densities of gums reported in Table 6 show that the *A. senegal* gum has low value (1.465 g cm^{-3}). This indicates that the distance between the molecules in *A. senegal* and the compactness of the molecules is less

than the distance between the molecules and the compactness of the molecules in both: *A. seyal* and *A. polyacantha*. This agrees with the ratio of weight average molecular weight to the radius of gyration [17] as shown in Table 7, the less ratio (*A. senegal* gum 0.22×10^5) shows the lowest compactness of the molecule and hence the lowest density. The highest density of *A. seyal* (1.485gm-3) and highest ratio of Mw/Rg ratio (0.41×10^5) indicate the high compactness of the molecules i.e. *A. seyal* molecules have less distances between them followed by *A. polyacantha* which is followed by *A. senegal*. This is consistence with Hassan, radius of gyration(Rg) measurements (Rg for *seyal* = 28 nm, Rg for *sengal* = 32 nm) [31].

The changes in chemical potential of the three gum samples were calculated by plotting weight fractions of water divided by weight fractions of gum (ω_1/ω_2) versus the change in chemical potential of water ($\Delta\mu_1$) (Fig.10,11 and 12). The areas under the curve, bound by ordinates corresponding to $\Delta\mu_2^{/}$ which was less than the true areas values were obtained by $\Delta\mu_2$ (true change chemical potential of gum), to correct these areas [22], a graph of $\Delta\mu_2^{/}$ versus weight fraction of water (ω_1) was plotted to obtain segment A from the intercepts of Fig. 13-15, and then to obtain the true values of gums ($\Delta\mu_2$) by using equation (5). Table 8 shows that the change in chemical potential of *A. seyal*, *A. senegal* and *A. polyacantha*, the results show that *A. polyacantha* gum has great changes in chemical potential. This means it interacts with water more than *A. senegal* which interacts with water more than *A. seyal*.

$$\Delta\mu_2 = \Delta\mu_2^{/} + A \qquad (5)$$

The change in free energy of the mixing of the three gum samples were calculated by using equation (6) [22]. The obtained results were reported in Table 9. The results show that *A. polyacantha* and *A. sengal* have closed values, those were higher than the values obtained for *A. seyal* values. This means that *A. polyacantha* interacts with water more strongly than*A. senegal*, which interacts more strongly than *A. seyal*.

$$\Delta G^m = \omega_1\Delta\mu_1 + \omega_2\Delta\mu_2 \qquad (6)$$

The three *A.* gums samples under studies have large values of osmotic pressure, great changes in chemical potential and free energy of mixing of the entire system and positive values of second virtial coefficient. This indicates that water is a good solvent for the three types of A. gums samples. The order of the interaction with water is greater in *A. polyacantha*, then in *A. senegal*and the smaller is in *A. seyal.*

The gum solvent interaction parameter χ_1 for the gums samples under studies were calculated from equation (7) [22] using the density of gums and their molecular masses. The results obtained were reported in the Table 10. The results show that the values of parameter χ_1 of the three gums samples are less than 0.5.

This indicates that water have good thermodynamically affinity to the three types of gum under studies. Moreover, water has better affinity to *A. polyacantha* rather than *A. senegal* and *A.seyal*.

$$A_2 = (\rho_1/M_1\rho_2^2)(1/2 - \chi_1) \qquad (7)$$

Where:
ρ_1&ρ_2 are the density of solvent (water) and solute (gum) respectively.
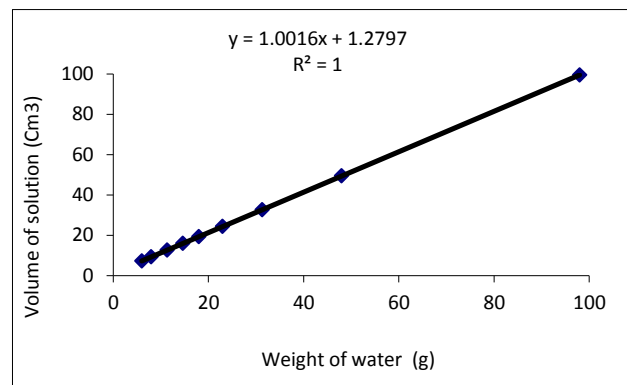M_1 is molecular mass of solvent (water).

Fig. 1 The partial specific volume of water for *A.seyal* gum solutions
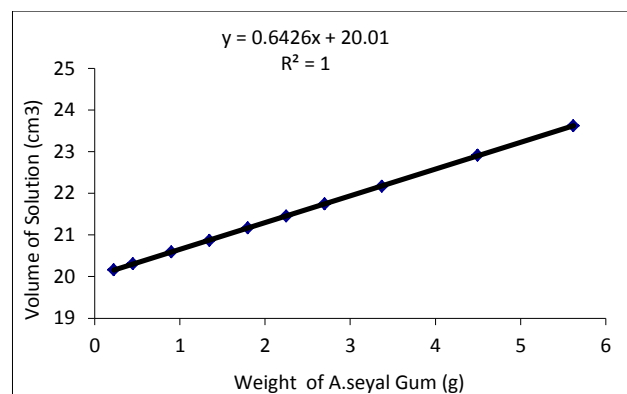

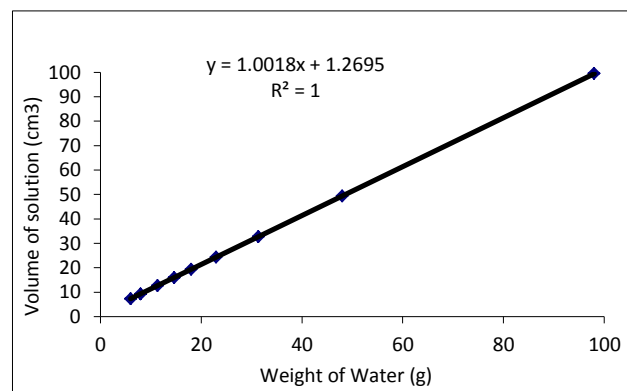**Fig. 2** The partial specific volume of *A. seyal*gum for*A. seyal* gum solutions

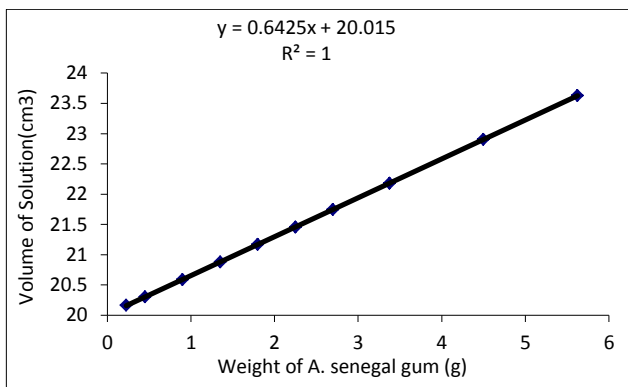**Fig. 3** The partial specific volume of water for *A. senegal* gum solutions

Fig.4 The partial specific volume of *A. senegal* gum for *A. senegal* gum solution
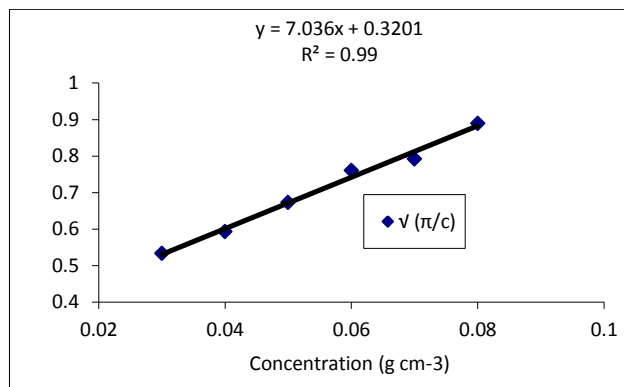

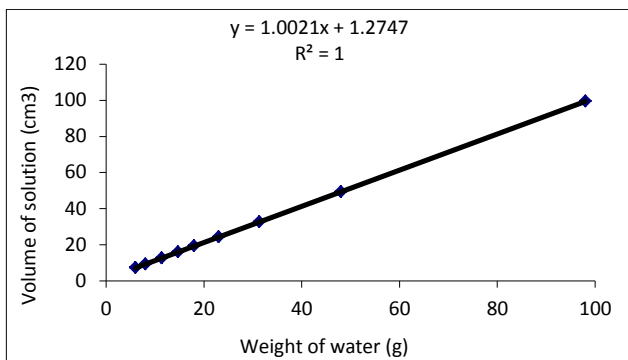**Fig. 8** The osmotic pressure concentration profile of *A. senegal* gum solutions

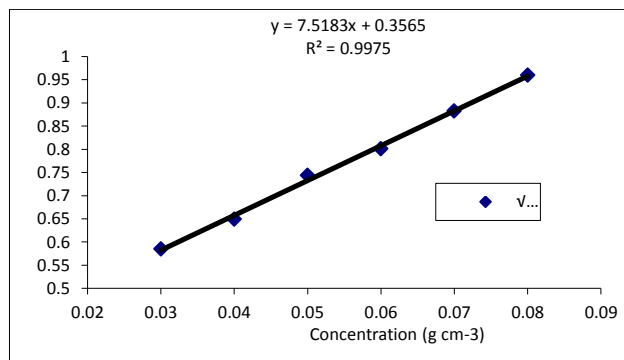**Fig. 5** The partial specific volume of water for *A. polyacantha* gum solutions

Fig. 9 The osmotic pressure concentration profile of *A. polyacantha* gum solutions
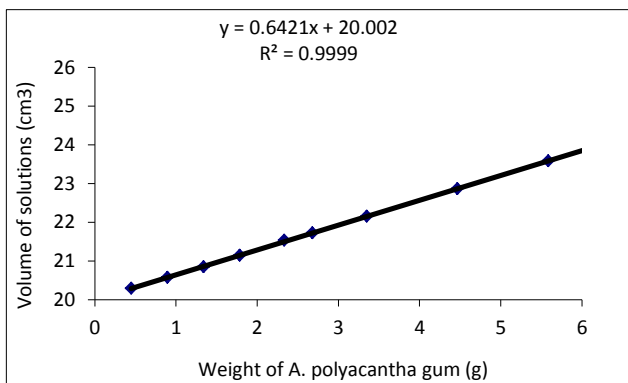

Fig. 6 The partial specific volume of *A. polyacantha* gum for *A. polyacantha* gum solutions

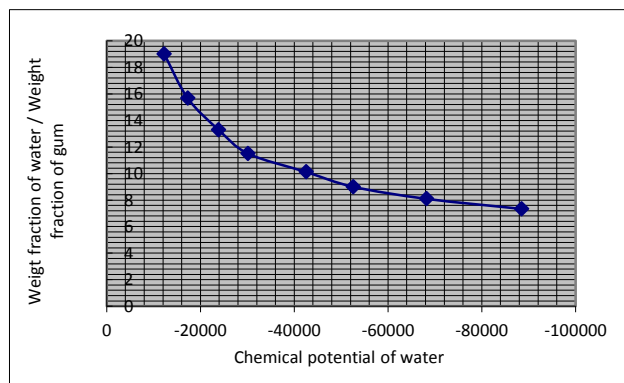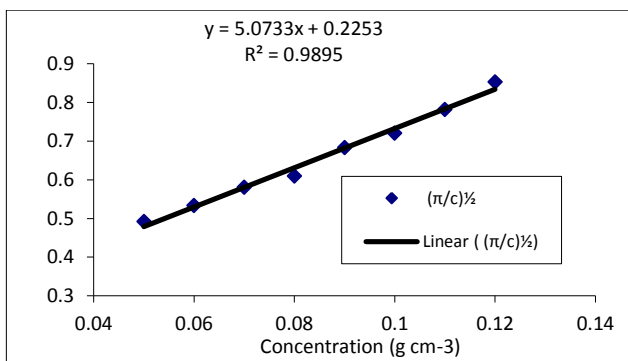**Fig.10** The change in chemical potential of a *A. seyal* gum $(\Delta\mu_2^{/})$

Fig. 7 The osmotic pressure concentration profile of *A. seyal* gum solutions
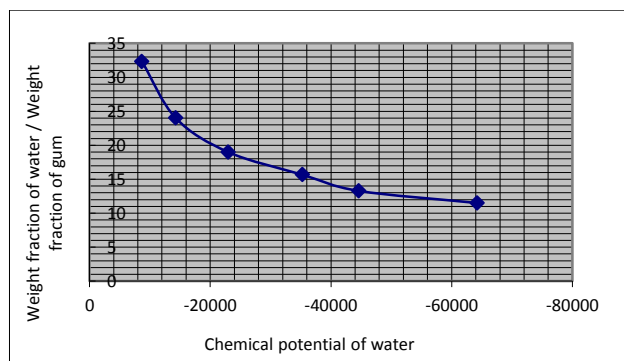

Fig. 11 The change in chemical potential of *A. senegal* gum $(\Delta\mu_2^{/})$

Table 1 The volume fractions of water (ϕ_1) and *A*.gums (ϕ_2) in different gums sample solutions

Gum sample	Water volume fraction(ϕ_1)	Gum volume fraction(ϕ_2)
*A. seyal*solution	0.6092	0.3908
*A. senegal*solution	0.6093	0.3907
*A. polyacantha*solution	0.6095	0.3905

Table 2 The osmotic pressures of *A. seyal, A. senegal* and *A. polyacantha*at different concentrations

Concentration (g cm^{-3})	Osmotic pressure Π (mmHg)		
	A. seyal	*A. senegal*	*A. polyacantha*
0.03	*ND	6.50	7.80
0.04	ND	10.70	12.80
0.05	**9.20**	**17.20**	**21.00**
0.06	**12.95**	**26.40**	**29.25**
0.07	**17.90**	**33.40**	**41.40**
0.08	**22.60**	**48.10**	**56.00**
0.09	31.90	ND	ND
0.10	39.40	ND	ND
0.11	51.10	ND	ND
0.12	66.30	ND	ND

* ND = not determined.

Table 3 The changes in chemical potentials of water ($\Delta\mu_1$) for different *A.* gums solutions

Concentration (g cm^{-3})	$\Delta\mu_1$(joule g^{-1})		
	A. seyal	*A. senegal*	*A. polyacantha*
0.03	*ND	-0.86801×10^{-3}	-1.0419×10^{-3}
0.04	ND	-1.4289×10^{-3}	-1.7098×10^{-3}
0.05	**-1.2283×10^{-3}**	**-2.2969×10^{-3}**	**-2.8059×10^{-3}**
0.06	**-1.7290×10^{-3}**	**-3.5255×10^{-3}**	**-3.9072×10^{-3}**
0.07	**-2.3900×10^{-3}**	**-4.4602×10^{-3}**	**-5.5302×10^{-3}**
0.08	**-3.0174×10^{-3}**	**-6.4233×10^{-3}**	**-7.4805×10^{-3}**
0.09	-4.2591×10^{-3}	ND	ND
0.10	-5.2604×10^{-3}	ND	ND
0.11	-6.8225×10^{-3}	ND	ND
0.12	-8.8519×10^{-3}	ND	ND

* ND = not determined.

Table 4 The second virial coefficients of the *A. seyal* gum, *A. senegal* gum and *A. polacantha* gum

Gum sample	Second virial coefficient (A_2)
A. seyal	0.97×10^{-4}
A. Senegal	1.85×10^{-4}
A. polyacantha	2.22×10^{-4}

Table 5 The number average molecular weight (Mn) of samples by osmotic method and GPC method[17]

Gum sample	Mn	
	Osmotic pressure method	GPC method
A. seyal	4.7×10^5	5.16×10^5
A. Senegal	2.4×10^5	2.86×10^5
A. polyacantha	1.9×10^5	1.43×10^5

Table 6 The densities of *A. seyal, A. senegal* and *A. polyacantha* gums

Particle size (μm)	Density of solid sample (g cm^{-3})		
	A. seyal	*A. senegal*	*A. polyacantha*
Less than 125	1.488	1.479	1.482
Between 125–500	1.483	1.461	1.476
Larger than 500	1.483	1.456	1.465
The average value	**1.485**	**1.465**	**1.474**

Table 7 The ratio between weight average molecular weight and radius of   gyration of the gums [17]

Gum sample	Weight average molecular weight (Mw)	Radius of gyration Rg (nm)	The ratio Mw/Rg
seyal	15.5×10^5	38	0.41×10^5
senegal	8.64×10^5	40	0.22×10^5
polyacantha	3.18×10^5	10	0.32×10^5

Table 8 The changes in chemical potentials *of A. seyal,A. Senegal* and *A. polyacantha* ($\Delta\mu_2$)

Concentration (g cm^{-3})	$\Delta\mu_2$(joule g^{-1})		
	A. seyal	*A. senegal*	*A. polyacantha*
0.03	*ND	-14.1093×10^{-2}	-23.5969×10^{-2}
0.04	ND	-13.9637×10^{-2}	-23.3534×10^{-2}
0.05	**-7.1739×10^{-2}**	**-13.8181×10^{-2}**	**-23.1103×10^{-2}**
0.06	**-7.0982×10^{-2}**	**-13.6729×10^{-2}**	**-22.8668×10^{-2}**
0.07	**-7.0228×10^{-2}**	**-13.5273×10^{-2}**	**-22.6237×10^{-2}**
0.08	**-6.9471×10^{-2}**	**-13.3817×10^{-2}**	**-22.3804×10^{-2}**
0.09	-6.8718×10^{-2}	- ND	- ND
0.10	-6.7961×10^{-2}	- ND	- ND

* ND = not determined.

Table 9 The changes in free energy of mixing (ΔG^m) of *A. seyal, A. senegal* and *A. polyacantha* with water at different weight fraction

Concentration (g cm^{-3})	ΔG^m(joule g^{-1})		
	A. seyal	*A. senegal*	*A. Polyacantha*
0.03	*ND	-0.50747×10^{-2}	-0.80897×10^{-2}
0.04	ND	-0.69572×10^{-2}	-1.0983×10^{-2}
0.05	**-0.475×10^{-2}**	**-0.90911×10^{-2}**	**-1.4220×10^{-2}**
0.06	**-0.588×10^{-2}**	**-1.1518×10^{-2}**	**-1.7393×10^{-2}**
0.07	**-0.714×10^{-2}**	**-1.3617×10^{-2}**	**-2.0980×10^{-2}**
0.08	**-0.833×10^{-2}**	**-1.6615×10^{-2}**	**-2.4786×10^{-2}**
0.09	-1.01×10^{-2}	ND	ND
0.10	-1.15×10^{-2}	ND	ND

* ND = not determined.

Table 10 The gum– water interaction parameters χ for *A. seyal, A. senegal* and *A. polyacantha*

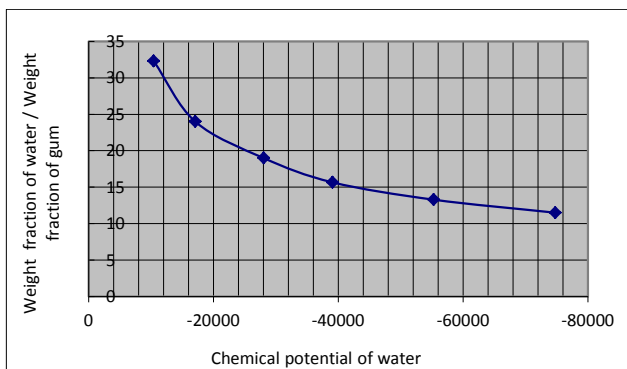| Gum sample	Parameter χ_1
A. seyal	0.4961
A. senegal	0.4928
A. polyacantha	0.4913

Fig. 12 The change in chemical potential of a *A. polacantha* gum ($\Delta\mu_2^{/}$)
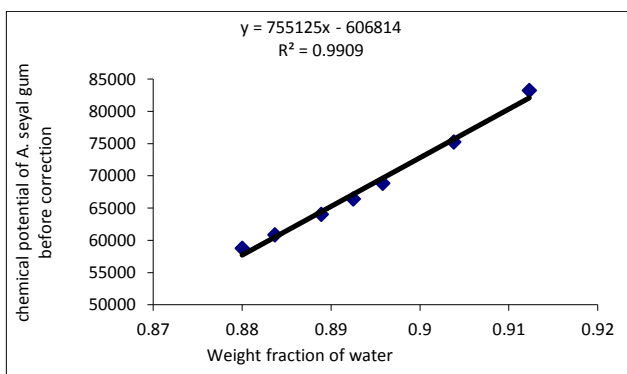

Fig. 13 The segment A to correct the chemical potential of *A. seyal* gum ($\Delta\mu_2$)
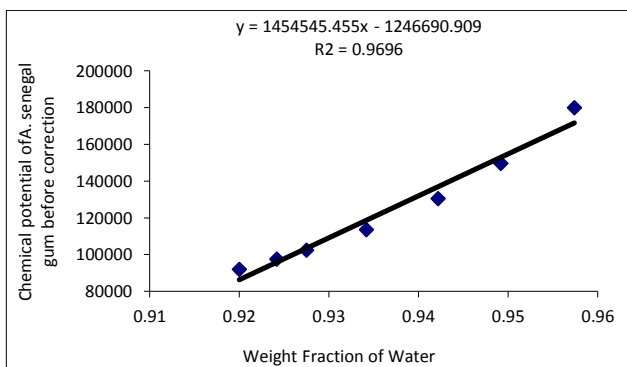

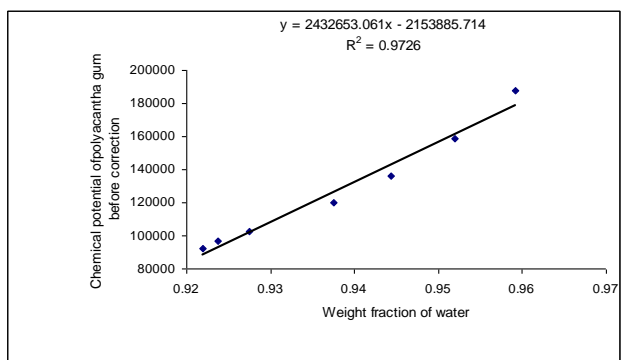**Fig. 14** The segment A to correct the chemical potential of *A. senegal* gum ($\Delta\mu_2$)

Fig. 15 The segment A to correct the chemical potential of *A. polyacantha* ($\Delta\mu_2$) of *A. polyacantha* gum solutions

Conclusion and recommendation

Comparative studies were carried out to measure and calculate the thermodynamic properties such as the density of solid gum, the partial specific volume, the volume fraction, the osmotic pressure, the chemical potential, the second virial coefficient, the free energy of mixing and the gum–solvent interaction parameter χ of the three gum solutions. The results show that water is a good solvent for the three types of gums although *A. polyacantha* interacts with water more strongly than *A. senegal*, and this interacts more than *A. seyal*. Thermodynamically, it was found that water has good affinity for the three types of gum samples. The results also show that *A. seyal* molecules are compact compared with *A. senegal* and *A. polyacantha*, and A. *senegal* molecule expands in water more than *A. seyal* molecule. Blending studies of those gum solutions depending on the thermodynamic properties, physicochemical properties, and rheological properties may lead to proper blending and wide application for the blend gums.

Acknowledgments

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