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Rheological behavior of high altitude Indian honey varieties as affected by temperature

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Abstract The rheological properties of high altitude India honeys were analyzed in honeydew (pine) and nectar honey (multifloral and acacia) varieties at a wide range of temperatures (0, 5, 10, 20, and 30 °C). All the honey samples were significantly dominated by loss modulus (G'') which displayed their viscous nature. Irrespective of geographical origin and temperature, all the honey varieties showed a Newtonian behavior. The viscosity of all honey varieties showed a strong dependence on temperature, and Arrhenius model was examined to describe this.

1. Introduction

According to Codex Alimentarius (2001), honey is the natural sweet substance produced by honeybees from the nectar of plants or from secretions of living parts of plants or excretions of plant sucking insects on the living parts of plants, which the bees collect, transform by combining with specific substances of their own, deposit, dehydrate, store and leave in the honeycomb to ripen and mature. Honey is considered as oldest sweetening substance mainly dominated by 70% of sugars primarily glucose and fructose (Nayik et al., 2014; Smanalieva and Senge, 2009; Nayik et al., 2016a). It is rich source of amino acids, organic acids, some macro- and microelements and polyphenols (Nayik and Nanda, 2016a). The polyphenols (phenolic acids and flavonoid compounds) are primarily responsible for antioxidant activity of honey (Nayik and Nanda, 2015a; Nayik and Nanda, 2016b; Suhag et al., 2016; Nayik et al., 2016b). Viscosity is one of the most important and significant characteristics of honey which affects its quality as well as the design of processing equipment (Anupama et al., 2003). The viscosity of honey depends on several factors, especially on temperature, moisture content and chemical constitution (Yanniotis et al., 2006; Kang and Yoo, 2008). The viscosity is considered as an important parameter in all stages of honey production, starting from the extraction of honey from combs to straining, pumping, processing and packing as well. The presence of sugars, acids, proteins and colloids makes the honey a material with changing molecular structure. Studies on rheological behaviors of honey are important for applications related to handling, storage, processing, pumping, transport, quality control, and sensory analysis of foods. The effect of temperature on rheological properties also needs to

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be analyzed since the honey experiences a wide range of temperatures during processing and storage (Rao, 1999).

In the literature, most honey varieties have showed Newtonian behavior irrespective of their geographical origin (Al-Malah et al., 2001; Junzheng and Changying, 1998; Mossel et al., 2000; Nayik and Nanda, 2015b) but few authors have reported non-Newtonian behavior of honey as in case of heather, manuka and buckwheat honeys (thixotropic), and eucalyptus honeys (dilatant) which may be due to presence of high-molecular compounds such as proteins or polysaccharides (dextran) (Smanalieva and Senge, 2009; Witczak et al., 2011; Yanniotis et al., 2006).

In past few decades, the rheological properties of honey from different countries have been briefly analyzed: Australia (Mossel et al., 2000), China (Chen et al., 2009), Korea (Kang and Yoo, 2008), Greece (Yanniotis et al., 2006), India (Kumar and Mandal, 2009; Nayik et al., 2015; Saxena et al., 2014), Spain (Gomez-Diaz et al., 2009), Jordan (Abu-Jdayil et al., 2002), and Poland (Witczak et al., 2011).

Although we have studied the rheology of few varieties of honey from fruit sources belonging to Kashmir valley of India (Nayik et al., 2015), still few varieties from non-fruit sources are yet to be studied. Thus, the main aim of the study was to determine rheological behavior of three high altitude Indian honey varieties from Kashmir valley (Altitude: 1850 m ASL) of India as a function of temperature.

2. Material and methods

2.1. Honey sample collection and pollen analysis

Three different raw and fresh honey varieties ($n = 24$) of acacia honey (*Robinia pseudoacacia*), pine honeydew (*Pinus wallichiana*) and multifloral honey samples were collected directly from beekeepers from September 2012 to May 2014 from different areas (Pulwama, Budgam and Srinagar) of Kashmir valley. All the honey samples were packed and stored at 4 °C prior to analysis. The origins of each honey sample were confirmed by melissopalynology. Honey samples were classified according to their botanical origin using the method described by Von der Ohe et al. (2004). The following terms were used for frequency classes: predominant pollen (>45% of pollen grains counted), secondary pollen (16–45%), important minor pollen (3–15%) and minor pollen (<3%).

Presence of the air bubbles and sugar crystals in honey can influence the rheological properties of honey (Abu-Jdayil et al., 2002); thus, the honey samples were heated to 50 °C for 1 h in a water bath to dissolve crystals, and then kept in 30 °C to remove air bubbles.

2.2. Rheological measurement

A Modular Compact Rheometer (MCR-102, M/s. Anton Paar, Austria), equipped with parallel plate system (50 mm diameter) at a gap of 0.5 mm at different temperatures (0, 5, 10, 20, and 30 °C) was used to study the dynamic rheological characteristics of honey samples. A strain sweep test was performed in the range of 0.01% and 50% in order to establish % strain since viscoelastic material is independent of strain up to a critical strain level. In this case, the critical strain obtained was at 3%.

After this, the frequency sweep test was employed over the range of 0.63–63 rad/s at 3% strain in order to determine the rheological data. A Rheoplus data analysis software (32 V3.40.) was used to obtain the experimental data and to calculate storage modulus (G') and loss modulus (G''). Shear stress/shear rate were evaluated in order to measure the viscosity. Results were obtained as an average of two measurements.

3. Results and discussion

3.1. Pollen analysis

The pollen spectra of honey samples studied have been briefly described and the percentages are related to pollen of nectar producing plants. Acacia honey contained 54–60% pollen of *R. pseudoacacia* sp. The honeydew element/pollen grain (HDE/P) ratio was in a range of 2.79–3.01 in pine honeydew (*P. wallichiana*), which was in good agreement with Louveaux et al. (1978). The microscopic analysis revealed some fungal spores in pine honeydew which is in good agreement with those found in Greek pine honeys. Multifloral honey contained 2–5% pollens of *Plectranthus rugosus*, and other pollens found were from *Prunus* sp., *Brassica* sp., *Thyme* sp. and *Ailanthus* sp.

3.2. Rheological measurement

Frequency sweep test was carried out to determine the rheological parameters (G' and G'') and their dependence on angular frequency. The loss modulus (G'') magnitude was much greater than storage modulus (G') which displayed that all three honey varieties showed more viscous behavior than elastic i.e. $G'' \gg G'$. Similar rheological behavior for different types of honey has been obtained by different authors around the world (Gomez-Diaz et al., 2009; Juszczak and Fortuna, 2006; Kumar and Mandal, 2009; Yanniotis et al., 2006; Yoo, 2004).

With increase in angular frequency, both G' and G'' increased which is in agreement with Yoo (2004). Fig. 1a–c shows the loss modulus (G'') as a function of angular frequency (ω) for three honey varieties at different temperatures (0–30 °C). The values for the viscous component i.e. G'' varied between 0.19 Pa at 30 °C in acacia honey and 1085.49 Pa at 0 °C in multifloral honey while elastic component ranged between 0.01 Pa at 30 °C in acacia honey and 15.3 Pa at 0 °C in multifloral honey. The reason behind the differences in G'' and G' between three honey varieties could be due to variations in pollen spectra, sugar composition and moisture content (Lazaridou et al., 2004). Doublier and Cuvelier (1996) concluded that G' is less important than G'' due to its low value. Thus in our present study, the effect of temperature on dynamic rheological properties on honeys was characterized by G'' values only. To describe the variation in the rheological properties of all honey varieties, the data obtained for both parameters (G'' and G') at different temperatures (0–30 °C) were fitted to the well-known power law as suggested by Rao and Cooley (1992) (Eqs. (1) and (2)):

$$G'' = k''(\omega)^{n''} \quad (1)$$

$$G' = k'(\omega)^{n'} \quad (2)$$

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