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Measurement, prediction and correlation of density, viscosity, surface tension and ultrasonic velocity of different honey types at different temperatures

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ABSTRACT

The aim of this study is to investigate the influence of temperature (20, 25, 30, 35, 40, 45 and 50 °C) and concentration on the evolution of different physical properties (density, viscosity, ultrasonic velocity and surface tension) of honeys. Temperature influences negatively all the physical properties of honey. A good correlation between density, viscosity, ultrasonic velocity and surface tension using linear and polynomial equations was observed. Also a good correlation between density, ultrasonic velocity and surface tension for all the honey samples ($R^2 = 0.954$) was obtained. The correlation between surface tension and viscosity was made using the Pelofsky model and an exponential model; the latter one describes better the correlation between surface tension and viscosity.

1. Introduction

Honey is the natural substance produced by honeybees, *Apis mellifera*, in almost every country of the world, from nectar of plants, as well as from honeydew (Blasa et al., 2006). The latter one is a sugar-containing substance excreted by some plant-sucking insects. Chemically, honey comprises sugars (70–80%), water (10–20%) and other minor constituents such as organic acids, mineral salts, vitamins, proteins, phenolic compounds and free amino acids. Monosaccharides, fructose and glucose are the main sugars found in honey (Nagai et al., 2002; Terrab et al., 2001; Ouchemoukh et al., 2007). The quality of honey is mainly determined by its sensorial, chemical, physical and microbiological characteristics. Honey physicochemical quality criteria are well specified by the EC Directive 2001/110 (EU, 2001). The major criteria of interest are moisture content, electrical conductivity, ash content, reducing and non-reducing sugars, free acidity, diastase activity and hydroxymethylfurfural (HMF) content (Gomes et al., 2010).

The rheological properties of honey are important qualities that influence the sensory quality of the product and also affect a number of technological operations, such as honey heating, mixing, filtering, hydraulic transport and bottling (Yanniotis et al., 2006).

The same as surface tension and viscosity, ultrasonic velocity is also an important parameter for materials' characterization. Ultrasonic velocity is usually correlated with density fluctuations of pure liquid; thereby it is closely related to compressibility, which is a response function of the density to the mechanical pressure. This is extremely sensitive to molecular organization and molecular interaction, hence gives accurate information about the processes occurring in the time scale 10^{-3}–10^{-10} s. Ultrasonic velocity can be used with other properties by thermodynamic relationship, resulting many important thermophysical properties viz. attenuation, relaxation time, isentropic compressibility, specific acoustic impedance, thermal expansion coefficient, etc. (Singh and Singh, 2011).

In the last decades some physical properties of honey have been investigated by other scientists: rheological properties (Yanniotis et al., 2006; Mossel et al., 2000; Kang and Yoo, 2008; Oroian et al., 2012; Oroian et al., 2013); but to the author's knowledge no other studies regarding the influence of temperature on density, viscosity, surface tension and ultrasonic velocity of honey samples have been reported in the literature yet.

The aim of this study is to evaluate the influence of the temperature honey density, viscosity, surface tension and ultrasonic velocity and to evaluate the correlation between these parameters.

2. Materials and methods

2.1. Materials

The honey samples (7 types of honey with different concentration and source type) are from Suceava county, the second county

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as surface from Romania; all the samples were harvested in 2012. The rheological and physical properties of honey can be influenced by the presence of crystals and air bubbles (Bhandari et al., 1999; Mossel et al., 2000; Abu-Jdayil et al., 2002). Before being used they were warmed up to 55 °C for 48 h into a temperature-controlled oven (Memmert, Germany) to dissolve any crystals, and kept in flasks at 30 °C to remove air bubbles that could interfere in rheological studies.

2.2. Physicochemical parameters measurements

The moisture contents of honey samples were obtained by measuring the refractive index at 20 °C using a digital refractometer (Leica Mark II Plus). The water content and °Brix concentration were determined based on a Chataway table (Bogdanov, 2002).

The determination of glucose and fructose in honey samples was made by a HPLC 10ADVP SHIMADZU, with RI-detector, according to a method described by Bogdanov (2002). The compounds were separated on an Alltech type Alltima amino column, 250 × 4.6 mm i.d. and particle size 5 μm. The samples were prepared as: 5 g of honey were dissolved in water (40 ml) and transferred quantitatively into a 100 ml volumetric flask, containing 25 ml methanol and filled up to the volume with water. The solution was filtered through a 0.45 μm membrane filter and collect in sample vials. Flow rate 1.3 ml/min, mobile phase: acetonitrile/water (80:20, v/v), column and detector temperature 30 °C, sample volume 10 μl. A calibration curve was made for each sugar using standard solutions of different concentrations (0.5–80 mg/ml). The linear regression factor of the calibration curves was higher than 0.9982 for all sugars. Sugars were quantified by comparison of the peak area obtained with those of standard sugars. The results for each sugar were expressed as g/100 g honey.

2.3. Density measurements

Density (ρ) of the honey samples was measured using pycnometer with an accuracy 10^{-5} gm/cc. The calibration of pycnometer was made with ultrapure water. Temperature was kept constant within ±0.1 °C using PID controller and circulating water using thermostatic-fluid bath. The density of the seven samples of honeys was measured at 20 °C, 25 °C, 30 °C, 35 °C, 40 °C, 45 °C and 50 °C. The values of parameters were expressed as the mean ± standard deviation to a confidence interval for mean of 95%.

2.4. Ultrasonic velocity measurement

The ultrasonic speed measurement was carried out using a flow detector USM 35 X (GE Measurement and Control, USA) with a dual-element (TR) probe working at 4 MHz. The measurements were carried out at 20 °C, 25 °C, 30 °C, 35 °C, 40 °C, 45 °C and 50 °C. The values of parameters were expressed as the mean ± standard deviation to a confidence interval for mean of 95%.

2.5. Viscosity measurement

Viscosity measurements were carried out on the honey samples at different temperatures (20, 25, 30, 35, 40, 45 and 50 °C), with Brookfield RV-DV II Pro Viscometer with RV spindles, RV 6 spindle, with temperature controlled water bath. The honey sample was allowed to reach the desired temperature 20 min. Each measurement was taken in duplicate. The values of parameters were expressed as the mean ± standard deviation to a confidence interval for mean of 95%.

2.6. Surface tension determination

The surface tension was computed using the Auerbach́s equation (Auerbach, 1948):

$$u = \left(\frac{\sigma \times 10^{10}}{6.33 \times \rho}\right)^{2/3} \tag{1}$$

where u is the ultrasonic velocity (m/s), σ is the surface tension in N/m and ρ is the density in kg/m^3. Therefore, for the calculation of the surface tension, first we measured the ultrasonic velocity and density and later on by using the Auerbach equation we were able to compute the surface tension.

2.7. Prediction accuracy

The mean relative deviation modulus, D, was used to verify the suitability of model for experimental data:

$$D\% = \frac{100}{n}\sum_{i=1}^{n}\frac{|x_{\exp,i} - x_{cal,i}|}{x_{\exp,i}} \tag{2}$$

where n is the total number of data in the number of data in the sample. Subscript exp and cal denote experimental and calculated values, respectively. X represents density, ρ, or viscosity, η, surface tension, σ, and ultrasonic velocity, u.

2.8. Statistical analysis

The parameters were fitted to the proposed equation using SPSS 16.00 and Excel 2007.

3. Results and discussion

Table 1 presents some physicochemical parameters (moisture content, °Brix concentration, fructose, glucose, fructose/glucose ratio) of the seven honeys analyzed. The moisture content ranged between and met the threshold requirements (20% maximum) for honey moisture content as defined by the Codex Alimentarius (Codex Standard, 2001). In all the cases of honeys analyzed, the total content of glucose and fructose was over 60 g/100 g honey, in agreement with 2001/110/CE Directive. The ratio fructose/glucose was always higher than 1, which confirmed the crystallization state of the analyzed honeys (all the honeys were in liquid state) (Oroian, 2012).

3.1. Effect of temperature on density

The measured densities of honey samples from 20 to 50 °C in function of temperature and concentration, c_H (expressed as °Brix concentration), are presented in Figs. 1 and 2 respectively. The density of honey samples was higher than water density in all the samples; it ranged (at 20 °C) between 1386.055 and 1403.222 kg/m^3, being strongly influenced by concentration (r = 0.914). The density is slightly influenced by temperature; the values of the honey density decreased with the increasing of temperature.

It can be observed that the density is linear with regard to temperature or concentration of the honey samples. The densities of the honey samples decreased linearly as the temperature increased, and increased linearly with an increase of the concentration of honey samples.

The evolution of density with temperature shows a small change of the density with the change of the temperature under a constant concentration of the solution. The evolution of the den-