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Modelling the Rheological behaviour of tender coconut (*Cocos nucifera* L) water and its concentrates

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Abstract

The rheological behaviour of tender coconut (*Cocos nucifera* L) water was studied as a function of total soluble solid (TSS) content (5.3 to 52.9 °Brix) and its corresponding water activity (a_w) (0.982 to 0.870) at a wide range of temperature (10 to 85°C) by controlled stress rheometer using concentric cylinders. The rheological parameter shear stress was measured up to the shear rate of 1000 s^{-1}. The investigation showed that tender coconut water and its concentrate behaved like a Newtonian fluid and the viscosity (η) was in the range 3.80 to 34.88 mPa s depending upon the concentration and temperature used. The temperature dependency on viscosity of tender coconut water was described by Arrhenius equation (r > 0.96) and the activation energy (E_a) of viscous flow was in the range 5.268 to 20.798 kJ/mol depending upon the total soluble solid content. The effect of total soluble solid content on flow activation energy was described by exponential equation (r>0.92, rmse%=11.4, p<0.05) and that of water activity was described by power law equation (r>0.98, rmse %=5.54, p<0.01). The effect of total soluble solid content on viscosity of tender coconut water followed second order polynomial exponential equation (r >0.99, rmse %< 3.98) at the temperature used. The effect of water activity on viscosity was described by both power law as well as exponential type relationship (r>0.99). The combined effect of temperature and total soluble solid content/water activity on viscosity was described by the equations

$\eta = 2.007\text{x}10^{-2} \exp(1710.5/T - 0.0157C + 0.000728C^2)$, (r >0.93, rmse%=3.95, p<0.05)
$\eta = 1.42 \times 10^{-2} (a_w)^{-11.217} \exp(1705.96/T)$, (r >0.93, rmse%=3.95, p<0.05)
$\eta = 2891.7^* \exp(1704.20/T - 12.256a_w)$, (r>0.93, rmse%=3.97, p<0.05)

where η is viscosity in mPa s, T is temperature in °K and C is the total soluble solid content in °Brix, a_w is the water activity (-).

Introduction

The rheological properties of fluid foods is an important property which has several applications in food science and technology, such as, in developing food processes, processing equipments, quality evaluation and structural understanding of food and raw agricultural materials. The rheological behaviour of food products cannot be predicted theoretically, due to complicated physical, chemical and biological structure of the food material. Therefore, experimental measurements of rheological properties are important in characterisation of fluid foods (Krokida *et al*., 2001). The processing and preservation of fluid foods by thermal treatment such as, heat pasteurisation, evaporation and sterilisation is favoured for longer periods of storage. The processing of fluid foods involved several unit operations, such as, mixing, pumping, evaporation, pasteurisation, sterilisation etc where the fluid properties such as, viscosity, soluble solid content and temperature vary during processing. The rheological properties and its behaviour during processing of fluid food products are important in determining the power requirements for unit operations such as, pumping, sizing of pipes, design of processing equipment of heat exchangers, mixing, filling etc. It is also important in the calculation of heat, mass and momentum transfer phenomena (Telis-Romero *et al*., 1999; Kimball *et al*., 2004). The parameter of fluid food quality related to rheology is known as mouth-feel and defined as the mingled experience derived from the sensation of skin of the

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mouth after ingestion of a food or beverage and is related to viscosity, density, surface tension and other physical properties of the fluid foods. The physical properties of fluid foods have gained importance as textural attributes of fluid foods have been developed and quantified (Ingate and Christensen, 2007). Fluid foods were subjected to different temperatures and concentrations during processing, storage, transportation, marketing etc. where the rheological properties were essential in processing and handling of the food material. The development of concentrated fluid foods is more advantageous than single strength liquid as concentration of liquid foods leads to decrease in water activity. The reduction in water activity of food leads to better stability; which is convenient during storage, handling, transportation and preparation of novel products with suitable dilution/modifications.

Water activity of food is defined as the ratio of the equilibrium vapour pressure exerted in the food to the vapour pressure of pure water at the same temperature and also defined as the amount of water available for microbial growth. It is the escaping tendency of water fugacity in the system by the escaping tendency of pure water. The fugacity of food systems was closely approximated by vapour pressure. The water activity is a measure of the energy state of the water in the system and it is a measure of free, unbound and available water in the food system. There are several factors that control the water activity in a food system, the colligative effect of dissolved species (salts, sugars and acids) interact with water through dipole-dipole, ionic and hydrogen bonds. Influence of water activity may induce profound changes in the quality and stability of a food product and it is also an important requirement for packaging of food material. Water activity is a critical factor that determines the shelf life of the food. The water activity of food is important rather than total moisture content for deciding the quality and stability of foods (Fennema, 2005).

The rheological behaviour of fluid foods in original and concentrated forms is vital to understanding the pumping and flow requirements in fluid food processing industries including aseptic processing industries. In general, liquid food such as fruit and vegetable juices behave like Newtonian fluids so their flow behaviour would be Newtonian in nature. Several investigators reported that clarified and depectinated juices and their concentrates exhibit Newtonian flow behaviour. (Ibarz *et al*., 1987, 1992a, 1992b; Cepeda and Villaran, 1999; Juszczak and Fortuna, 2004).

$$\sigma = \eta \dot{\gamma} \qquad \text{------ (1)}$$

where σ is shear stress (Pa), η is coefficient of viscosity (Pa s) and $\dot{\gamma}$ is the shear rate (s^{-1}). Several authors had used Newtonian equation for describing rheological behaviour of food products like pomegranate juice (Altan and Maskan, 2005), Pekmez (Kaya and Belibagli, 2002), liquorice extract (Maskan, 1999). However certain juices with low pulp content and soluble solid content less than 30 ^{o}Brix also behaved like a Newtonian liquid (Krokida *et al*., 2001).

Food habits of people are rapidly changing towards the natural, soft and safer drinks, with lower amount of calories free from added ingredients. Coconut water is a natural beverage product with distinct sensory attributes such as aroma, taste and nutritional values. It is also considered as energy drink for sport personals due to its nutritional characteristics. Coconut water, the liquid endosperm that fills in the central cavity of solid endosperm, is the most important nutritious wholesome beverage that nature has provided to alleviate the sultry heat. The natural form of the tender coconut water is sterile and it is used as a natural drink, as an oral rehydration fluid for children and elderly people suffering from gastroenteritis. The medicinal properties of tender coconut water are well documented (Alleyne *et al*., 2005). Tender coconut water is mainly consumed as a soothing natural beverage in its natural form throughout the year in tropical countries. It is a nutritive beverage and contains greater amount of minerals such as sodium, potassium, phosphorus, chlorides, magnesium, ascorbic acid and also sugars (Magda, 1992; Campos *et al*., 1996; Sabapathy and Kumar, 1999). It also contains a variety of trace elements such as zinc, selenium, iodine, sulphur, manganese, boron, molybdenum etc, these were derived from volcanic soils and sea water where coconut palms were grown. All these minerals are in the form of electrolytes, easily absorbable by the human body. Over the past two decades, coconut water has been used extensively for the treatment of cholera, dysentery, influenza and other infectious diseases that result in dehydration. The minerals potassium and magnesium are known to help reduce high blood pressure. One of the useful components in coconut water is cytokinins, which are a class of phytohormones (Kende and Zeevaari, 1997). The tender coconut water has several functional activities such as hydrating electrolytic fluid, anticarcinogenic, antimyocardial infarction, hepatoprotective, antioxidant, anti-ageing, and anti-thrombotic effects (Sylianco *et al*., 1992; Rattan and Clark, 1994; Falck *et al*., 2000; Vermeulen et al., 2002; Loki and Rajamohan, 2003; Anurag and Rajamohan, 2003a, 2003b). The tender coconut water can be used as natural substrate for production of microbial metabolites like bacterial cellulose (Nata-de-coco), pullulan etc (Jagannath *et al*., 2008; Thirumavalavan *et al*., 2009).

The presence of sugars and a number of plant enzymes, tender coconut water has a stronger tendency to undergo chemical, biochemical and microbiological changes which result in spoilage. To reduce the transportation volume and cost associated with whole fruit and to improve the shelf life of green coconut water, processing is essential. The preservation of green coconut water is a worldwide demand processing technology. Different techniques of preservation of tender coconut water are reported in literature such as, pasteurization, filtration, increasing soluble solid content, pH regulation, addition of preservatives, ultrasonic treatment, concentration by reverse osmosis and ultra filtration, carbonation, microwave treatment and spray drying (Bergonia *et al*., 1982; Rosario *et al*., 1986; Dutta, 1995; Reddy, 1995; Reddy *et al*., 2005; Matsui *et al*., 2007, 2008). The tender coconut water is processed by high temperature short-time pasteurization in several countries; the pasteurization temperature being in the range 60 to 100^{o}C (Magda, 1992). Chowdhury *et al*., (2009) studied the processing and preservation of green coconut water by thermal treatment. Several reports are available in literature for development and evaluation of tender coconut water based non alcoholic beverages blended with other fruit juices. The tender coconut water based beverages with stimulant were stabilised by adjusting sugar, citric acid, stimulating agents, suitable thermal treatment and these beverages had a shelf life of six months (Carvalho *et al*., 2007a, 2007b, 2007c; Lima *et al*., 2009). Reddy *et al*., (2007) studied the non thermal sterilisation of green coconut water by two-stage filtration using ashless filter paper and cellulose nitrate membrane. The concentration of tender coconut water is one important process to increase stability and storage and has commercial potential due to the reduction in water content that facilitates minimised processing, handling and transportation costs. The heat treatment of tender coconut water alters its organoleptic qualities such as taste and flavour. In order to obtain better organoleptic quality of tender coconut water, design of process, standardisation, and optimisation are important. The rheological properties of tender coconut water are crucial for processing, handling and storage of tender coconut water. It is also helpful in the designing of equipments and process standardisation of tender coconut water and their products in large scale industrial production. Fonton *et al*., (2009) studied the thermophysical properties of coconut water at different temperatures. There is a lack of information on rheological properties of tender coconut water which is a pre-requisite for the processing, design of processing equipments and process optimisation for development of novel tender coconut water products. The present investigation was carried out to study the effect of temperature and total soluble solid content/water activity on rheological properties of green tender coconut water and modelling of these properties.

Material and Methods

Raw material

Green coconuts were procured from local market, Mysore, India. The selected fruits were washed with a brush and rinsed in 100 mg/litre chlorinated water using sodium hypochlorite. A special type of stainless steel knife was used for perforating the fruit mesocarp. The water from individual fruits were collected by sucking using vacuum filtration system and collected in stainless steel vessel. The tender coconut water was filtered through muslin cloth. The colleted water was subjected to concentration process.

Concentration

Tender coconut water was concentrated by vacuum evaporation technique using laboratory rotary vacuum evaporator (Model; Laborata 4001, Heidolph, Germany) at 60^{o}C with reduced pressure. Tender coconut water was concentrated to different concentration levels and subjected to rheological measurements.

Analytical methods

Moisture content

Moisture content of tender coconut water was estimated gravimetrically using vacuum oven method (Ranganna, 1986).

Ash content

The ash content of tender coconut water was measured gravimetrically by drying the tender coconut water taken in silica crucible placed in a hot air oven and ignited on hot plate. The sample was later placed in a muffle furnace set at 550^{o}C for 16 hours. The ash content was calculated by difference in weight and expressed in % (Ranganna, 1986).

Mineral content

Mineral content of tender coconut was estimated by dissolving the ash in 3N HCl and the analysis was carried out by flame atomization technique using atomic absorption spectrometer (Varian, model; Varian spectra A280 FS, Germany) and expressed as mg/100ml.

Titrable acidity

The acidity was determined by titration method with standard 0.01M NaOH solution using phenolphthalein as indicator and expressed as % citric acid (Ranganna, 1986).

Total soluble solids

The total soluble solids was determined using a digital hand held refractometer (Atago co., Ltd., Tokyo, Japan) and the total soluble solid content was expressed as °Brix at 25°C.

Sugars

Reducing sugar and total sugar of tender coconut water were determined colorimetrically using 3,5-dinitro salisalicylic acid reagent and expressed as percentage (Miller, 1959)

pH

The pH of tender coconut water was measured using digital pH meter (Cyber scan, India) at 25^{0}C.

Density

The density of tender coconut water was measured using pycnometer at 25°C, the pycnometer was previously calibrated with distilled water and expressed as kg/m^3

Water activity

The water activity of tender coconut water at different concentrations was measured using digital water activity meter at 25°C (Aqua Lab, model 3T E, Decagon devices, USA). The instrument was calibrated using standard solutions provided by manufacturer.

Rheological measurements

The rheological measurements were carried out using MCR100 controlled stress rheometer (Paar Physica, Anton Paar, Gmbh, Germany) equipped with coaxial cylinders (CC 27) and the radii ratio of coaxial cylinders was 1.08477. The rheometer was equipped with an electric temperature controlled peltier system (TEZ-15P-C) to control the experimental temperature and a circulating water bath was used to maintain constant temperature (Viscotherm VT-2, Paar Physica, Anton Paar Gmbh, Germany). The rheological parameter shear stress (Pa) was measured linearly increasing up to a shear rate of 1000 s^{-1} with 10 minutes duration of time and 30 shear stress - shear rate data points were collected and analyzed using universal software US200 (Paar Physica, Anton Paar Gmbh, Germany).The rheological measurements were carried out at 10, 25, 40, 55, 70 and 85°C temperatures. All the measurements were carried out in triplicate and fresh sample was used in each measurement.

Statistical analysis

The experimental results, data analysis and different mathematical models were fitted using statistical software (Statistica 7.0, Stat Soft Tulsa, USA). The fitting and estimates were calculated at $p \leq 0.05$ significance level. The suitability of the models fitting was evaluated by determining the correlation coefficient (r) and root mean square error percent (rmse %) which was evaluated by the following equation

$$\text{rmse }\% = 100/n\ [\sum(W_{exp} - W_{cal}/W_{exp})^2]^{1/2} \quad \text{----- (2)}$$

where W_{exp} is the experimental value, W_{cal} is the calculated value and n is number of data sets. The suitability of the model was decided based on higher correlation coefficient (r) and low percent root mean square error (rmse %) values and level of significance ($p<0.05$).

Results and Discussions

Physico-chemical characteristics

The physico-chemical characteristics of tender coconut water are shown in Table 1. The moisture content of tender coconut water was 94.57% and soluble solid content was 5.3 °Brix which indicated that solids present in tender coconut water was mainly soluble solids such as sugars. The ash content was found to be 0.385% which indicated that tender coconut water had appreciable amount of minerals. Potassium was the major mineral in tender coconut water followed by sodium and calcium. The physico-chemical characteristics of tender coconut water was highly comparable and within the range as reported in literature (Santoso *et al*., 1996; Sabapathy and Kumar, 1999; Reddy *et al*., 2007; Yong *et al*., 2009; Jayanti *et al*., 2010).

Flow behaviour

Figure 1 shows the rheogram of tender coconut water at 26.2 °Brix at 40°C. The rheogram of tender coconut water showed that there was linear increase in shear stress with respect to increase in shear rate, passing through origin and viscosity-shear rate curves almost parallel to X-axis which indicated that the flow was Newtonian in nature. The Newtonian model was able to describe the relationship between shear stress-shear rate data and the viscosity of tender coconut water could be estimated using Newtonian model

Table 1. Physico-chemical characteristics of tender coconut water

Parameter	Quantity
Moisture (%)	94.57±0.07
Ash (%)	0.385±0.007
Total soluble solids (°Brix)	5.30±0.00
pH	4.913±0.006
Acidity (% citric acid)	0.0864±0.001
Water activity (a_w)	0.982±0.001
Density (kg/m^3)	1018.26±2.84
Reducing sugar (%)	4.69±0.016
Total sugar (%)	5.20±0.014
Minerals (mg/100ml)	
K	299.06±14.32
Na	106.16±4.04
Ca	33.69±2.55
Mg	22.49±3.52
Fe	0.22±0.01
Zn	0.28±0.02
Cu	0.025±0.006

Mean± S D (n=3)

Table 2. Viscosity values of tender coconut water at different total soluble solid content, water activity and temperatures

Total soluble solids (°Brix)	Water activity (a_w)	Temperature (°C)	Viscosity (mPa s)	r
5.3	0.982	10	6.02±0.01	0.9852
		25	5.13±0.01	0.9873
		40	4.61±0.01	0.9876
		55	4.25±0.01	0.9884
		70	4.01±0.00	0.9887
		85	3.80±0.01	0.9901
17.0	0.972	10	7.29±0.00	0.9780
		25	5.95±0.00	0.9856
		40	5.27±0.01	0.9868
		55	4.82±0.01	0.9870
		70	4.47±0.01	0.9876
		85	4.26±0.01	0.9867
26.2	0.959	10	8.28±0.01	0.9764
		25	6.97±0.01	0.9810
		40	5.99±0.01	0.9850
		55	5.40±0.01	0.9865
		70	4.97±0.01	0.9868
		85	4.74±0.01	0.9856
36.4	0.935	10	10.55±0.01	0.9791
		25	8.82±0.01	0.9746
		40	7.70±0.01	0.9779
		55	6.64±0.01	0.9835
		70	5.95±0.01	0.9856
		85	5.75±0.01	0.9850
42.6	0.910	10	14.69±0.01	0.9991
		25	10.27±0.01	0.9761
		40	8.92±0.01	0.9722
		55	7.82±0.01	0.9773
		70	6.88±0.01	0.9821
		85	6.39±0.01	0.9840
52.9	0.870	10	34.88±0.01	0.9997
		25	17.22±0.02	0.9991
		40	11.79±0.02	0.9847
		55	10.24±0.02	0.9726
		70	9.22±0.01	0.9706
		85	8.34±0.01	0.9744

Mean± S D (n=3)

(equation 1). The correlation coefficient of the model were greater than 0.97. The magnitude of viscosity values of tender coconut water and its concentrates at different temperatures are given in Table 2. The viscosity values were in the range 3.80 to 34.88 mPa s depending upon the concentration and temperature studied. The results showed that temperature and

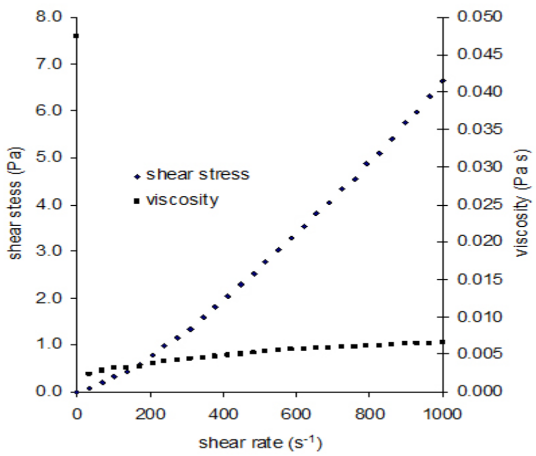

Figure 1. The sample rheogram of tender coconut water at 26.2 °Brix and 40 °C temperature

total soluble solid content/water activity had a significant effect on viscosity of tender coconut water. The magnitude of viscosity of tender coconut water increased significantly ($p<0.05$) with increase in soluble solid content, whereas it was decreased significantly ($p<0.05$) with increase in water activity. The viscosity of tender coconut water decreased significantly ($p<0.05$) with increase in temperature. The concentrations of the soluble solids, insoluble solids and temperature were reported to have a strong effect on the viscosity of Newtonian fluids. The viscosity of tender coconut water strongly depended on inter-molecular forces between molecules and water-solute (sugars and acids) interactions, which result from the strength of hydrogen bonds and inter-molecular spacing as both were strongly dependent on concentration and temperature. An increase soluble solid content leads to increase in hydrated molecules and hydrogen bonding with hydroxyl groups of solute, which would enhance the flow resistance that leads to increase in viscosity of liquid. In case of tender coconut water soluble solids mainly sugar content plays an important role in magnitude of viscosity. The increase in temperature of liquid the magnitude of viscosity significantly decreased, because of increase in thermal energy of the molecules which leads to increase in mobility of molecules and also increases in inter-molecular spacing which decrease the flow resistance (Krokida *et al.*, 2001). Several authors have reported similar type of results for different juices and other products with similar magnitude of viscosity values such as Liquorice extract (Maskan, 1999), cherry juice (Juszczak and Fortuna, 2004), pomegranate juice (Kaya and Sozer, 2005), Pineapple juice (Shamsudin *et al.*, 2007), Orange juice (Ibarz *et al.*, 2009) and Gooseberry juice (Manjunatha *et al.*, 2012).

Table 3. Parameters of Arrhenius equation relating to viscosity and temperature for different total soluble solid content/ water activity

Total soluble solid content (^{o}Brix)	Water activity (a_w)	η_∞ (mPa s)	Activation energy (E_a) (kJ/mol)	r
5.3	0.982	$0.626^{f}\pm0.003$	$5.268^{a}\pm0.012$	0.9884
17.0	0.972	$0.493^{e}\pm0.001$	$6.261^{b}\pm0.007$	0.9847
26.2	0.959	$0.485^{d}\pm0.002$	$6.631^{c}\pm0.009$	0.9922
36.4	0.935	$0.469^{c}\pm0.002$	$7.298^{d}\pm0.014$	0.9952
42.6	0.910	$0.211^{b}\pm0.002$	$9.864^{e}\pm0.020$	0.9765
52.9	0.870	$0.005^{a}\pm0.000$	$20.798^{f}\pm0.010$	0.9607

Mean± S D (n=3), Different superscripts with in a column shows significantly different at p<0.05

Table 4. Parameters of different models relating to flow activation energy (E_a) and total soluble solid content/water activity of tender coconut water (n=3)

Model	a (kJ/mol)	b (Brix^{-1})	c (Brix^{-2})	r	rmse %
E_a=a+bC+cC^2	8.384^{ns}	-0.434^{ns}	0.0121^{ns}	0.9573	9.18
E_a=a (C)^b	0.0430^{ns}	1.520^{ns}	-	0.8115	19.09
E_a=a exp(bC)	2.151^{*}	0.0411^{**}	-	0.9206	11.94
E_a=a exp(bC +cC^2)	6.874^{*}	-0.0338^{ns}	0.00103^{*}	0.9906	4.88
E_a=a + ba_w+ca_w^2	1510.9^{ns}	-3117.2^{*}	1613.9^{*}	0.9925	3.99
E_a=a (a_w)^b	3.593^{**}	-12.363^{***}		0.9807	5.54
E_a=a exp(ba_w)	2561241.4^{ns}	-13.52^{**}	-	0.9777	5.95
E_a=a exp(ba_w+ ca_w^2)	63.06^{ns}	9.667^{*}	-12.643^{ns}	0.9718	6.64

*** p<0.001, ** p<0.01, * p<0.05, ns p>0.05

Effect of temperature

The temperature had a major effect on the Newtonian viscosity similar to the effect on the consistency coefficient for non-Newtonian fluids. The increase in temperature of fluid leads increased in mobility of the molecules and increase in intermolecular spacing, which decreases the flow resistance. The viscosity of tender coconut water decreased markedly with increase in temperature. The variation in viscosity of tender coconut water with temperature was significantly high at higher soluble solid content. The effect of temperature on the viscosity of tender coconut water with different soluble solid contents/water activity was described using the Arrhenius equation

$$\eta = \eta_\infty \exp(E_a/RT) \qquad \text{------- (3)}$$

where η = Viscosity (Pa s), η_∞ = Material constant/ pre-exponential coefficient/frequency factor (Pa s), E_a= Flow activation energy (J/mol), R= Gas constant (J/mol K) and T= Temperature (^{o}K)

Table 3 shows the parameters of Arrhenius equation which was determined by the method of least square approximations. The correlation coefficient was greater than 0.96 and the activation energy was in the range 5.268 to 20.798 kJ/mol depending upon the soluble solid content. The flow activation energy (E_a) was defined as minimum energy required which overcomes the energy barrier before the elementary flow can occur. The viscous flow occurs as a sequence of events which are shift of particles in the direction of shear force action from one equilibrium position to another position by overcoming a potential energy barrier. The barrier height determines the free activation energy of viscous flow. Higher activation energy values indicate a greater influence of temperature on the viscosity, i.e. more rapid change in viscosity with temperature. The magnitude of energy of activation for viscous flow increased significantly (p<0.05) with increase in soluble solid content of the tender coconut water, indicating that higher energy was required to overcome potential energy barrier at higher soluble solids content. Therefore, temperature had a greater effect on viscosity at higher soluble solid contents. When temperature increased, the thermal energy of the molecules and intermolecular spacing increased significantly, which lead to decrease in the magnitude of viscosity (Steffe 1996; Rao 2007). The magnitude of activation energy was in conforming to values reported for other fluid foods (Juszczak and Fortuna, 2004; Altan and Maskan, 2005; Kaya and Sozer, 2005; Juszczak *et al*., 2009; Vandresen *et al*., 2009; Ibarz *et al*., 2009).

Effect of soluble solid content and water activity on flow activation energy

The activation energy for viscous flow of tender coconut water increased significantly (p<0.05) with increase in soluble solid content whereas decreased significantly (p <0.05) with increase in water activity and both trends were non-linear in nature. The variation of activation energy with concentration and water activity could be described by different models, such as

$$E_a = a + bC + cC^2$$
$$E_a = a\,(C)^b$$
$$E_a = a \exp(bC)$$
$$E_a = a \exp(bC + cC^2) \qquad \text{-------- (4)}$$
$$E_a = a + b^* a_w + c^* a_w^2$$
$$E_a = a\,(a_w)^{b^*}$$
$$E_a = a \exp(b^* a_w)$$
$$E_a = a \exp(b^* a_w + c^* a_w^2)$$

where E_a is activation energy (kJ/mol), a is empirical constant (kJ/mol), C is total soluble solid content (^{o}Brix), a_w is the water activity (-), a, b, c, b*, c* are empirical constants.

These models were fitted with activation energy values which were obtained by Arrhenius equation with soluble solid content and water activity by the method of least squares at 5% significant level. The

Table 5a. Parameters of the power law model relating total soluble solid content to viscosity of tender coconut at different temperature

Power law model: η=a $(C)^b$				
Temperature (°C)	a (mPa s)	b ($Brix^{-1}$)	r	rmse%
10	0.00087^a±0.00001	2.659^f±0.005	0.9121	22.52
25	0.4968^b±0.0051	0.8502^e±0.003	0.8667	11.55
40	1.2305^c±0.0034	0.5371^d±0.001	0.9028	7.17
55	1.3133^d±0.0041	0.4829^c±0.001	0.8893	6.74
70	1.3776^e±0.0054	0.4403^b±0.001	0.8696	6.52
85	1.4650^f±0.0073	0.4035^a±0.001	0.8855	5.69

Mean± S D (n=3), Different superscripts with in a column shows significantly different at p<0.05

Table 5b. Parameters of the first order exponential model relating to total soluble solid content to viscosity of tender coconut water at different temperatures

Exponential model: η=a exp(bC)				
Temperature (°C)	a (mPa s)	b ($Brix^{-1}$)	r	rmse%
10	1.522^a±0.009	0.0581^f±0.0001	0.9590	14.36
25	3.217^b±0.009	0.0304^e±0.0001	0.9661	5.62
40	3.590^c±0.003	0.0219^d±0.00003	0.9900	2.50
55	3.341^d±0.005	0.0205^c±0.00004	0.9856	5.93
70	3.146^e±0.006	0.0194^b±0.0001	0.9774	2.88
85	3.107^f±0.008	0.0179^a±0.0001	0.9842	2.21

Mean± S D (n=3), Different superscripts with in a column shows significantly different at p<0.05

Table 5c. Parameters of second order exponential model relating total soluble solid content to viscosity of tender coconut water at different temperatures

Second order exponential model: η=a exp(bC +cC^2)					
Temperature (°C)	a (mPa s)	b ($Brix^{-1}$)	c ($Brix^{-2}$)	r	rmse %
10	7.826^a±0.007	-0.0333^a±0.00009	0.00116^a±0.000002	0.9962	3.98
25	5.482^b±0.008	-0.00663^b±0.00016	0.00053^b±0.000003	0.9970	1.64
40	4.360^c±0.010	-0.00749^c±0.00016	0.000215^c±0.000003	0.9995	0.51
55	4.126^d±0.010	-0.00445^d±0.00024	0.000241^d±0.000004	0.9996	0.35
70	4.009^e±0.007	0.000706^e±0.00006	0.000285^e±0.000004	0.9998	0.33
85	3.725^f±0.004	0.00364^f±0.00015	0.000218^e±0.000003	0.9995	0.35

Mean ± S D (n=3), Different superscripts with in a column shows significantly different at p<0.05

magnitudes of the parameters of above eight models, (equations 4) correlation coefficient (r) and percent root mean square error (rmse%) are reported in table 4. The results indicated that exponential model (r=0.9206, rmse%=11.4, p<0.05) was more effective to describe the influence of total soluble solid content on flow activation energy of tender coconut water. The other models had lower values of correlation coefficient (r), higher root mean square error values and non significant (r<0.99, rmse%>9.1, p>0.05). This indicated that the flow activation energy increased exponentially with total soluble solid content. The effect of water activity on flow activation energy was described by power law equation (r>0.98, rmse %=5.54, p<0.01), where as in the other models the correlation coefficient was lower and the percent root mean square error values are higher. The results showed that the relation between activation energy for viscous flow of tender coconut water with total soluble solid content/water activity was non-linear. The relationship between flow activation energy and total soluble solid content/ water activity were given by

E_a= 2.151*exp (0.0411C), (r>0.92, rmse%= 11.4, p<0.05)
E_a= 33.593 $(a_w)^{-12.363}$ (r>0.98, rmse%=5.54, p<0.01)

where E_a is the flow activation energy (kJ/mol), C is total soluble solid content (°Brix), a_w is the water activity (-). Similar type of exponential equation was reported relating to flow activation energy to total soluble solid content The magnitude of the coefficient concentration was found to be 0.0411 and it was with in the range as reported for different fruit juices such as peach, cherry, pomegranate, pineapple, chokeberry and gooseberry (Ibarz *et al*., 1992a; Giner *et al*., 1996; Altan and Maskan, 2005; Kaya and Sozer, 2005; Shamsudin *et al*., 2007; Juszczak *et al*., 2009; Manjunatha *et al*., 2012). It was reported that flow activation energy increased significantly with square of total soluble solid content for blue berry and raspberry juices, where as it increased quadratically with soluble solid content in case of liquorice (*Glycyrrhiza glabra*) extract (Maskan, 1999; Nindo *et al*., 2005). The deviation in models and model coefficient was due nature solute, size, shape, solute-solvent interactions, hydration state and range of temperature and soluble solid content studied. The flow activation decreased significantly with increased in water activity by power law relation and magnitude of decrease was high, which indicated that flow activation energy sensitive to water activity of tender coconut water.

Effect of total soluble solid content

The concentration of the soluble solids and insoluble solids had strong effect on the viscosity of the Newtonian fluids, where as consistency index and apparent viscosity of non-Newtonian fluids (Krokida *et al*., 2001). The viscosity of a liquid depends on the nature of solvent, nature of solute, their interaction and amount of solid content in solution, solute shape, size and state of hydration. The viscosity of tender coconut water increased significantly with increase in total soluble solid content. The variation in viscosity with soluble solid content was due to variation in degree of hydration of solute molecules, increase in hydrogen bonding with hydroxyl groups of solute and decrease in inter-molecular spacing. The variation of viscosity of tender coconut water with total soluble solid content was non-linear in nature. The different models namely power law and exponential model of different orders were used to investigate the variation in viscosity with soluble solid content at particular temperature used. Several investigators had used these models to investigate the effect of soluble solid content on viscosity of different fluids (Ibarz *et al*., 1989; Ibarz *et al*., 2009).

Power law type; $\eta = a\,(C)b$ (5)

Exponential type; First order $\eta = a \exp(bC)$ (6)

Exponential type; second order $\eta = a \exp(bC + cC^2)$ (7)

where η is the viscosity (mPa s), a is constant (mPa s), b is constant ($Brix^{-1}$), c is a constant ($Brix^{-2}$) and C is total soluble solid content (^{o}Brix).

The parameters of the models were estimated by the method of least squares at 5% significant level. The parameters of variation in viscosity of tender coconut water with soluble solid content by three models namely power law; exponential first order and exponential second order at different temperatures are shown in Table 5a, 5b and 5c respectively. The correlation coefficients were $0.86 \leq r \leq 0.91$, $0.95 \leq r \leq 0.99$ and $0.996 \leq r \leq 0.999$ for power law, exponential and exponential second order respectively. The root mean square error percentage values $5.69 \leq$ rmse % ≤ 22.5, $2.21 \leq$ rmse % ≤ 14.3 and $0.33 \leq$ rmse % ≤ 3.98 for power law, exponential first order and exponential second order models respectively. The parameter 'b' in power law and exponential models decreased significantly ($p \leq 0.05$) with increase in temperature. This indicated that at lower temperatures, the viscosity of tender coconut water increases rapidly when concentration increases, which could be due to change in thermal energy of the molecules and inter-molecular spacing. The exponential type of second order was better to describe the influence of total soluble solid content on viscosity of tender coconut water at different temperatures ($r \geq 0.99$, rmse % ≤ 3.98). The parameters of second order exponential model are shown in table 5c and parameter 'a' decreasing significantly ($p<0.05$) with increasing temperature. The suggested model indicated that the viscosity of tender coconut water was sensitive to total soluble solid content because the parameter 'c' which relates the viscosity quadratically with concentration. The second order exponential model was better to describe the relation between effects of soluble solid content on viscosity of tender coconut water at different temperatures. Similar type of result was reported for clarified pear juice (Ibarz *et a*l., 1989).

Effect of water activity

The water activity of fluid was dependent on amount of solid content, nature of solute, its physico-chemical properties and solute-solvent interactions. The variation in viscosity of tender coconut water to water activity was described by two equations namely power law and exponential type models

Power law; $\eta = a\,(a_w)^b$ (8)

Exponential type; $\eta = a \exp(ba_w)$ (9)

The parameters of the power law and exponential models, correlation coefficient and percent root mean square error were estimated by the method of least squares at 5% significant level and are shown in Tables 6a and 6b respectively. The correlation coefficient was in $0.991 \leq r \leq 0.998$ and $0.990 \leq r \leq 0.998$ for power law and exponential models respectively, where as percent root mean square error values $0.92 \leq$ rmse % ≤ 5.00 and $0.79 \leq$ rmse % ≤ 5.61 for power law and exponential models respectively. The results indicated that both models were suitable for describing the viscosity of tender coconut water with specific water activity. The parameter 'b' was negative which indicated that the viscosity would decrease with increase in water activity as water activity mainly dependent on solid content of the tender coconut water. The water activity of liquid foods is dependent on concentration of the soluble solids, insoluble solids, nature of solute and solute-solvent interactions and state of hydration reported to have a strong non-linear effect on the viscosity of Newtonian fluids (Krokida *et al*., 2001) The magnitude of parameter 'b' of both the models decreased significantly ($p<0.05$) with increase in temperature which indicated that the effect of water activity on viscosity markedly high at lower temperatures. At lower temperatures the change in viscosity of tender coconut water was more rapid compared to that at higher temperatures. The variation in magnitude of coefficient of water activity of power law model was found to be -15.662 to -6.046 whereas exponential model was -17.244 to -6.584 at temperature studied. Similar type of results was reported for other juices. The power law was slightly better to describe the relation between viscosities of orange and blackcurrant juices, the magnitude of coefficient of water activity was in the range -27.75

Table 6a. Parameters of the power law model relating water activity to viscosity of tender coconut water at different temperatures

Power law model: $\eta=a\ (a_w)^b$				
Temperature (°C)	a (mPa s)	b (-)	r	rmse %
10	$3.841^a\pm0.005$	$-15.662^a\pm0.033$	0.9925	5.00
25	$4.442^b\pm0.007$	$-9.640^b\pm0.019$	0.9955	1.71
40	$4.431^b\pm0.002$	$-7.163^c\pm0.013$	0.9917	2.09
55	$4.030^c\pm0.004$	$-6.799^d\pm0.013$	0.9954	1.48
70	$3.724^d\pm0.004$	$-6.542^e\pm0.015$	0.9981	0.92
85	$3.626^e\pm0.007$	$-6.046^f\pm0.020$	0.9940	1.43

Mean ± S D (n=3), Different superscripts with in a column shows significantly different at $p<0.05$

Table 6b. Parameters of the exponential model relating water activity to viscosity of tender coconut water at different temperatures

exponential model: $\eta=a\ \exp(ba_w)$				
Temperature (°C)	a (mPa s)	b (-)	r	rmse %
10	$111639208.2^f\pm1236970.6$	$-17.244^a\pm0.012$	0.9908	5.61
25	$159878.2^e\pm3108.9$	$-10.520^b\pm0.021$	0.9947	1.78
40	$10735.2^d\pm115.0$	$-7.813^c\pm0.011$	0.9935	1.85
55	$6537.4^c\pm86.43$	$-7.411^d\pm0.014$	0.9967	1.26
70	$4539.2^b\pm86.8$	$-7.123^e\pm0.021$	0.9987	0.79
85	$2579.2^a\pm51.1$	$-6.584^f\pm0.022$	0.9951	1.28

Mean ± S D (n=3), Different superscripts with in a column shows significantly different at $p<0.05$

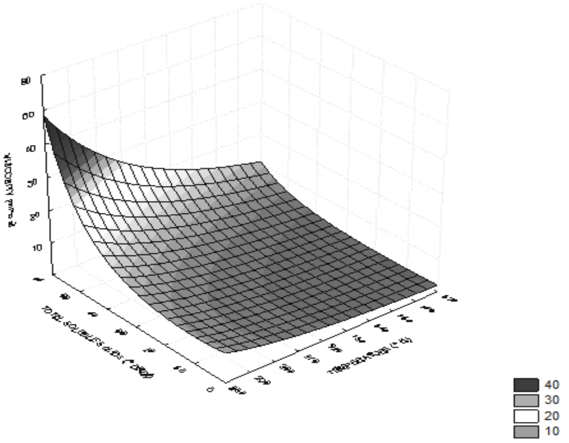

Figure 2. Surface plot for combined effect of total soluble solid content and temperature on viscosity of tender coconut water

to -14.08 at temperature 5 and 75 °C respectively for orange juice and the magnitude was in the range -23. 96 to -13.24 at temperature 5 and 60 °C respectively in case of blackcurrant juice (Ibarz *et al*., 1992; Ibarz *et al*., 1994).

Combined effect of temperature and total soluble solid content

From the food process engineering point of view, it is important to obtain a single equation which describes both temperature and soluble solid content on viscosity of tender coconut water. Several authors had used different equations to describe the combined effect of temperature and soluble solid content on viscosity of the fluids (Giner *et al*., 1996; Ibarz *et al*., 1996; Altan and Maskan, 2005; Kaya and Sozer, 2005: Nindo *et al*., 2005; Juszczak *et al*., 2009; Ibarz *et al*., 2009; Manjunatha *et al*., 2012). The model equations were

Power law type: $\eta = a \exp(E_a/RT)^*(C)^c$ (10)
Exponential type first order: $\eta = a \exp(E_a/RT + cC)$ (11)
Exponential type second order: $\eta = a \exp(E_a/RT + cC + dC^2)$ (12)

Where η is the viscosity (mPa s), a is pre-exponential constant (mPa s), b= E_a/R, E_a is the flow activation energy (J/mol), R is universal gas constant (J/mol K), T is absolute temperature (°K), c is constant (Brix^{-1}), d is constant (Brix^{-2}) and C is total soluble solid content (°Brix).

The values of viscosity shown in table 2 were fitted to these equations by method of least squares using multiple regression analysis. The fits and estimates of the parameters were determined at 5% significance level. The suitability of the model was decided based on correlation coefficient (r) and percent root mean square error (rmse%) values. Table 7a shows the parameters for the different models, correlation coefficients and percent root mean square errors. The correlation coefficients were 0.8426, 0.9074 and 0.9352 for power law, exponential first order and exponential second order models, where as percent root mean square error values 6.80, 4.83 and 3.95 respectively. The second order exponential equation was better to describe the combined effect of temperature and total soluble solid content on viscosity of tender coconut water, because of high correlation coefficient and low percent root mean square error values, where the values of other models were higher compared to second order exponential model. The final equation which represents the combined effect of temperature and total soluble solid content on viscosity tender coconut water was given by

$$\eta=2.007 \times 10^{-2} \exp(1710.5/T - 0.0157C + 0.000728C^2),$$
$(r=0.9352,\ \mathrm{rmse}\%=3.95,\ p<0.05)$

where η is viscosity in m Pa s, T is temperature in °K and C is total soluble solid content in °Brix. Similar type of result was reported for clarified pear juice (Ibarz *et al*., 1989). Several authors reported exponential first order exponential equation to describe the relation between viscosity to total soluble solid content and temperature (Giner *et al*., 1996; Juszczak and Fortuna 2004; Kaya and Sozer 2005; Altan and Maskan 2006; Ibarz *et al*., 2009; Juszczak *et al*., 2009; Juszczak

Table 7a. Parameters of the different models relating to combined effect of temperature and total soluble solid content on viscosity of tender coconut water

Model	a (mPa s)	b=Ea/R	c (Brix^{-1})	d (Brix^{-2})	r	rmse%
η=a exp(Ea/RT)*(C)^c	$6.26 \times 10^{-4*} \pm 4.0 \times 10^{-6}$	$1655.21^{***} \pm 0.42$	$1.188^{***} \pm 0.001$	-	0.8426	6.80
η=a exp(Ea/RT+cC)	$0.0102^{*} \pm 0.00002$	$1677.50^{***} \pm 0.79$	$0.0365^{***} \pm 0.00002$	-	0.9074	4.83
η=a exp(Ea/RT+cC+dC^2)	$0.02007^{*} \pm 0.00004$	$1710.5^{***} \pm 0.33$	$-0.0157^{*} \pm 0.00007$	$0.000728^{***} \pm 0.000001$	0.9352	3.95

Mean± S D (n=3), *** p<0.001, ** p<0.01, * p<0.05

Table 7b. Parameters of the different models relating to combined effect of temperature and water activity on viscosity of tender coconut water

Model	a (mPa s)	b=Ea/R	C (-)	r	rmse%
η=a exp(Ea/RT)*(a_w)^c	$0.0142^{*} \pm 0.00002$	$1705.96^{***} \pm 0.33$	$-11.217^{***} \pm 0.005$	0.9339	3.95
η=a exp(Ea/RT+ca_w)	$2891.7^{*} \pm 12.7$	$1704.20^{***} \pm 0.34$	$-12.256^{***} \pm 0.005$	0.9329	3.97

Mean± S D (n=3), *** p<0.001, ** p<0.01, * p<0.05

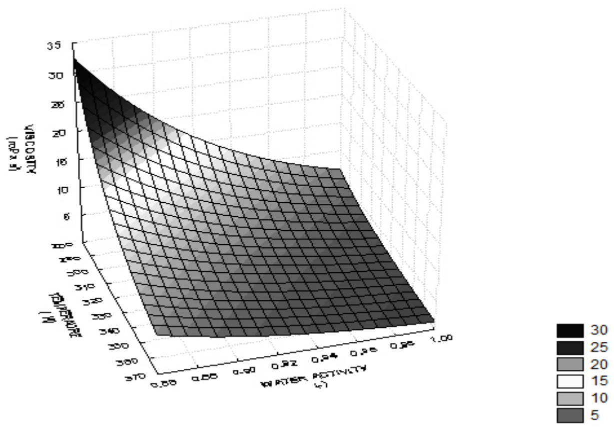

Figure 3. Surface plot for combined effect of water activity and temperature on viscosity of tender coconut water

et al., 2010). The surface plot that described the combined effect of temperature and total soluble solid content on viscosity of tender coconut water at different temperatures and concentrations is shown in Fig 2. The magnitude of viscosity depends on both temperature and total soluble solid content of tender coconut water. At lower temperatures the magnitude of viscosity rapidly increased with soluble solid content and increased marginally at higher temperatures, this was due to increase in thermal energy of the molecules and increase in intermolecular spacing at higher temperatures. Similar type of results was reported for other fluid foods (Ibarz *et al*., 1989; Nindo *et al*., 2005). Altan and Maskan (2005) reported that the viscosity of pomegranate juice was strongly depends on total soluble solid content and temperature irrespective of method of concentration. The viscosity of fluid depends on nature solute, size, shape and state of hydration of the molecules in juice. The solute and solvent interaction was different for different types of solutes (Nindo *et al*., 2005). In case of tender coconut water the soluble solids were mainly sugars, such as glucose, fructose and sucrose and viscosity of tender coconut water dependent on different fractions of sugars present.

Combined effect of temperature and water activity

It was also very important to establish a combined single equation relating temperature and water activity on viscosity of tender coconut water. The two models were used to obtain a single equation for describing the combined effect of temperature and water activity on viscosity of tender coconut water. Generally, power law and exponential type equation were used to describe the combined effect of temperature and water activity on viscosity of juices.

Power law model: $\eta = a \exp(E_a/RT)*(a_w)^c$ (13)

Exponential model: $\eta = a \exp(E_a/RT + ca_w)$ (14)

where η is the viscosity (m Pa s), a is pre-exponential constant (m Pa s), b=Ea/R, E_a is flow activation energy (J/mol), R is the universal gas constant, T is absolute temperature (°K), aw is the water activity (-) and c is constant (-).

The viscosity of tender coconut water at different temperature and water activity in Table 2 were fitted using multiple regression analysis by method of least squares at 5% significant level. The parameters of combined effect of temperature and water activity were given in Table 7b. The correlation coefficients were 0.9339 and 0.9329 for power law and exponential models where as percent root mean square error was 3.95 and 3.97 respectively. Both the models were able to explain the combined effect of temperature and water activity on tender coconut water, since the correlation coefficients and root mean square value were almost similar magnitude. The parameter 'c' of the model was negative, which indicated that viscosity of tender coconut water decreased with increase in water activity. The water activity of fluid mainly depends on nature of solute and its concentration. The combined equations which related to temperature and water activity are given by

$\eta = 1.42 \times 10^{-2} (a_w)^{-11.217} \exp(1705.96/T)$, ($r > 0.93$, rmse%=3.95, $p < 0.05$)

$\eta = 2891.69* \exp(1704.2/T - 12.256a_w)$, ($r > 0.93$, rmse%=3.97, $p < 0.05$)

Figure 3 shows the surface plot for the combined effect of temperature and water activity on viscosity of tender coconut water. The magnitude of viscosity of tender coconut water increased rapidly at lower water activities whereas increased marginally at higher water activity levels. This indicated that both temperature and water activity had significant effect on viscosity of tender coconut water and at higher temperatures the mobility of molecules was higher due to its higher kinetic energy and also increase in inter-molecular spacing. Similar type of results was reported for different fruit juices (Ibarz et al., 1992b; Ibarz *et al*., 1994). These results were very useful in processing, designing of equipments and upscaling of process of tender coconut water and their concentrates in large scale commercial production.

Conclusions

The rheological behaviour of tender coconut (*Cocos nucifera* L) water was studied as a function of total soluble solid (TSS) content (5.3 to 52.9 °Brix), corresponding water activity (a_w) (0.982 to 0.870) at a wide range of temperature (10 to 85°C). The results indicated that the tender coconut water and its concentrate behaved like a Newtonian fluid and the Newtonian viscosity (η) was in the range 3.80 to 34.88 mPa s depending upon the concentration and temperature studied. The Arrhenius equation was able to describe temperature dependency on viscosity of tender coconut water (r > 0.96) and the energy of activation (E_a) for viscous flow was in the range 5.268 to 20.798 kJ/mol depending upon the total soluble solid content. The effect of total soluble solid content on flow activation energy was described by exponential equation (r>0.92. rmse %=11.4, p<0.05) and that of water activity was described by power law equation (r>0.98, rmse %=5.54, p<0.01). The effect of total soluble solid content on viscosity of tender coconut water followed second order polynomial exponential equation (r >0.99, rmse%< 3.98) at the temperature used., where as, the effect of water activity on viscosity was described by both power law as well as exponential type relationship (r>0.99). The combined effect of temperature and total soluble solid content/water activity on viscosity of tender coconut water was described by the equations

$\eta = 2.007 \times 10^{-2} \exp(1710.5/T - 0.01567C + 0.000728C^2)$, (r >0.93, rmse%=3.95)

$\eta = 1.42 \times 10^{-2} (a_w)^{-11.217} \exp(1705.96/T)$, (r >0.93, rmse%=3.95)

$\eta = 2891.69^* \exp(1704.2/T - 12.256a_w)$, (r>0.93, rmse%=3.97)

where η is viscosity in mPa s, T is temperature in °K and C is the total soluble solid content in °Brix, a_w is the water activity (-).

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