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Potravinarstvo Slovak Journal of Food Sciences

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Slovak Journal of Food Sciences

Potravinarstvo Slovak Journal of Food Sciences
vol. 11, 2017, no. 1, p. 203-209
doi: https://dx.doi.org/10.5219/735
Received: 10 February 2017. Accepted: 15 March 2017.

ISSN 1337-0960 (online)

STUDY OF RHEOLOGY AND FRICTION FACTOR OF NATURAL FOOD HYDROCOLLOID GELS

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ABSTRACT

Differences in the rheology and friction factor of natural food hydrocolloid gels were studied in this paper. The practical importance of the knowledge of the rheological parameters is quite evident. The experimental data were carried out using a concentric cylinder rotary viscometer. It was prepared 1% hydrocolloid solutions (hydrogels). Hydrogels of the natural gums extracted from the seeds of the plants and plant tubers have been used – carob gum (from the seeds of *Ceratonia siliqua*), guar gum (from the seeds of *Cyamopsis tetragonoloba*) and tara gum (from the seeds of *Caesalpinia spinosa*). Rheological behaviour has non–Newtonian pseudoplastic character and the flow curves were fitted using the Otswald – de Waele (power law) model and Herschel – Bulkley model. The hydrogels exhibit shear thinning behaviour. The meaning of the rheological parameters on the friction factors during flow of hydrocolloid gels in the tube has been shown. Information on time dependent behaviour of tested liquids has been also obtained. Time dependent curves were fitted by the Gaussian model. Preliminary results obtained for a constant shear rate showed the thixotropic and time–dependent behaviour of the hydrogels. These parameters can be especially used in much software dealing with a numerical simulation of the flow problems.

Keywords: viscosity; shear thinning; non-Newtonian fluid; velocity profile; Reynolds number

INTRODUCTION

The knowledge of the rheological properties of food products is essential for the product development, quality control, sensory evaluation and design and evaluation of the process equipment. The flow behaviour of a fluid can be varied from Newtonian to time dependent non-Newtonian in nature depending on its origin, composition and structure behaviour and previous history **(Rao, 2005)**. The knowledge of this behaviour is also very important for natural hydrocolloids owing to an increasing demand on the processed hydrocolloids products. These products can be classified as the refrigerated liquid and especially as the dried products. Hydrogels are polymer networks formed from polymers that absorb water to a significant extent **(Wientjes et al., 2000; Mandala et al., 2004; Abd Alla et al., 2012)**. Their porous structure and elastic properties make them useful for applications such as tissue engineering, drug delivery systems and functional coatings **(Kono et al., 2014; Jamshidian et al., 2014)**. Hydrogels can be formed by proteins, peptides, or other biopolymers such as alginates, or chitosan **(Sittikijyothin et al., 2007, Amundarain et al., 2009)**. In recent years, the hydrocolloids consumption in the form of food products has increased **(Hayakawa et al., 2014; Tárrega et al., 2014)**. Several researchers **(Alves et al., 1999; Sandolo et al., 2009; Karaman et al. 2014)** studied the rheological characteristics of natural hydrocolloids and reported Newtonian as well as time–dependent non–Newtonian flow behaviour of hydrogels (hydrocolloids solutions) **(Sandolo et al., 2010)**.

A non-Newtonian fluid is a fluid with properties that are different in any way from those of Newtonian fluids. Most commonly, the viscosity (the measure of a fluid's ability to resist gradual deformation by shear or tensile stresses) of non-Newtonian fluids is dependent on shear rate or shear rate history. Some non-Newtonian fluids with shear-independent viscosity, however, still exhibit normal stress-differences or other non-Newtonian behaviour **(Bourriot et al., 1999)**. In a Newtonian fluid, the relation between the shear stress and the shear rate is linear, passing through the origin, the constant of proportionality being the coefficient of viscosity. In a non-Newtonian fluid, the relation between the shear stress and the shear rate is different and can even be time-dependent (Time Dependent Viscosity). Therefore, a constant coefficient of viscosity cannot be defined **(Kumbár et al., 2015a)**. There are underlying differences in flow behaviour which can cause problems in transport, processing, manufacturing, and/or storage.

Rheological properties are depended of concentration of hydrogels. With increasing concentration of hydrocolloid the solution (gel) exhibits non-Newtonian behaviour **(Zhu et al., 2012; Torres et al., 2013)**.

Considering this lack of published information on fluid dynamics of the natural hydrogels, the main purpose of this work was to determine rheological behaviour of these products for three hydrogels of the natural gums extracted from the plants and plant tubers. The carob gum is extracted from the seeds of *Ceratonia siliqua*, the guar gum is extracted from the seeds of *Cyamopsis tetragonoloba*, and the tara gum is extracted from the seeds of *Caesalpinia spinose* **(Wientjes et al., 2000; Sittikijyothin et al., 2007; Sandolo et al., 2010)**. In the Europe Union are these natural hydrocolloids labelled. The carob gum is labelled as E410, guar gum as E412 and the tara gum as E417.

The meaning of the reological data for the calculation of friction factors for the tube flow is discussed in details. Information on time dependent behaviour of the tested solutions has been obtained.

MATERIAL AND METHODOLOGY

Natural hydrocolloids carob gum, guar gum and tara gum were purchased from specialized manufacturer.

The carob gum (also know as locust bean gum) is a galactomannan vegetable gum extracted from the seeds of the carob tree, mostly found in the Mediterranean region. The long pods that grow on the tree are used to make this gum. The pods are kibbled to separate the seed from the pulp. The seeds have their skins removed by an acid treatment. The deskinned seed is then split and gently milled. This causes the brittle germ to break up while not affecting the more robust endosperm. The two are separated by sieving. The separated endosperm can then be milled by a roller operation to produce the final locust bean gum powder **(Mandala et al., 2004)**.

The guar gum, also called guaran, is a galactomannan. It is primarily the ground endosperm of guar beans. The guar seeds are dehusked, milled and screened to obtain the guar gum. It is typically produced as a free-flowing, off-white powder **(Sandolo et al., 2009)**.

The tara gum is a white or beige, nearly odorless powder that is produced by separating and grinding the endosperm of C. spinosa seeds. Tara gum consists of a linear main chain of (1-4)-ß-D-mannopyranose units attached by (1-6) linkages with α-D-galactopyranose units. The major component of the gum is a galactomannan polymer similar to the main components of guar and locust bean gums that are used widely in the food industry. The ratio of mannose to galactose in the tara gum is 3:1 **(Sittikijyothin et al., 2007)**.

It was prepared 1% solutions from the dried gum and the distilled water. Exactly 500 mL of samples for each hydrogel were prepared and stored (couple of minutes) at 20 °C before measurement. From the physical parameters which have been measured only the densities of the tested liquids are presented in the Table 1.

The rheological measurements were carried out using the DV3-P viscometer (Anton Paar, Austria), equipped with a coaxial cylinder sensor system. Rotational speeds ranged between 0.3 rpm and 12 rpm, which is corresponds with shear strain rate from 0.102 s^{-1} to 4.08 s^{-1}, because standard spindle TR 9 (1 rpm = 0.34 s^{-1}) was used. Viscosity η [Pa·s] is the ratio of shear stress σ [Pa] and shear strain rate $\dot{\gamma}$ [s^{-1}] as is described in publication **(Kumbár et al., 2015b)**:

$$\eta = \frac{\sigma}{\dot{\gamma}}. \tag{1}$$

All measurements were performed at the constant temperature 20 °C.

RESULTS AND DISCUSSION

Shear stress and apparent viscosity

In the Figure 1 the flow curves, i.e. shear stress vs. shear strain rate, are shown. These curves can be fitted by using

Table 1 Density of hydrogels.

Product	Concentration	Density ρ (kgm^{-3})
carob gum	1%	1005
guar gum	1%	1022
tara gum	1%	1041

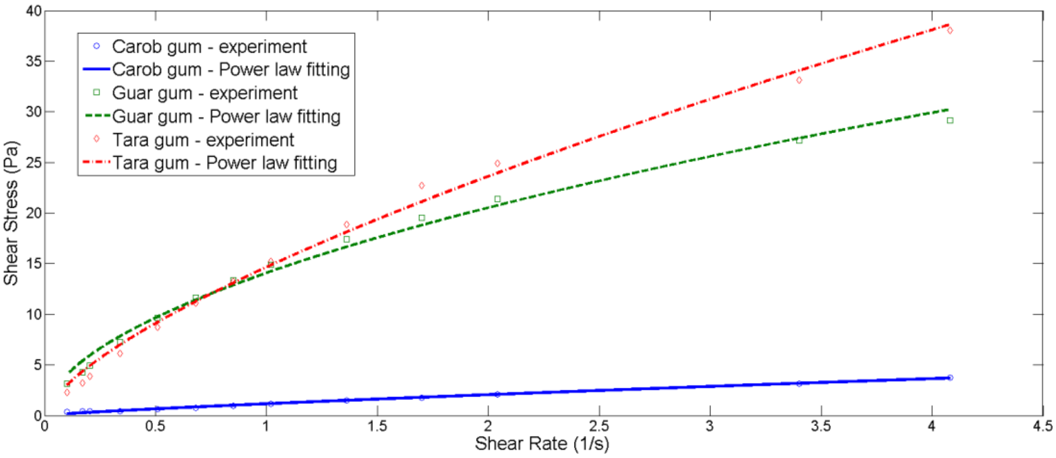

Figure 1 Effect of shear strain rate on the shear stress.

Table 2 Parameters of the Ostwald – De Waele model for hydrogels (R^2 is the coefficient of determination).

Hydrogel	K (Pas^n)	n (–)	R^2
carob gum	1.158	0.8253	0.9934
guar gum	14.07	0.5441	0.9919
tara gum	14.65	0.6887	0.9943

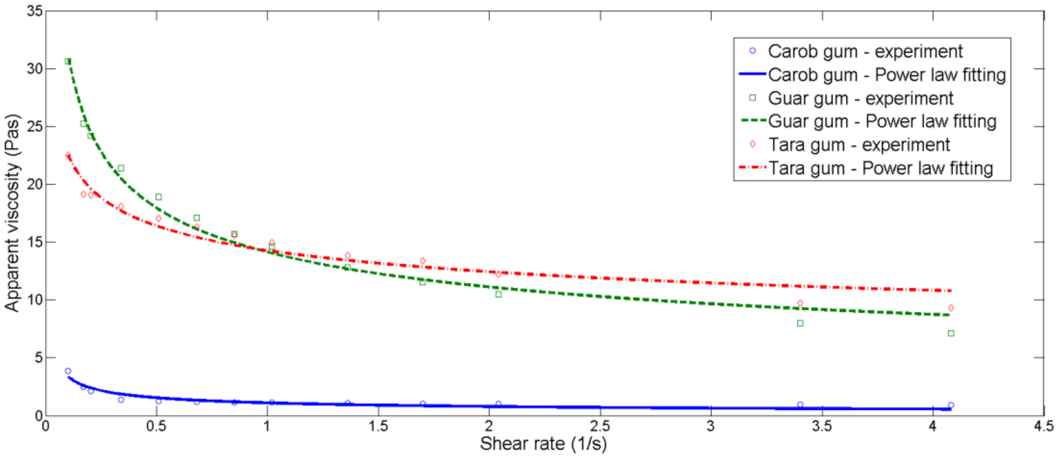

Figure 2 Apparent viscosity of the hydrogels.

of Herschel-Bulkley model **(Saravacos and Kostaropoulos, 1995)**:

$$\sigma = \sigma_0 + K\dot{\gamma}^n. \qquad (2)$$

In Eq. (2), K is the consistency index [Pa·s^n], n is the flow behaviour index [–] and σ_o is the yield stress [Pa].

For all three hydrogels this model reduces to the Ostwald – De Waele model, given by Eq. (3), also known as the power-law model **(Rao, 1982)**:

$$\sigma = K\dot{\gamma}^n. \qquad (3)$$

Eqs. (2) and (3) can be used for both Newtonian and power law fluids, since for Newtonian fluids n equals 1, and K equals η and /or ($\eta + \sigma_o$), respectively.

The Eq. (3) can be used for all three hydrogels. Parameters of Eq. (3) are given in the Table 2.

The apparent viscosity is than given by the Eq. (1). The apparent viscosity of tested liquids is shown in the Figure 2.

Time dependence of the apparent viscosity

In order to study of the time on the hydrogels these liquids were sheared at constant shear rates (3.4 s^{-1}) for about 4000 and 5000 s and changes of apparent viscosity with time was considered as time dependence.

Results of time–dependences of the apparent viscosity for the hydrogels are shown in the Figures 3 – 5.

It is obvious that be apparent viscosity decreases with the time. The experimental data were fitted by Gaussian model with different coefficient k.

$$\eta = \sum_{k=1}^{k=4} a_k \exp\left[-\left(\frac{t-b_k}{c_k}\right)^2\right] \text{ (carob gum solution)}, \quad (4)$$

$$\eta = \sum_{k=1}^{k=2} a_k \exp\left[-\left(\frac{t-b_k}{c_k}\right)^2\right] \text{ (guar gum and tara gum solution)}. \quad (5)$$

Parameters of the Eq. (4) for Carob gum solution are given in the Table 3. Parameters of the Eq. (5) for Guar and Tara gum solution are given in the Table 4.

Friction factor and flow velocity

The hydrogels exhibit shear thinning behaviour. There are occurred differences between solutions of different gums. The highest values of the apparent viscosity were achieved for the guar gum, following by the tara gum and the minimum values exhibited carob gum.

The obtained rheological parameters have great meaning in many problems of industry. For example the design of piping and pumping systems requires knowledge of the pressure drop due to flow in straight pipe segments and through valves and fittings. Friction losses caused by the presence of valves and fittings usually result from disturbances of the flow, which is forced to change direction abruptly to overcome path obstructions and to adapt itself to sudden or gradual changes in the cross section or shape of the duct. This problem is described e.g. in **(Cabral et al., 2011)**. The pressure drop is calculated using of the friction factor, f. The friction factor is defined as **(Garcia and Steffe, 1987)**:

$$f = \frac{2\sigma_w}{\rho v^2}, \qquad (6)$$

where ρ is the fluid density, v is the average flow velocity, and σ_w is the stress in the wall, given by

$$\sigma_w = \frac{D\Delta P}{4L}. \qquad (7)$$

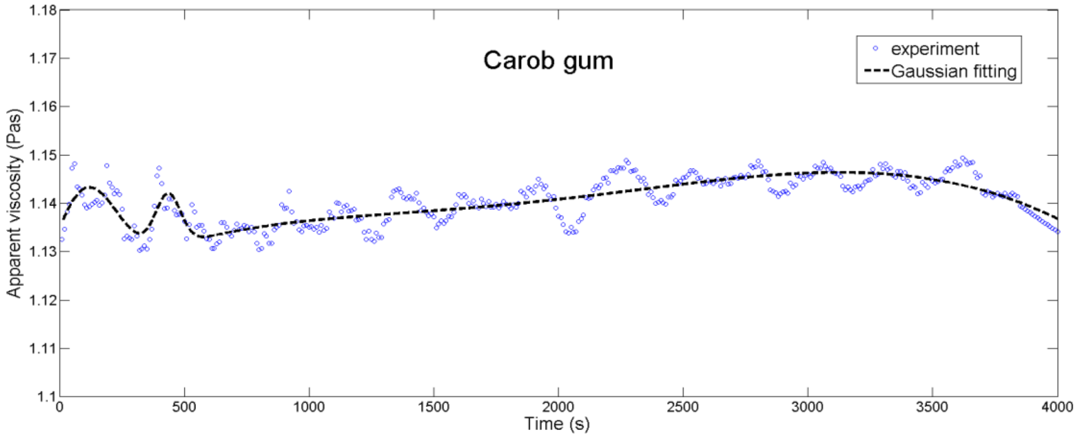

Figure 3 Time-dependence of the apparent viscosity (carob gum solution).

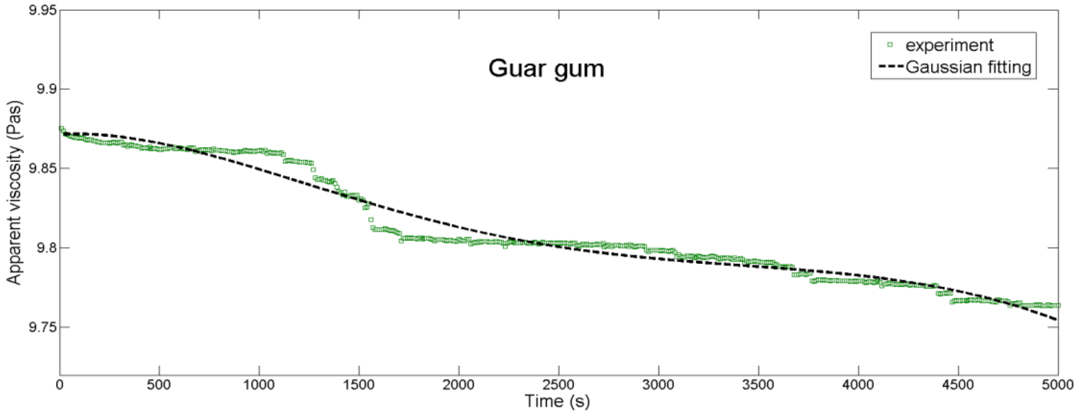

Figure 4 Time-dependence of the apparent viscosity (guar gum solution).

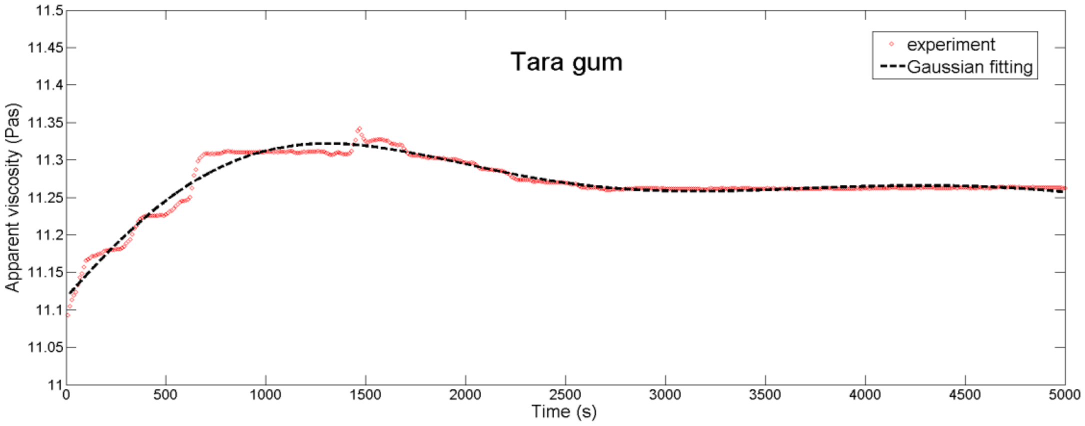

Figure 5 Time-dependence of the apparent viscosity (tara gum solution).

In Eq. (7) D is the tube diameter and ΔP is the pressure drop observed in a length L of the tube. For laminar flow, the friction factor can be obtained from a simple function of the generalized Reynolds number, which is identical to the dimensionless form of the Hagen-Poiseuille equation **(Darby, 1996)**:

$$f = \frac{16}{Re}, \tag{8}$$

in which

$$Re = \frac{D^n v^{2-n} \rho}{8^{n-1} K} \left(\frac{4n}{1+3n}\right)^n. \tag{9}$$

Eqs. (8) and (9) can be used for both Newtonian and power law fluids, since for Newtonian fluids n equals 1, and K equals η, so that the generalized Reynolds number (Eq. (7)) reduces to well–known number:

$$Re = \frac{Dv\rho}{\eta}. \tag{10}$$

The values of generalized Reynolds number for hydrogels tested in this paper are given in the Table 5. These numbers have been calculated for values of D = 0.1 m and v = 1 m.s^{-1}.

Under turbulent flow conditions, the existing correlations to estimate the friction factor are semi-empirical. For

Table 3 Parameters of fitting for Eq. (4) (*SSE* is sum squared error, *RMSE* is root–mean–square error and R^2 is the coefficient of determination).

carob gum	a_1 **(Pas)**	b_1 **(s^{-1})**	c_1 **(s^{-1})**	a_2 **(Pas)**	b_2 **(s^{-1})**
	10.47	4430	1.553E4	0.02026	96
	c_2 **(s^{-1})**	a_3 **(Pas)**	b_3 **(s^{-1})**	c_3 **(s^{-1})**	a_4 **(Pas)**
	180.4	0.01093	431.4	69.48	1.462
	b_4 **(s^{-1})**	c_4 **(s^{-1})**	***SSE***	***RMSE***	R^2
	-2863	3949	0.00283	0.002702	0.6809

power law fluids, probably the best-known correlation is that presented by **(Dodge and Metzner, 1959)**:

$$\frac{1}{\sqrt{f}} = \frac{4}{n^{0.75}}\ln\left(Ref^{1-\frac{n}{2}}\right) - \frac{0.4}{n^{1.2}}. \tag{11}$$

Let us consider a cylindrical tube of diameter $D = 0.1$ m and an average flow velocity $v = 1$ m.s^{-1}. The values of generalized Reynolds numbers (Eq. 9) are given in the Table 5.

The Reynolds number describes namely the transitive from laminar to turbulent flow. The behaviour of hydrogels is different. The maximum value exhibits the carob gum solution. Laminar flow of a power law fluid exists in the tube **(Kumbár et al., 2015c)** when:

$$Re \leq (Re)_{critical} \tag{12}$$

The critical value of the power law Reynolds number depends on the value of the flow index behaviour n according to **(Steffe and Daubert, 2006)**:

$$(Re)_{critical} = 2100 + 875(1-n) \tag{13}$$

Values of critical Reynolds number vary from 2888 at $n =$ 0.1 to the familiar value 2100 for Newton liquids ($n = 1$). In all cases the flow is laminar.

The next application of the rheological properties is connected with the continuous thermal processing system. Such system generally involves a heat exchanger in form of a tube. A length of this tube is known as a “hold tube“, must be sufficient in order to achieve sufficient fluid residence time. Because the hold tube is a critical part of the system understanding velocity profiles found in tube flow is important for the numerical simulation of thermal process.

For power law fluid in laminar flow the velocity $v(x)$ is function of the distance x from the centre of the pipe:

Table 4 Parameters of fitting for Eq. (5) (*SSE* is sum squared error, *RMSE* is root–mean–square error and R^2 is the coefficient of determination).

guar gum	a_1 **(Pas)**	b_1 **(s^{-1})**	c_1 **(s^{-1})**	a_2 **(Pas)**	b_2 **(s^{-1})**
	2.139	-3.661	2.948	9.68	1.853
	c_2	***SSE***	***RMSE***	R^2	
	9.844	0.0196	0.00629	0.9667	
tara gum	a_1 **(Pas)**	b_1 **(s^{-1})**	c_1 **(s^{-1})**	a_2 **(Pas)**	b_2 **(s^{-1})**
	0.2941	691.9	1579	11.26	4403
	c_2 **(s^{-1})**	***SSE***	***RMSE***	R^2	
	2.34E4	0.0289	0.00765	0.9587	

Table 5 Reynolds numbers given by the Eq. (9).

Hydrogel	***Re***
carob gum	179
guar gum	48
tara gum	26

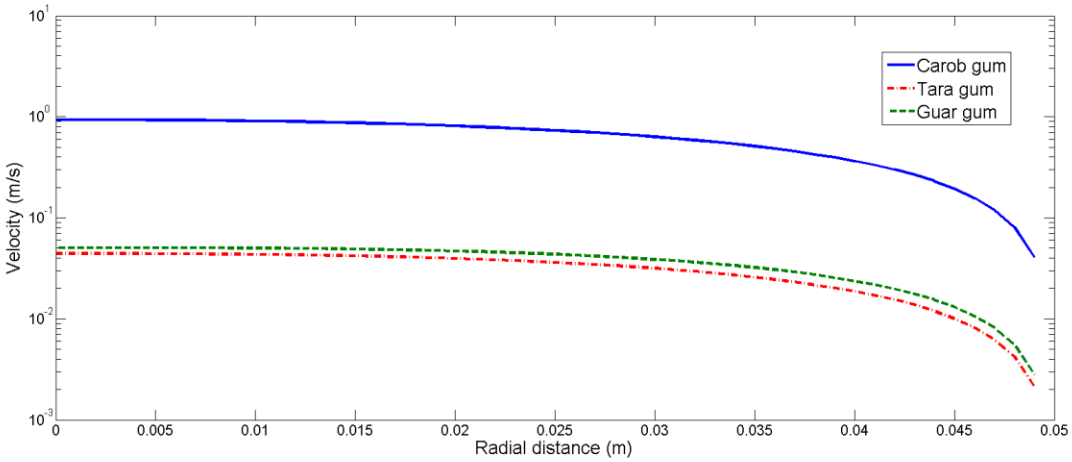

Figure 6 Velocity profiles of hydrogels flow.

$$v(x) = \left(\frac{\Delta P}{2KL}\right)^{\frac{1}{n}} \left(\frac{n}{n+1}\right) \left(R^{\frac{n+1}{n}} - x^{\frac{n+1}{n}}\right), \qquad (14)$$

where ΔP denotes the driving over pressure, L is the tube length and R is its radius. For the illustration the values of R = 0.05 m, L = 1 m and ΔP = 1000 Pa were chosen. Results are displayed in the Figure 6.

The highest values of flow velocities are observed for the flow of carob gum solution. The lowest one was achieved for the flow of the tara gum solution. Velocities are different for different hydrogels but velocity of the tara gum and guar gum solutions are very close.

The velocity equation given above is valid for fully developer undisturbed flow in straight, horizontal tubes. Reals processing systems contain many elements like valves, tees, elbows, etc. that cause fluid mixing during flow **(Das et al., 1991; Telis-Romero et al., 2005)**.

In addition pipe vibration caused by energy inputs from pumps may contribute to mixing. It means that the equation given above represents only general guidelines in examining velocity profiles during tube flow.

CONCLUSION

Rheological properties of natural hydrocolloids solutions (carob gum – from the seeds of *Ceratonia siliqua*, guar gum – from the seeds of *Cyamopsis tetragonoloba* and the tara gum – from the seeds of *Caesalpinia spinosa*) were studied using rotary viscometer with coaxial cylinder sensor system. Prepared were 1% solutions of hydrocolloids (hydrogels). Experimental data were successfully fitted with the Ostwald-De Waele model and Herschel-Bulkeley model. The hydrogels exhibit shear thinning behaviour.

The highest values of the apparent viscosity were achieved for the guar gum solution, following by the tara gum solution and the minimum values exhibited carob gum solution.

The differences between the guar gum and tara gum solutions are not too significant. In order to study of the time on the hydrogels these liquids were sheared at constant shear strain rates for about 4000 and 5000 s and changes of apparent viscosity with the time was considered as time dependence. Preliminary results obtained for a constant shear strain rate showed the thixotropic and time–dependent behaviour of the hydrogels. The behaviour of the hydrogels was complicated – Gaussian model it was used. The practical importance of the knowledge of the rheological parameters was outlined. These parameters can be used in much software dealing with a numerical simulation of the flow problems.

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Acknowledgments:

This work was supported by TP 2/2017 “Effect of addtives on the rheological behaviour of foodstuffs and product and raw materials for their production” financed by IGA AF MENDELU.

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