

Effect of Ultrasonication Process on the Physical Properties of Three Different Honey Varieties

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ABSTRACT

Honey is widely known for its nutraceutical properties and purity. It is a sweet and viscous food in nature which is prepared by Honey bees, and have been recommended for consumption purpose. Honey possesses antioxidant activities, anti-inflammatory properties, antiseptic, anti-allergen properties, and thus used as a portion of therapeutic food for skin disease, intestinal problems and anemia. These qualities of honey get attention for use of honey to human consumption. To date, available honey in the market is a pasteurized product that is thermally treated. The thermal treatment (pasteurization) process leads to degradation of honey quality by changing the Color and also influences the physical properties. An increase in temperature during the thermal process leads to the inversion of sugars and also increases hydroxymethylfurfural (HMF) which affects the color of honey. The use of non-thermal technologies like ultrasound treatment overcomes the quality-related aspects of honey. In the present study, three varieties of Honey (Jungle, Tulsi, Eucalyptus honey) were selected to see the effect of the Ultrasonication process on the rheological properties. Experimentation results have not demonstrated any significant change in color among varieties, but Eucalyptus honey had a minimum color difference ΔE of 15.87 after ultrasound treatment. The viscosity of the Honey sample (Tulsi) was found to be decreased (97.45%) for temperature variation from 20 °C to 100 °C.

Keywords: Ultrasound, Honey, Viscosity, Colour, Total soluble solids

INTRODUCTION

Honey is a naturally occurring sweet substance classified by Codex Alimentarius Commission, that is produced by honey bees from plant nectar. The definition was modified later as "the natural sweet substance produced

by *Apis mellifera* bees" to separate it from the honey produced by other bee species such as *Micrapis*, *Megapis*, and *Meliponines* (Thrasylvoulou et al., 2018). From ancient times, honey has been used for various therapeutic purposes.

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Honey is used in food mixture as a sweet Indian product called "Gur" or "Jamun Ghutti" (homemade sugar water), a herbal decoction with honey is used as feed to 1st day of life to a newborn baby (Brown, 1978). According to Ayurveda, honey is a boon to the people suffering from the digestive related problems, irritating cough, teeth and gum health, skin disorder, anemia cardiac pain, and palpitation, and also used to treat insomnia. Insomnia is known to its hypnotic actions for human beings.

Ayurveda, states 8 different varieties of honey that can be differentiated by its treatment and functionality (Ediriweera & Premarathna, 2012). In Egyptian culture, honey is known for the antibacterial properties due to healing of the wound, like tropical ointment. Treatment of Diarrhea and tuberculosis were also suggested by ancient Islamic culture and by Avicenna respectively (Eteraf-Oskouei & Najafi, 2013). Presently, honey is used at household level due to its antiseptic, antioxidant, and anti-allergens properties. The use of honey has increased for its other properties such as healing, moisturizing, and anti-anemic. Contamination of Honey leads to many problems associated with human health. According to European union regulations (Council directives 2001/110/EC 2002) states that "honey must be free from any outside organic and inorganic residue" (Qi et al., 2018) in between 2009 to 2013, RASFF (Rapid system for food and feed) portal revealed that, about 71% of antimicrobial residue in honey sample collected worldwide.

Consumers now days are conscious about their health that is associated with nutritious intake of food. Concern over food quality has also been grown rapidly for maintaining a good health. Present research has focused on the preservation of Physico-chemical properties of honey for better quality and nutrition (Galarini, Saluti, Giusepponi, Rossi, & Moretti, 2015; Machado De-Melo, Almeida-Muradian, Sancho, & Pascual-Maté, 2018). Non-thermal techniques currently are the best technique for the preservation and maintaining

the quality aspects of food. Ultrasound is a type of non-thermal technology that also considered as the green technology (Qi et al., 2018). Ultrasound technology is used to perform many functions in food industries such as mixing, grinding, heat and mass transfer, energy transfer, extraction of selective oils and oleoresins. Ultrasound units generally is smaller in size but faster in response time, use less energy while providing better quality compared to other non-thermal methods (Ruiz-Matute, Brokl, Soria, Sanz, & Martínez-Castro, 2010) (Tewari & Irudayaraj, 2004). Ultrasound technology leads to conversion of Honey into liquid form as sugar crystals get dissolved and hinder the process of crystallization. Hinder effect caused by ultrasound increases the shelf life of honey as it kills present yeast cells, while other microbes lose their ability to grow (Alqarni, Owayss, Mahmoud, & Hannan, 2014). Ultrasound treatment at 60 and 100 amplitude, 20KHz for 8 min with pulsation time of 5 seconds nominally change the pH, viscosity, color, HMF value, and diacetase values.

MATERIALS AND METHODS

Jungle, Tulsi, and Eucalyptus honey were three different varieties of Honey samples procured from the local market of Thanjavur District (Tamil Nadu). Procured sample of honey was not processed and had the same quality as raw Honey. These samples were stored in a dark bottle at the low temperature of about 4°C. These samples were used to conduct the experiments. Further, due to the application of ultrasound treatment, the differences like change in color, viscosity, Brix, and moisture content were recorded for all three varieties of honey.

2.1 Ultrasonic treatment of honey

Honey samples of these varieties were taken in 100 ml of the beakers, and was placed in an ultrasound treatment chamber. Ultrasound treatment parameters were used from the studies conducted by (Janghu, Bera, Nanda, & Rawson, 2017). The ultrasound system (750 ultrasonic processor, sonic and materials, Inc., Newton, CT, USA) with a constant frequency

of 20KHz and 19 mm diameter food-grade probe was attached to the transducer for pulsation. The ultrasound treatment of honey was used at 60% amplitudes. The volumes of the samples were kept constant at 50ml. Distance between the sample and probe was adjusted as per the experiment requirement, it was made sure initially that all the samples placed at the same distance and the probe should not touch the bottom of the beaker. Above all, it was considered that the temperature of all the honey samples during processing and after processing should not increase 40°C.

2.2 Color measurement

$$\Delta E = [(L_0^* - L^*)^2 + (a_0^* - a^*)^2 + (b_0^* - b^*)^2]^{1/2} / 2 \quad (1)$$

2.3 pH value, Brix %, and Moisture content

Acidity of the honey is vitally important parameter and also plays a important role for increasing the shelf life. Therefore, acidity of honey was checked using a digital pH meter (pH tutor, Eutech Instruments, Thermo Fisher Scientific, Singapore).

RX7000 α refractometer (ATAGO, Tokyo, Japan) was used to measure Brix % of honey samples at 20°C. The moisture content of the

HunterLab (Hunter Associates Laboratory, Inc., Reston, VA, USA) was used to quantify color parameters for control and ultrasound treated honey samples. The values of 3-D color space are determined in L*, a* and b*. Here L* represents luminance towards brightness (100% black to 100% white values), a* axes represent range from greenness (-a*) to redness (+a*) and b* axes range from blueness(-b*) to yellowness (+b*) respectively. All the readings were taken in replication of triplicate form.

The following formula was used for calculating ΔE^* value (color difference)

honey was determined using the standard reference table of honey refractive indices and water content, according to Harmonized Method of the International Honey Commission (Bogdanov & Commission, 2009). The reading of each sample was taken in triplicate form and then the average was taken for the study purpose.

2.4 Viscosity



Fig. 1: MCR 52 modular compact rheometer with 25 mm parallel plate type of probe- for analysing viscosity with respect to temperature.

Viscosity is associated with the flow properties or this can be defined as the ability of honey to resistance of flow. One gram of control and ultrasound treated honey was taken on to the temperature-controlled stage of MCR 52 modular compact rheometer with 25 mm parallel plate type of probe. The viscometer was connected to the PC with Rheoplus software. Using rheomanager application - Temperature Ramp: Constant Shear, Temperature- dependency liquid-like sample was chosen for running the sample. The sample was run from minimum temp i.e, 20°C to 100°C. The readings were recorded every 2 sec of time duration. Viscosity values at every 2 sec were recorded in rheometer as viscosity vs temperature graph.

RESULT AND DISCUSSION

Honey has all nutritious composition known for its health benefits for human beings that were expected in its processed form. Basically, the quality of honey not only depends on its internal parameters but also on the external factors like the variety of honey bees used during honey production, the orchids or plant source from where they collect nectar, climatic condition, floral origin, processing technique and one of the major factor in acceptability among consumers. Changes in these parameters can be sometimes noticeable as the change in the color, aroma and taste (Önür et al., 2018). Janghu et al., 2017 have discussed

the tendency of honey to form crystals that was formed at room temperature, due to the presence of glucose was converted into glucose monohydrate. And this becomes a limitation for the honey industry as it increases the viscosity thereby increases the stickiness. To date, the common commercial approach which was opted is thermal processing. This may liquefy honey but it may degrade the nutritional quality of the honey as well.

3.1 Colour measurement

Light color honey has higher acceptability and commercial value than dark-colored honey (Gonzales, Burin, & del Pilar Buera, 1999). **Fig. 2** shows the significant increase in the lightness of colour for all the three varieties of honey. (Chaikham, Kemsawasd, & Apichartsrangkoon, 2016) has reported similar results at 40% amplitude and 20kHz for 30 min and also reported a minimal alteration in the color, pH, enzymatic activity and noticeably improved the mass fractions of phenols, flavonoids in and antioxidant activity of logan, lychee and wildflower honey as compared to thermal treatment. In the current study Eucalyptus, thermally treated honey sample showed redness value and thereby in tulsi, whereas yellowness was much more observed in jungle honey and thereafter in tulsi and eucalyptus. Change in colour (ΔE) value was found to be maximum in jungle honey variety followed by Tulsi Honey then by Eucalyptus honey.

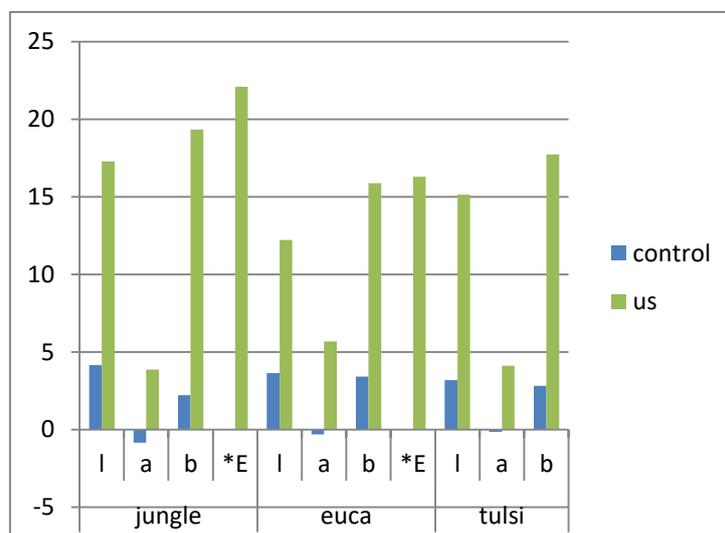


Fig. 2: Colour profile of control and ultrasound treated honey sample (60 % amplitude).

Thus, this clearly indicates that the browning reaction occurs due to the high temperature. The Jungle variety of honey showed a highest ΔE value, these changes could be due to the cavitation effect producing physicochemical reactions. The decrease in lightness was the lowest in the ultrasonically treated sample compared to the control sample. The decreases

of ΔE were found to be attributed due to Maillard reaction, fructose caramelization or/and polyphenol reactions] or/ and the increase in flavonoids that contain the honey they are related to the color, and the increased effect could be due to the reduction in crystal size.

3.2 pH, TSS and Moisture content

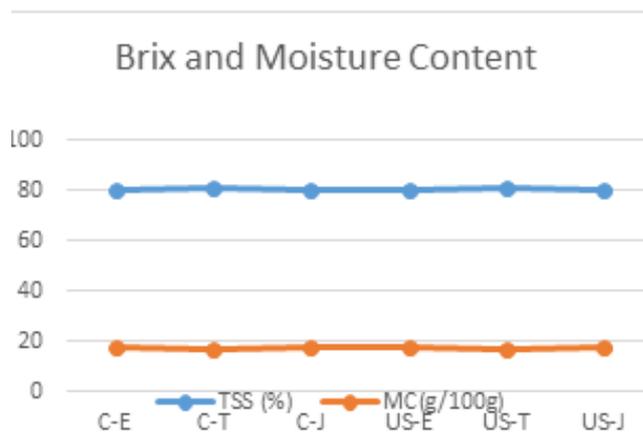


Fig. 3: TSS and Moisture Content of the control and ultrasound honey (60% amplitude). Here, C-E: Control Eucalyptus; C-T: Control Tulsi; C-J: Control Jungle; US-E: Ultrasound Eucalyptus; US-T: Ultrasound Tulsi; US-J: Ultrasound Jungle

Ultrasound-treated eucalyptus honey, tulsi honey, and jungle honey had 3.41, 3.22 and 3.6 pH respectively shown in **Fig 3**. The ultrasound treated honey did not showed any significant effect on pH as compared to the control sample (eucalyptus honey (3.48 pH); tulsi honey (3.5 pH); jungle honey(4.0 pH)) . (Janghu et al., 2017) pH decreases with increase in exposure to heat for a longer period of time. The decrease in pH can be due to an increase in hydrogen ion concentration release while heating or it may also be due to the presence of pollens which releases organic acids while it is thermally processed, and this affects the pH of the final sample (Chaikham & Apichartsrangkoon, 2012).

Moisture content of hiney is a vital parameter in terms of deciding shelf life and rheological parameters. Maximum moisture content were found in Eucalyptus Honey and jungle honey at 18% amplitude, followed by Tulsi honey in the control sample showed a negligible change in all three varieties. In commercial samples due to thermal

processing, some of the moisture was lost, which was reported as negligible when honey treated with ultrasound as shown in **Fig. 3**. Amount of TSS correlates with the amount of moisture present in the honey sample. Ultrasonic treatment samples for all the honey showed no significant difference in moisture content and TSS value than the control sample as shown in **Fig.3**. But this can be possible that the bubbles which form during the ultrasound treatment affects the TSS of the honey.

3.3 Viscosity measurement

Honey is known as a viscous fluid and this makes it difficult to flow. The rheological property of honey is a very important parameter to control and process design in the honey for the food industry. Rheology studies reviled the flow and deformation properties of the material with changing conditions internally or externally. The flowing property of any food material is dependent on the viscosity of the material. The reciprocal of viscosity is fluidity or flow ability, where if viscosity indicates opposition to flow, the

fluidity is the measure of the easy flow of material. The viscosity of the material depends on the internal friction between the molecules

and this friction opposes the development of velocity differences within the fluid.

Table 1: Data and Result in determining the effect of temperature on control Jungle, Tulsi, and Eucalyptus Honey Viscosity

Sample	T(°C)	T(K)	1/K	μ (mPaS)	$\ln \mu$	$\ln \mu_0$	μ_0	Ea/R	R (J/mole K)	Ea (Kj/mole)
CJ	20	293.15	0.003411	4.497	1.503411	-13.8155	1.00E-06	9.343917	8.3145	77.69
CE	20	293.15	0.003411	9.853	2.287776					
CT	20	293.15	0.003411	5.808	1.759236					
CJ	60	333.15	0.003002	2.832	1.040983					
CE	60	333.15	0.003002	0.8623	-0.14815					
CT	60	333.15	0.003002	0.95	-0.05129					
CJ	100	373.15	0.00268	0.56	-0.57982					
CE	100	373.15	0.00268	0.5773	-0.54939					
CT	100	373.15	0.00268	0.702	-0.35382					

Honey is known as a non-Newtonian fluid because this does not follow Newton's law of viscosity. The viscosity of the honey depends on the total solid content in honey and moisture content present. Honey contains a good amount of concentrated sugars which is responsible for its viscous nature. In the present study, experiments were conducted to understand the nature of viscosity changes in all three varieties of honey with respect to changes in temperature **Table 1**. In this study, viscosity of a controlled sample of all varieties of honey was compared with an ultrasound treated sample of all three varieties of honey. There was a constant linear decrease of viscosity with increasing temperature. Initially,

the eucalyptus honey was having a higher viscosity of about 9.853 PaS and thereby of tulsi and jungle honey (5.808 and 4.497 respectively). After the ultrasound treatment of these samples, the ultrasound treated Tulsi honey was having a higher viscosity of about 14.98 PaS and thereby of the jungle and eucalyptus honey 9.65 and 7.28 respectively **Table 2**. The sudden increase may be caused due to the resistance caused by the small bubbles which are formed during ultrasound treatment. **Fig 4** revealed that there is a significant difference in the drop-down of viscosity as compared to control samples this may be due to the damage of bubbles caused once the temperature is given to the sample.

Table 2: Data and Result in determining the effect of temperature on ultrasound treated Jungle, Tulsi, and Eucalyptus Honey Viscosity

Sample	T(°C)	T(K)	1/K	μ (PaS)	$\ln \mu$	$\ln \mu_0$	μ_0	Ea/R	R (J/mole K)	Ea (Kj/mole)
US-J	20	293.15	0.003411	9.65	2.266958	-13.8155	1.00E-06	9.343917	8.3145	77.69
US-E	20	293.15	0.003411	7.28	1.985131					
US-T	20	293.15	0.003411	14.95	2.704711					
US-J	60	333.15	0.003002	0.38	-0.96758					
US-E	60	333.15	0.003002	0.81	-0.21072					
US-T	60	333.15	0.003002	0.81	-0.21072					
US-J	100	373.15	0.00268	0.48	-0.73397					
US-E	100	373.15	0.00268	0.45	-0.79851					
US-T	100	373.15	0.00268	0.377	-0.97551					

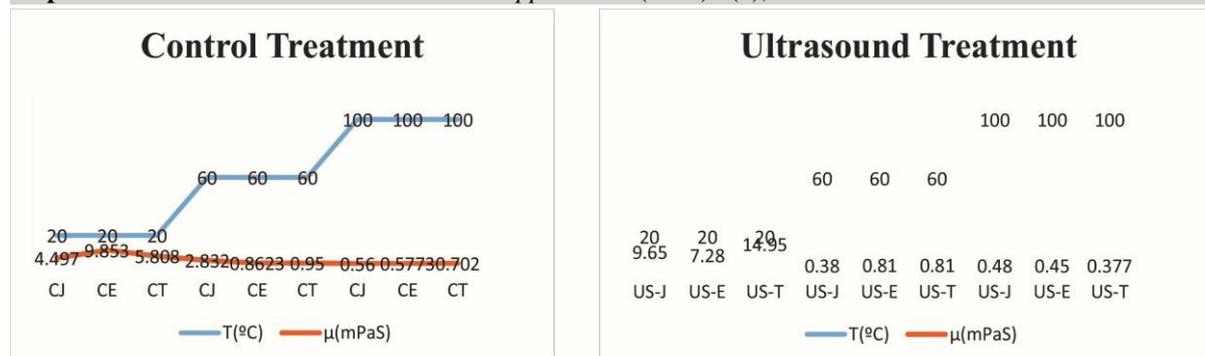


Fig. 4: Viscosity of Control and Ultrasound treated honey at different temperatures

The Arrhenius equation is the simplest model to relate the effect of temperature on the physical or chemical properties of honey (Bambang, Ikhsan, & Sukri, 2019).

$$\mu = \mu_0 e^{E_a/RT} \tag{2}$$

after linearization the eq. becomes

$$\ln \mu = \ln \mu_0 + (E_a/R)(1/T) \tag{3}$$

The following table represents the parameters required for the measurement of honey. (Rao, 1986) reported that the activation energy of honey obtained was about 77.69 kJ/mole. The model relates the dependency viscosity to temperature. (Önür et al., 2018) reported that natural crystals of honey were broken down or liquefied. Pre-liquefaction exhibited a non-Newtonian shear thinning behavior this can be related to the presence of sugar crystal in samples, however, after ultrasound treatment, the post-liquefaction, the viscosity of all honey samples exhibited Newtonian behavior. The following author reported a decrease in the viscosity after ultrasound treatment of honey, this was found to be opposite to the results of the present study. This can be the result of using probe type of ultrasound treatment instead of water bath type of ultrasound treatment. Where probe directly comes in contact with the sample and sound waves given to the sample unlike water bath ultrasound where the sound waves travel through water medium and then are passed on to sample.

CONCLUSION

The present study showed that there was no significant differences in the physical properties of eucalyptus, tulsi and jungle

honey. The current research showed that the physical properties like moisture, pH, color, brix% were least affected at 60% amplitude. the effect of the temperature on honey viscosity was best modeled with the Arrhenius model. Ultrasound-treated tulsi honey had a 97.45% decrease in viscosity which was to rapidly drop down to 1.37 PaS at 100°C which was found to be the lowest among all the samples. This phenomenon can be due to the firm bubbles formed during ultrasound treatment. Unlike heat-treated honey, ultrasound treated honey didn't darken the honey color which means that it resists millard reaction and the formation of Hydroxymethyl furfural leading to a better commercial sample. However, there is further research needed for the microbiological and storage study at various temperature of ultrasound treated honey.

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