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Journal of Food Engineering 72 (2006) 372–377

JOURNAL OF FOOD ENGINEERING

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Effect of moisture content on the viscosity of honey at different temperatures

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Received 9 February 2004; revised 20 July 2004; accepted 21 December 2004
Available online 17 February 2005

Abstract

Viscosity of honey was measured in two honeydew honeys (pine and fir) and four unifloral nectar honeys (thymus, orange, helianthus and cotton) at their initial moisture content as well as at 17%, 19% and 21% water content at 25, 30, 35, 40 and 45 °C. It was found that viscosity varied between 0.421 and 23.405 Pa s. Shear stress varied linearly with shear rate for all the samples indicating Newtonian behaviour. Shear stress was also measured at a constant shear rate as a function of time. Viscosity was time independent. Arrhenius equation was used to express the variation of viscosity with temperature. The activation energy and the constant μ_o of the Arrhenius equation were determined as a function of moisture content from regression analysis of the experimental results. Activation energy decreased linearly as the moisture content increased varying between 70.8 and 96.3 kJ/mol. The constant μ_o increased exponentially as the moisture content increased.

Keywords: Honey; Viscosity; Moisture content; Temperature; Arrhenius model

1. Introduction

Viscosity is one of the most significant physical and sensory characteristics of honey, which affects the quality of the product as well as the design of honey-processing equipment. The importance of this parameter in process engineering is crucial in all stages of honey production, starting from the extraction of honey from comps, straining, mixing of different honey types, pumping, processing and packing.

Viscosity of honey is influenced by temperature, moisture content, as well as the presence of crystals and colloids in the product. Various researchers have studied viscosity of honey as function of temperature at specific moisture content (Junzheng & Changying, 1998; Lazaridou, Biliaderis, Bacandritsos, & Sabatini, 2004; Mossel, Bhandari, D'Arcy, & Caffin, 2000; Sopade et al., 2003). Moisture content of honey depends on the environmental conditions and the manipulation from beekeepers at the period of harvest. It can vary from year to year. High moisture content could accelerate crystallisation in certain types of honey and increase its water activity to values where certain yeasts could grow. Anupama, Bhat, and Sapna (2003) found moisture content in 11 commercial samples to vary from 17% to 22.6%, Lazaridou et al. (2004) from 13.0% to 18.9%, Sopade et al. (2003) from 15.8% to 18.0% and Junzheng and Changying (1998) from 19.8% to 29%.

In all recent papers, honey is reported as Newtonian liquid (Abu-Jdayil, Ghzawi, Al-Malah, & Zaitoun, 2002; Bhandari, D'Arcy, & Chow, 1999; Junzheng & Changying, 1998; Lazaridou et al., 2004). However, there are some reports in the literature, as cited in Mossel et al. (2000), for non-Newtonian behaviour. Thus, heather honey (*Calluna vulgaris*), buckwheat honey (*Fagopyrum esculentum*), white clover honey

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doi:10.1016/j.jfoodeng.2004.12.017

(*Trifolium repens*), New Zealand manuca honey (*Leptospermum scoparium*) and Indian Karvi honey (*Carvia callosa*) have shown thixotropic behaviour, while dilatancy has been detected in Nigerian honey (*Opuntia engelmanni*) and several eucalyptus honeys (e.g. *Eucalyptus fisifolia*). The non-Newtonian behaviour has been attributed to the presence of colloids or high-molecular weight dextrans.

The aim of the present work was to study the effect of water content on the viscosity of six different unifloral honeys at various temperatures.

2. Materials and methods

Six types of Greek unprocessed honey were studied, namely, two honeydew honeys from *Pinus halepensis* (pine honey) and *Abies cephalonica* (fir honey) and four nectar honeys from *Thymus* spp. (thymus honey), *Citrus sinensis* (orange honey), *Gossypium hirsutum* (cotton honey) and *Helianthus annuus* (helianthus honey). The botanical source of each honey was identified by pollen analysis (Louveax, Maurizio, & Vorwohl, 1978). The samples were obtained directly from the beekeepers. The weight of each sample was approximately 1000 g.

Water content of the samples was measured with a digital ABBE WAY-1S refractometer. The refractive index values were converted to moisture contents (AOAC, 1990). The viscosity of each sample was measured initially at the original moisture content. The necessary amount of distilled water, determined by mass balance, was added to each sample to adjust its moisture content to the predetermined values of 17%, 19% and 21%.

Sugar analysis was performed by HPLC (Waters, RI 410) using a Hichrom 250 mm × 4.6 mm column with 1.6 ml/min flow rate. The mobile phase was acetonitrile and water (80:20). Five grams of honey was dissolved in acetonitrile and water (50:50) and transferred into 100-ml flask. The sample was poured through a 0.45 μm filter and collected in sample vials. Twenty microlitres at 30 °C was used for the analysis. The method was after sugar analysis of the Apimodia Honey Commission (Bogdanov, Martin, & Lullman, 1997).

Viscosity was measured using a HAAKE model VT 500 concentric cylinder rotational viscometer with MVDIN sensor with 21 mm internal radius of the outer cylinder, 19.36 mm radius and 58.08 mm length of the inner cylinder. The viscometer was connected to a PC with VT500 version 1.3 software. The PC automatically was increasing the rotational speed of the sensor so that a shear stress vs. shear rate curve was obtained for shear rates from 5 to 100 s^{-1}.

The presence of air bubbles and crystals can affect the viscosity of honey. To dissolve any crystals present in the samples, each sample of about 150 ml was heated to 45 °C for 3 h in a thermostatically controlled container. Then each sample was heated to 50 °C for 30 min, while the sensor, dipped in the sample, was slowly rotating at 8 rpm to facilitate the release of any bubbles that might have been trapped in the sensor. After 24 h, without removing the sensor from the sample, the sample was heated to the selected temperature for 40 min and the viscosity was measured.

Viscosity was measured at 25, 30, 35, 40 and 45 °C at the initial moisture, at 17%, 19% and 21% water content. All experiments were run in duplicate and the average values are reported here.

3. Results and discussion

The initial moisture content of the samples varied from 15% to 17.1% as shown in Table 1. Moisture content of honey usually varies from 14% to 18%, but it must not exceed 20% according to the Greek law. Sugar analysis is shown in Table 2. Fructose and glucose represent the major part of sugars in honey. Pine, fir and thymus honeys contain small amounts of disaccharides and trisaccharides with pine and fir having the higher concentration of disaccharides and trisaccharides.

Viscosity values are shown in Table 3. As expected, viscosity decreases substantially as moisture content and temperature increases. The effect of moisture is more pronounced for moisture content up to about 19%. Above this level the effect is weak. The effect of temperature is more pronounced for temperatures up to 30 °C. The effect is much less at temperatures 35–45 °C.

A plot of ln μ vs. $1/T$ gave a linear relationship for all samples indicating that the Arrhenius equation (Eq. (1)) can be applied to describe the variation of viscosity of honey with temperature.

$$\mu = \mu_o \exp(E_a/RT) \quad (1)$$

where μ is the viscosity (Pa s), μ_o a constant (Pa s), E_a the activation energy (kJ/mol), R the gas constant (0.00831434 kJ/mol K) and T the temperature (K). Bhandari et al. (1999), Mossel et al. (2000), Lazaridou et al. (2004) have used Arrhenius equation to predict temperature dependence in honey.

The best-fitting straight lines through all the experimental points are shown in Fig. 1, where the logarithm

Table 1
Initial moisture content of the samples

Sample	Moisture (%)
Pine	17
Fir	15
Cotton	15
Helianthus	17.1
Orange	15.9
Thymus	16.4

Table 2
Sugar composition of the samples (%)

Sample	Fructose	Glucose	Sucrose	Turanose	Maltose	Isomaltose	Rafinose	Erlose	Melezitose
Pine	31.3	26.9		2.3	2.7	1.6			0.3
Fir	29.9	20.1	0.8	0.6	4.0	0.6			
Cotton	44.1	33.6		2.2					
Helianthus	38.1	34.9	0.3						
Orange	41.0	31.0	0.7						
Thymus	41.6	30.8	0.6				1.7	0.2	

Table 3
Viscosity values for the different honeys as a function of moisture content at different temperatures

Honey	Viscosity (Pa s)					
	Moisture (%)	25 °C	30 °C	35 °C	40 °C	45 °C
Pine	17	20.765	11.465	6.453	3.646	2.284
	18	13.535	7.182	4.071	2.371	1.445
	19	9.688	5.289	3.045	1.815	1.119
	21	4.801	2.767	1.634	1.015	0.662
Fir	15	[a]	26.52	13.44	7.477	4.350
	17	17.46	9.110	4.999	2.889	1.810
	19	8.065	4.455	2.694	1.538	1.002
	21	4.384	2.538	1.552	1.007	0.696
Cotton	15	23.405	12.13	6.751	3.779	2.243
	17	8.064	4.413	2.587	1.568	1.010
	19	4.076	2.315	1.390	0.883	0.595
	21	2.541	1.494	0.928	0.612	0.421
Helianthus	17.1	12.28	6.497	3.690	2.203	1.376
	18	8.278	4.600	2.643	1.587	1.037
	19	5.120	2.894	1.170	1.057	0.676
	21	3.367	1.944	1.182	0.738	0.506
Orange	15.9	18.39	9.504	5.220	3.066	1.873
	17	10.67	5.736	3.249	1.952	1.247
	19	5.548	3.092	1.818	1.138	0.751
	21	3.202	1.826	1.123	0.721	0.502
Thymus	16.4	16.930	8.772	4.839	2.683	1.600
	17.4	11.040	5.874	3.310	1.928	1.187
	19	6.084	3.339	1.939	1.195	0.777
	21	3.184	1.825	1.114	0.735	0.502

[a] Viscosity value outside the range of the sensor.

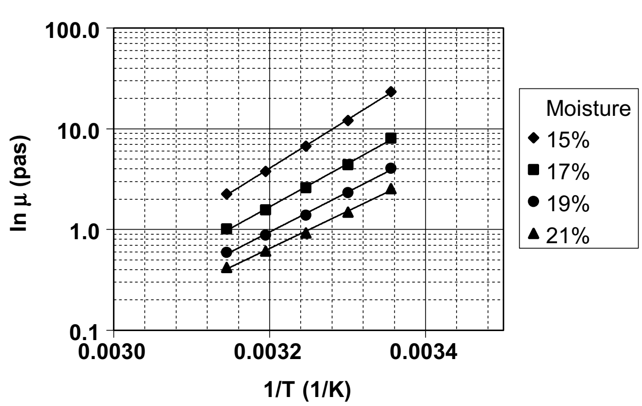

Fig. 1. Typical Arrhenius plot for cotton honey at various moisture content levels.

of viscosity is plotted vs. $1/T$ for cotton honey. Similar plots were obtained for all the samples with R^2 ranging between 0.9951 and 0.9999. The activation energy and the constant μ_o were obtained from these regression lines at each moisture content. The activation energy decreases as the moisture content increases (Fig. 2) indicating that the viscosity is more sensitive to temperature changes at low moisture contents. It varies between 70.8 kJ/mol (cotton honey at 21% moisture) and 96.3 kJ/mol (fir honey at 15% moisture). Pine, fir and thymus honey have the higher activation energy at the same moisture content. Mossel et al. (2000) found activation energies for some Australian honeys to vary between 66.315 and 124.493 kJ/mol. Lazaridou et al. (2004) found activation energies for some Greek honeys to vary from 69.1 to 93.75 kJ/mol. The constant μ_o increases exponentially as the moisture content increases. The change of the activation energy and the constant μ_o with moisture content are given by the equations shown in Table 4, which were obtained with regression

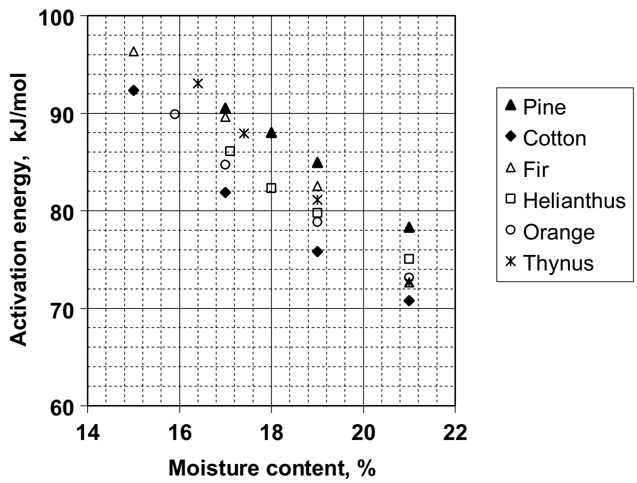

Fig. 2. Activation energy vs. moisture content for the different honey types.

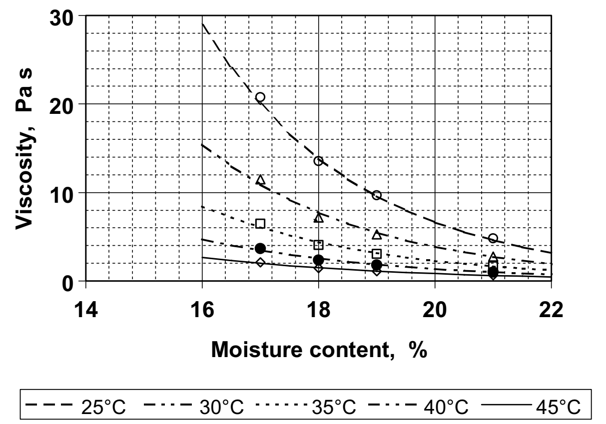

Fig. 3. Viscosity of pine honey vs. moisture content at 25, 30, 35, 40 and 45 °C (points, experimental values; lines, predicted values using the Arrhenius model).

analysis. Lines in Figs. 3–8 represent the predicted values of viscosity using Eq. (1) with E_a and μ_o values given by the equations of Table 4. The experimental values are also included in these plots. Average percent deviations between experimental and predicted values calculated by Eq. (2) for all temperatures and moisture contents for each honey type are: 3.75% for pine, 11.5% for fir, 8.77% for helianthus, 6.49% for orange, 3.65% for thymus, and 13.54% for cotton honey.

$$\text{Dev}\% = \frac{100}{n} \cdot \sum \frac{|\mu_{\text{exp}} - \mu_{\text{pred}}|}{\mu_{\text{exp}}} \tag{2}$$

where μ_{exp} experimental viscosity value, μ_{pred} predicted viscosity value (with Eq. (1) and equations from Table 4), n number of experimental points for each honey type.

A plot of viscosity for the six different honey types vs. moisture content at 25 °C, as calculated with Eq. (1), shows that pine and fir honey have the higher viscosity at a given temperature and moisture content followed by thymus honey (Fig. 9). At higher temperatures the difference in viscosity among the six honey types decreases but still exists even at 45 °C. The difference could be attributed to the composition of sugars and colloid materials present in the honey and reveals the importance of the botanical origin of honey on its viscosity.

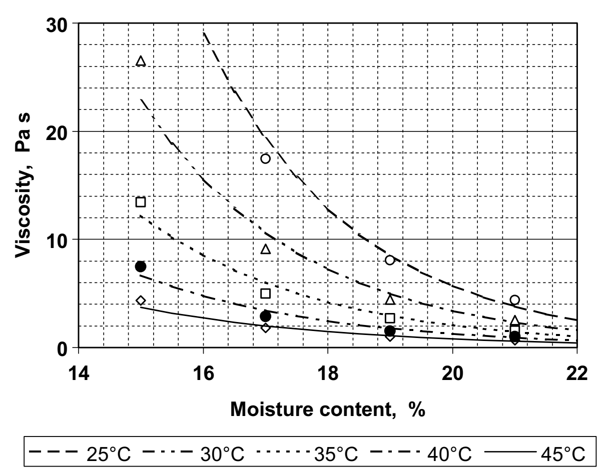

Fig. 4. Viscosity of fir honey vs. moisture content at 25, 30, 35, 40 and 45 °C (points, experimental values; lines, predicted values using the Arrhenius model).

Honey is known as Newtonian liquid although there are reports in the literature for dilatant behaviour and thixotropic behaviour of some types of honey. Non-Newtonian behaviour was tested by measuring the viscosity of all samples at various shear rates (5 s^{-1} to 100 s^{-1}). Shear stress was always a linear function of

Table 4
Activation energy (E_a) and constant μ_o of the Arrhenius equation for the different honeys as a function of moisture content M (in%)

	E_a (kJ/mol)	R^2	μ_o (Pa s)	R^2
Pine	143.29–3.0851*M	0.997	7.9115E–22*EXP(0.8776*M)	0.988
Fir	155.62–3.9073*M	0.991	1.0226E–23*EXP(1.1704*M)	0.973
Cotton	143.81–3.5339*M	0.969	3.1134E–22*EXP(1.0563*M)	0.969
Helianthus	132.25–2.7392*M	0.982	1.9798E–20*EXP(0.7766*M)	0.980
Orange	140.06–3.2046*M	0.990	9.7741E–22*EXP(0.9557*M)	0.990
Thymus	164.79–4.3955*M	0.999	8.5418E–26*EXP(1.4070*M)	0.999

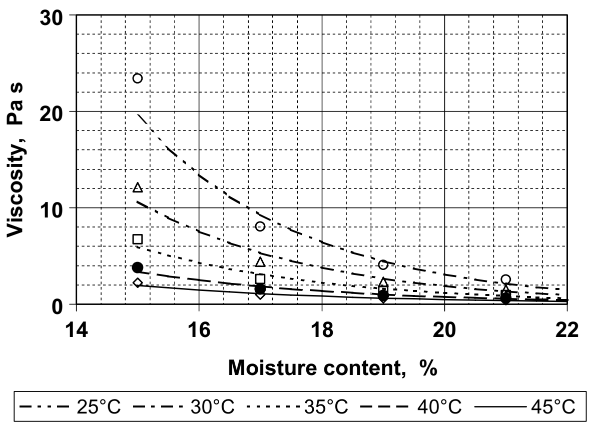

Fig. 5. Viscosity of cotton honey vs. moisture content at 25, 30, 35, 40 and 45 °C (points, experimental values; lines, predicted values using the Arrhenius model).

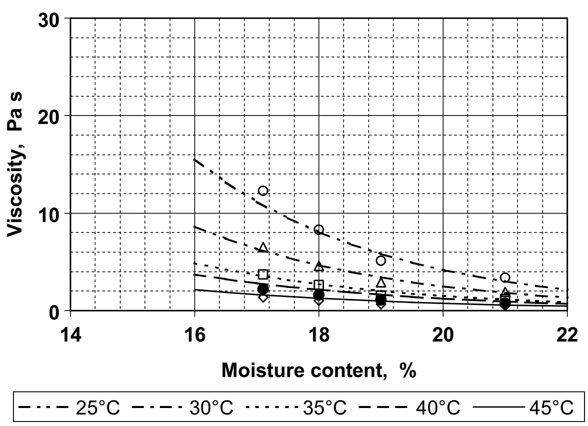

Fig. 6. Viscosity of helianthus honey vs. moisture content at 25, 30, 35, 40 and 45 °C (points, experimental values; lines, predicted values using the Arrhenius model).

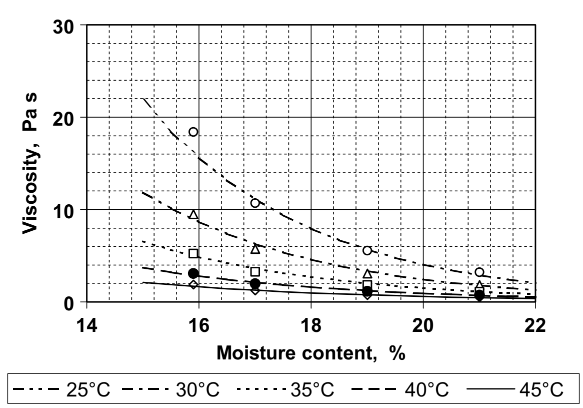

Fig. 7. Viscosity of orange honey vs. moisture content at 25, 30, 35, 40 and 45 °C (points, experimental values; lines, predicted values using the Arrhenius model).

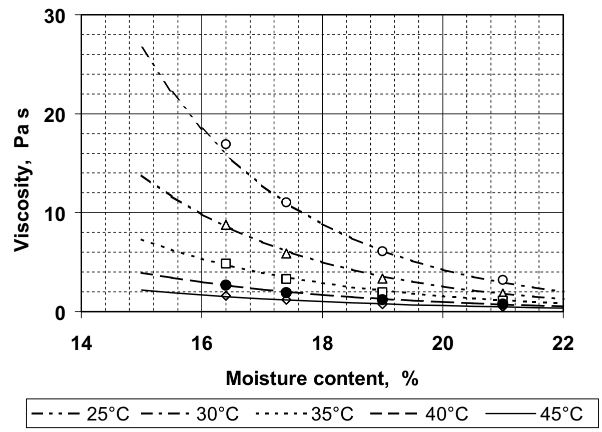

Fig. 8. Viscosity of thymus honey vs. moisture content at 25, 30, 35, 40 and 45 °C (points, experimental values; lines, predicted values using the Arrhenius model).

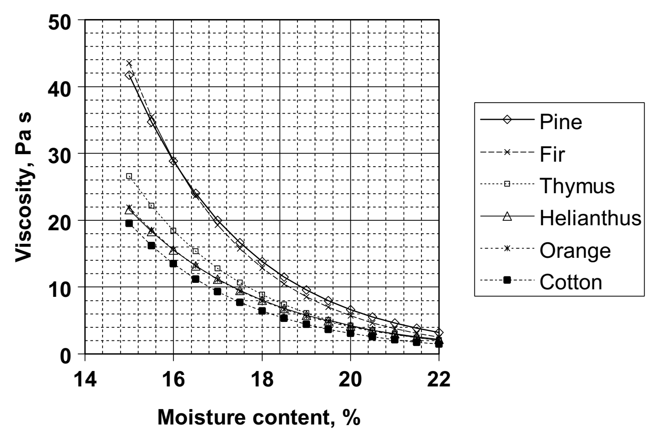

Fig. 9. Viscosity of the different honey types at 25 °C as predicted with Arrhenius model.

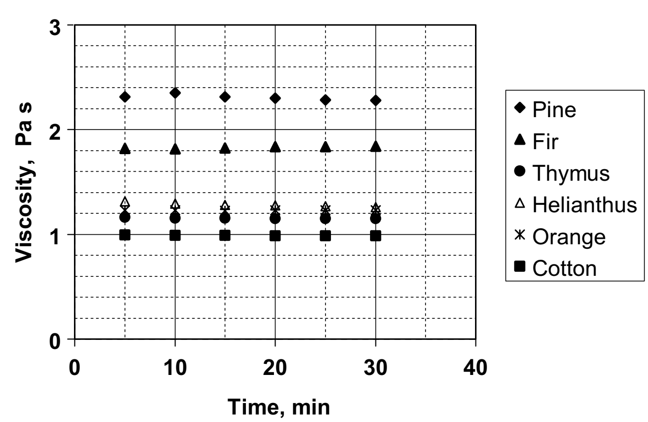

Fig. 10. Viscosity of the different honey types vs. time at 40 s^{-1} shear rate, 45 °C and 17% moisture content for pine, fir, orange, cotton, 17.1% for helianthus and 17.4% for thymus.

shear rate indicating Newtonian behaviour. The viscosity was also measured at constant shear rate (40 s^{-1}) as

a function of time at 45 °C at the original moisture content, at 17% (except for thymus which was at

17.4% and helianthus at 17.1%), 19% and 21% for all the samples to test for any time dependency behaviour. Viscosity was time independent, as shown in Fig. 10 where the viscosity vs. time is plotted for the samples with 17% moisture (except for thymus which was at 17.4% and helianthus at 17.1%). Similar plots where obtained for the other moisture levels.

4. Conclusions

Viscosity of honey varies with temperature, moisture content and its botanical origin. The viscosity of pine and fir honey was found to be substantially higher than the viscosity of thymus, cotton, helianthus and orange honeys at the same temperature and moisture content especially at the lower end of the moisture and temperature range tested (15–21% moisture and 25–45 °C). Arrhenius equation described satisfactorily the variation of viscosity with temperature. The activation energy decreased as the moisture content increased indicating that the viscosity is more sensitive to temperature changes at low moisture contents. All the samples showed Newtonian behaviour.

References

Abu-Jdayil, B., Ghzawi, A. M., Al-Malah, K., & Zaitoun, S. (2002). Heat effect on rheology of light- and dark-colored honey. *Journal of Food Engineering, 51*, 33–38.

Anupama, D., Bhat, K. K., & Sapna, V. K. (2003). Sensory and physico-chemical properties of commercial samples of honey. *Food Research International, 36*, 183–191.

AOAC (1990). *Official methods of analysis* (15th ed.). Arlington, VA: Association of Official Analytical Chemists.

Bhandari, B., D'Arcy, B., & Chow, S. (1999). Rheology of selected Australian honeys. *Journal of Food Engineering, 41*, 65–68.

Bogdanov, S., Martin, P., & Lullman, C. (1997). Methods of the Apimondia. *Honey Commission Apidologie Extra Issue*, 42–45.

Junzheng, P., & Changying, J. (1998). General rheological model for natural honey in China. *Journal of Food Engineering, 36*, 165–168.

Lazaridou, A., Biliaderis, C., Bacandritsos, N., & Sabatini, A. G. (2004). Composition, thermal and rheological behaviour of selected Greek honeys. *Journal of Food Engineering, 64*, 9–21.

Louveax, J., Maurizio, A., & Vorwohl, G. (1978). Methods of melissopalinology. *Bee World, 59*, 139–157.

Mossel, B., Bhandari, B., D'Arcy, B., & Caffin, N. (2000). Use of an Arrhenius model to predict rheological behaviour in some Australian honeys. *Lebensmittel Wissenchauft und Technologie, 33*, 545–552.

Sopade, P. A., Halley, P., Bhandari, B., D'Arcy, B., Doebler, C., & Caffin, N. (2003). Application of the Williams–Landel–Ferry model to the viscosity–temperature relationship of Australian honeys. *Journal of Food Engineering, 56*, 67–75.