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# Colour kinetics and rheology of coriander leaf puree and storage characteristics of the paste

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#### Abstract

Coriander leaves have been used as a food flavourant in various cuisines since ancient times. The colour degradation kinetics of coriander leaf puree was studied using a fraction conversion technique during thermal treatment at 50, 65, 80, 95 and 110  $\degree$ C for up to 60 min. Blanched, comminuted coriander leaf puree was subjected to heat treatment at selected temperatures in an oil bath with agitation. Treated samples were removed from the bath at selected time intervals (0–60 min after come-up period), cooled immediately and analyzed for colour using a Hunterlab colourimeter. The rheological characteristics of the puree were evaluated using a computer-controlled Haake rotational viscometer at 50, 60, 70 and 80  $^{\circ}$ C and it was found that the Herschel–Bulkley model adequately represented shear stress-shear rate data. Temperature dependency of the consistency index and apparent viscosity at a shear rate of 100 s<sup>-1</sup> followed the Arrhenius relationship and the flow activation energy ranged between 17.2 and 17.9 kJ/mol. Coriander puree was converted to paste by adding common salt (2%) and the required volume of citric acid to adjust the pH to 4.2. The paste was then filled into glass bottles and stored at selected temperatures (5, 25 and 37  $^{\circ}$ C) for 6 months and was periodically evaluated for colour, total aerobic plate count and physicochemical properties. Coriander puree/paste colour was expressed in terms of tristimulus colour value a and combination  $(L \times a \times b)$ . First order reaction kinetics adequately described the changes in colour values during both thermal treatment of puree and storage of paste. The process activation energies were 29.3 and 22.1 kJ/mol, respectively, for a and  $(L \times a \times b)$  during thermal treatment. The corresponding values during storage were 12.7 and 12.2 kJ/mol. The paste was microbiologically stable with no significant changes  $(P>0.05)$  in physicochemical characteristics during storage.  $\odot$  2003 Elsevier Ltd. All rights reserved.

Keywords: Coriander leaves puree and paste; Kinetic model; Colour; Flow behavior; Storage; Activation energy

# 1. Introduction

Coriander (Coriandrum sativum L.) is an umbelliferous annual plant of the parsley family, native to the east Mediterranean region and southern Europe. It is grown in Romania, Mexico, Argentina, Egypt, India, the United States, China, Russia and many other countries. The herb is used as a valuable spice for its exotic flavour, stimulant and carminative properties while fluid-extract (and oil) is used medicinally as an antispasmodic, for rheumatism and as a tonic. The plant produces a slender, hollow stem 30–60 mm high with bipinnate leaves

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and small pink flowers. It is valued for the fruits, called coriander seeds and the fresh green leaves called cilantro. The delicate young leaves are widely used in Latin-American, Indian and Chinese dishes to impart flavour. The fresh coriander leaves contain 2-decenal and 2 dodecenal.

Fresh coriander leaves are perishable in nature and require immediate processing or preservation. Drying is the major processing technology practised for coriander leaves so far. However, drying has a limitation on aromatic herbs as it results in considerable losses of flavonoid components and attractive colour pigments [\(Baranowski, 1985; Lee et al., 1992; Pezzutti & Crapiste,](#page-6-0) [1997\)](#page-6-0). Therefore, there is a need for alternate processes for shelf-life extension and for developing other coriander leaf-based products. Puree and pastes are such

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## Nomenclature



products that could retain colour and flavour in a semisolid form having qualities close to the fresh ones and they are convenient to use. Recently the market for various spices purees/pastes has been increased considerably due to their demand in fast food industries.

The kinetics of food colour degradation are complex. Dependable models that accurately predict the progress of a chemical reaction occurring in a homogeneous liquid or semi-solid phase during thermal processing and storage are useful in many engineering applications, including process optimization. Therefore experimental studies and application of simplified models to predict and interpret kinetic parameters (reaction order, rate constant, and activation energy) are needed.

Several colour scales have been used to describe colour. Those most often used in the food processing industries are the Hunter colour  $L$ ,  $a$ ,  $b$  system and CIE LAB colour scales ([Giese, 2000\)](#page-6-0). The kinetic parameters for a reaction should be independent of the process and instrument used. Several researchers have advocated the use of tristimulus colourimetry over spectrophotometric measurement of pigment to represent the colour changes during thermal processing ([Ahmed, Kaur, & Shiv](#page-5-0)[hare, 2002a; Gupte, El-Bisi, & Francis, 1964; Kajuna,](#page-5-0) [Bilanski, & Mittal, 1998; Rocha, Lebert, and Marty-](#page-5-0)[Audouin, 1993; Shin & Bhowmik, 1994; Steet & Tong,](#page-5-0)

[1996; Weemaes, Ooms, Leoy, and Hendrickx, 1999\)](#page-5-0). No information is, however, available on green and total colour degradation of coriander leaf puree/paste in terms of tristimulus colourimetry.

Knowledge of the rheological properties of food products is essential for product development, design and evaluation of process equipment such as pumps, piping, heat exchangers, evaporators, sterilizers and mixers. Rheological measurements have also been considered as an analytical tool to provide fundamental insight of the structural organization of food. Various factors affecting the rheological behaviour of fruit puree and concentrates include temperature ([Holdsworth, 1971;](#page-6-0) [Vitali & Rao, 1984](#page-6-0)), total soluble solids/concentration [\(Hernandez, Chen, Johnson, & Carted, 1995; Ilicali,](#page-6-0) [1985; Rao, 1977](#page-6-0)), particle size [\(Ahmed, Shivhare, &](#page-6-0) [Raghavan, 2000; Tanglertpaibul & Rao, 1987](#page-6-0)). It has been reported that fruit purees behave as non-Newtonian fluid ([Holdsworth, 1971](#page-6-0)) as a result of complex interactions among soluble sugars, pectic substances and suspended solids.

The objectives of this study were to evaluate: (1) the degradation kinetics of visual colour during thermal treatment of coriander leaves puree, (2) the rheological characteristics of puree, and (3) the storage stability of the paste.

# 2. Materials and methods

# 2.1. Raw material

Fresh coriander leaves were procured from a local firm of Amritsar in the state of Punjab, India.

#### 2.2. Preparation of puree

Coriander leaves were separated from stems, washed in running tap water and blanched for 2 min in hot water at 90 °C. The blanching time was based on preliminary trials that resulted in complete inactivation of peroxidase enzyme in coriander leaves. Coriander leaves were cooled immediately in chilled water, drained and comminuted in a laboratory size grinder fitted with a 14 mesh (1.19 mm aperture size) screen to obtain a puree of uniform size. The puree was stored at  $0^{\circ}$ C, immediately, until further use.

# 2.3. Thermal treatment

Thermal degradation kinetics of coriander leaf puree was studied by isothermal heating at selected temperatures (50, 65, 80, 95 and 110  $^{\circ}$ C) for a residence time of 0 to 60 min. The samples were sealed in vials (i.d. 1 cm, 8 ml) and immersed in thermostatted oil-bath for preset times (0, 15, 30, 45 and 60 min) following the

<span id="page-2-0"></span>method described by [Weemaes et al. \(1999\) and Ahmed](#page-6-0) [et al. \(2002a\)](#page-6-0). The desired temperature was considered to have been achieved when the correct temperature of the oil bath was reached. An oil-recirculating pump was used to facilitate rapid heat transfer. The samples were transferred to an ice water bath immediately after the thermal treatment.

#### 2.4. Preparation of paste

Total soluble solids and pH of coriander puree were 8.6 Brix and 6.21 respectively. Paste is characterized as the product obtained after addition of common salt and organic acid to the puree (Ahmed & Shivhare, 2001). The puree was converted to paste by addition of 2% sodium chloride (w/w) and the required volume of  $30\%$  $(w/v)$  citric acid to adjust the pH to approximately 4.2. Salt level was selected by a preliminary sensory test. Salt increased total soluble solids of puree. An acidified food  $(pH < 4.5)$  requires only pasteurization for shelf-stability and therefore the paste was heat-treated at 80  $\degree$ C for 15 min and immediately hot-filled into pre-sterilized glass bottles. The bottles were cooled by spraying chilled water and stored at selected temperatures (5, 25 and  $37 \pm 1$  °C) for 6 months. The samples were analyzed at intervals of 1 month for colour, TSS, pH and salt content.

## 2.5. Measurement of visual colour

Visual colour was measured with a Hunter colourimeter model D25 optical sensor (Hunter Associates Laboratory, Reston, VA) in terms of  $L$  (lightness),  $a$ (redness and greenness) and  $b$  (yellowness and blueness). The instrument  $(45^{\circ}/0^{\circ}$  geometry,  $10^{\circ}$  observer) was calibrated with a standard white tile  $(L=90.55,$  $a = -0.71$ ,  $b = 0.39$ ). A glass Petri dish containing the heat-treated puree was placed above the light source and covered with a white plate and post-process Hunter L, a, b values were recorded.

## 2.6. Kinetics model

Degradation of visual colour has been found to follow first order reaction kinetics [\(Ahmed et al., 2002a;](#page-5-0) [Gunawan, & Barringer, 2000; Hutchings, 1994; Shin &](#page-5-0) [Bhowmik 1994; Steet & Tong 1996; Weemaes et al.,](#page-5-0) [1999\)](#page-5-0). Following these reports, it was reasoned that the thermal degradation of colour of coriander leaf puree follows the first order kinetic model.

$$
\ln(C/C_o) = -k.t
$$
 (1)

Fractional conversion is a convenient variable, often used in place of concentration ([Levenspiel, 1974\)](#page-6-0), and has been reported to increase the accuracy of the calculation [\(Gunawan & Barringer, 2000](#page-6-0)). In reaction kinetics, it is important to know the fraction of reactants that has been converted to product at any time, t. For irreversible first order reaction kinetics, the rate constant at constant temperature can be determined through fractional conversion, f:

$$
f = (C_0 - C)/(C_0 - C_{\infty})
$$
\n<sup>(2)</sup>

First order reaction in terms of the fractional conversion may be represented as

$$
\ln(1 - f) = -k. \tag{3}
$$

Dependence of the degradation rate constant on temperature is represented by the Arrhenius equation:

$$
k = k_0 \exp(-E/RT) \tag{4}
$$

The infinite value of visual colour was determined by the methods of [Steet and Tong \(1996\), Weemaes et al.](#page-6-0) [\(1999\) and Ahmed et al. \(2002b\)](#page-6-0). Coriander puree was acidified using concentrated hydrochloric acid and thermally treated at selected temperatures for 24 h and the corresponding Hunter colour values were measured. It was found that the colour degradation at a constant temperature was nearly constant with respect to time on prolonged heating. This non-zero residue colour/pigment should be independent of reaction temperature and reaction path [\(Steet & Tong, 1996\)](#page-6-0).

## 2.7. Rheological measurement

A rotational viscometer (Haake Model RV20; Haake Mess-Technik, Karlsrulhe, Germany), equipped with an M-05 OSC measuring head and MV1 rotor (o.d. 20.04 mm and height 60 mm) in a concentric cylindrical cup (i.d. 21 mm) assembly, interfaced to a microcomputer for control and data acquisition, was used for rheological measurement. The sample compartment was maintained at a constant temperature using a water bath/ circulator (Haake, Model FK-2).

For each test, the filled sample cup and spindle were temperature-equilibrated for about 15 min and the samples were subjected to three-cycle shear changes from 0 to 300 s<sup>-1</sup> over 5 min, steady shear at 300 s<sup>-1</sup> over 5 min followed by back to  $0 s<sup>-1</sup>$  in the next 5 min. In order to perform a quantitative comparison of coriander puree sample various rheological models, based on shear stress-shear rate, were tested (Newtonian, power law, Casson, Bingham and Herschel Bulkley model) using software (Haake RV20 version 2.3) and the best fit model was selected on the basis of standard error.

#### 2.8. Physicochemical properties

Total soluble solids (TSS) ( $\degree$ Brix) and pH were measured with a refractrometer (Atago, Japan) and a pH meter (Systronics, India) with glass electrode at  $25 \text{ }^{\circ}\text{C}$ , <span id="page-3-0"></span>respectively. Sodium chloride was determined by titration with silver nitrate ([Ranganna, 1986](#page-6-0)).

# 2.9. Microbiological studies

Paste samples were analyzed for standard plate counts (SPC), coliforms, lactobacillus spp. and yeast and mold counts, following the procedure of the International Commission on Microbiological Specifications [\(ICMSF, 1992\)](#page-6-0).

#### 2.10. Statistical analysis

All tests were carried out in duplicate and mean values reported. Statistical analysis was carried out by the method described by [Gacula and Singh \(1994\)](#page-6-0). Trends were considered significant when means of compared sets differed at  $P < 0.05$ .

### 3. Results and discussion

# 3.1. Kinetics of thermal degradation

The Hunter colour values of fresh coriander puree were  $L = 16.57$ ,  $a = -3.65$  and  $b = 7.94$ . The tristimulus colour values decreased during thermal treatment and the corresponding post-process  $L$ ,  $a$  and  $b$  values ranged from 15.10 to 15.64,  $-1.03$  to  $-2.63$  and 6.05 to 7.23, respectively, after 1 h. The equilibrium Hunter  $L_{\infty}$ ,  $a_{\infty}$ and  $b_{\infty}$  values were 6.45,  $-0.29$  and 2.75, respectively, when the puree was acidified in concentrated hydrochloric acid and thermally processed for a prolonged time (24 h).

Since the major colour of coriander puree is green, Hunter  $-a$  value was considered as the physical parameters to describe the visual colour degradation during thermal processing. Several researchers [\(Ahmed et al.,](#page-5-0) [2002a,2002b; Steet & Tong, 1996](#page-5-0)) have used the Hunter colour-a value while studying the colour degradation kinetics of green vegetables. [Eq. \(3\)](#page-2-0) can be written for Hunter-a value using fraction conversion technique as:

$$
\ln[(-a) - (-a_{\infty})]/[(-a_{\infty}) - (-a_{\infty})] = -k \tag{5}
$$

The rate of degradation of colour of coriander leaf puree was determined by linear regression [Eq. (5)] and the coefficients are reported in Table 1. The  $R^2$  values were greater than 0.959 while the standard error were less than  $0.0002$  for the entire range. Solid lines in [Fig. 1](#page-4-0) represent the model values. It is obvious from [Fig. 1](#page-4-0) that degradation of visual colour represented by  $-a$ value of coriander leaf puree, followed first-order reaction kinetics.

In practice, any change in  $a$  value is associated with simultaneous change in  $L$  and  $b$  values. Representation of quality in terms of total colour may therefore be more relevant from the processing viewpoint [\(Ahmed et](#page-5-0) [al., 2002a; Shin & Bhowmik, 1994\)](#page-5-0). In the present study, both  $L$  and  $b$  decreased with decrease in  $-a$ value. Therefore the combination  $(L \times a \times b)$  was selected to represent the colour change.

The Eq. (5) takes the following form in terms of  $(L \times a \times b)$  colour combination:

$$
\ln\left[\left(L\times a\times b\right) - \left(L_{\infty}\times a_{\infty}\times b_{\infty}\right)\right]/\left[L_{0}\times a_{\infty}\times b_{\infty}\right) - \left(L_{\infty}\times a_{\infty}\times b_{\infty}\right)\right] = -k.t
$$
 (6)

The rate of degradation of total colour was determined by linear regression [Eq. (6)] (Table 1). The  $R^2$  values were greater than 0.955 while the standard error were less than 0.0005 for the entire range. It is obvious from [Fig. 2](#page-4-0) that degradation of visual total colour as represented by  $(L \times a \times b)$  of coriander leaf puree, followed first-order reaction kinetics.

Effect of temperature on the degradation rate constants, is shown in [Fig. 3](#page-4-0). Results indicated that the dependence of rate constants for both  $-a$  and  $(L \times a \times b)$ on temperature followed the Arrhenius relationship  $(R<sup>2</sup> > 0.96)$ . The activation energies for degradation of  $-a$  and  $(L \times a \times b)$  were 29.30 and 22.13 kJ/mol respectively. High activation energy signifies greater heat sensitivity of visual colour degradation during thermal

Table 1

Kinetic parameters for colour change during thermal processing (50–110 °C for 0–60 min) and storage (5, 25 and 37 °C for 0–6 months)

Temperature $(^{\circ}C)$	Eq. $(5)$			Eq. $(6)$			
	$k \, (min^{-1})$	$R^2$	S.E.	$k \, (min^{-1})$	$R^2$	S.E.	
50	0.0024	0.973	0.0001	0.0036	0.955	0.0002	
65	0.0035	0.959	0.0002	0.0047	0.969	0.0002	
80	0.0054	0.997	0.0001	0.0071	0.998	0.0001	
95	0.0069	0.972	0.0004	0.0085	0.990	0.0003	
110	0.0145	0.973	0.0007	0.0134	0.983	0.0005	
5	$2.5 \times 10^{-6}$	0.991	$5.44 \times 10^{-8}$	$2.62\times10^{-7}$	0.996	$3.93 \times 10^{-8}$	
25	$3.7 \times 10^{-6}$	0.914	$2.2 \times 10^{-7}$	$3.75 \times 10^{-6}$	0.932	$2.01 \times 10^{-7}$	
37	$4.5 \times 10^{-6}$	0.932	$2.4 \times 10^{-7}$	$4.5 \times 10^{-6}$	0.976	$2.57 \times 10^{-7}$	

<span id="page-4-0"></span>treatment. It may therefore be inferred that the Hunter colour  $-a$  value should preferably be used to represent colour degradation during thermal processing of coriander leaf puree.

# 3.2. Rheological characteristics of coriander leaf puree

Coriander leaf puree exhibited yield stress. Similar observations for puree foods have been reported by [Ahmed and Ramaswamy \(2003\)](#page-5-0). The magnitude for the yield stress depends on the evaluation technique and the



Fig. 1. First-order colour (Hunter  $-a$  value) degradation kinetics of coriander leaf puree at selected temperatures.



Fig. 2. First-order total colour (Hunter  $L \times a \times b$ ) degradation kinetics of coriander leaf puree at selected temperatures.



Fig. 3. Variation of colour degradation rate constants with temperature. Fig. 4. Rheogram for coriander leaf puree.

assumptions used during the measurement ([Steffe,](#page-6-0) [1992\)](#page-6-0). For coriander puree, the yield stress varied between 16.54 and 23.72 Pa and the variation was not systematic with temperature. Yield stress values were obtained directly from the software and verified from the Casson model [Eq. (7)] using the relationship  $K_0^2 = \tau_0$ .

Shear stress-shear rate data fitted the Herschel–Bulkley model adequately [Eq. (8)]. In all cases  $R^2$  values were greater than 0.87 and standard errors were less than 0.039.

$$
\tau^{0.5} = K_0 + K_c \; (\gamma)^{0.5} \tag{7}
$$

$$
\tau = \tau_0 + K\gamma^n \tag{8}
$$

Typical flow curves are shown in Fig. 4. No significant changes were observed in up and down curves and, hence, average values of rheological parameters were considered. The puree did not exhibit time-dependency during shearing at  $300s^{-1}$  for 5 min. The flow behavior index of puree varied between 0.61 and 0.70 and it behaved as a pseudoplastic fluid. Similar observations for various purees have been reported by various researchers [\(Duran & Costell, 1982; Ibarz et al., 1995;](#page-6-0) [Bhattacharya & Rostogi, 1998](#page-6-0)).

The consistency index decreased with increase of temperature. Decrease in K with increased temperature has been commonly reported by researchers [\(Hernandez](#page-6-0) [et al., 1995; Rao, 1977; Vitali & Rao 1984\)](#page-6-0). Dependencies of consistency index and apparent viscosity on process temperatures were adequately described by the Arrhenius equation [Eqs. (9) and (10)] ([Fig. 5\)](#page-5-0).

$$
K = A_k \exp(E_k / RT) \tag{9}
$$

$$
\eta = A_{\eta} \exp(E_{\text{n}}/RT) \tag{10}
$$

The  $R<sup>2</sup>$  values were greater than 0.943. The flow activation energies equalled 17.2and 17.9 kJ/mol for consistency index and apparent viscosity, respectively. These values are in agreement with those previously



<span id="page-5-0"></span>reported ([Ahmed et al., 2002b; Manohar et al., 1991;](#page-6-0) [Vitali & Rao, 1984\)](#page-6-0). The values of  $A_k$  and  $A_n$  were  $6.44 \times 10^{-4}$  Pa.s<sup>n</sup> and  $2.08 \times 10^{-4}$  Pa.s, respectively.

# 3.3. Effect of storage on physico-chemical characteristics of paste during storage

TSS, pH and sodium chloride did not change  $(P>0.05)$  during storage. Colour of paste was affected  $(P \le 0.05)$  by both storage temperature and time periods [\(Table 1,](#page-3-0) [Fig. 3](#page-4-0)). Predictably, the change in colour was minimum at  $5 \degree C$ , while that stored at higher temperatures (25 and 37  $\degree$ C) showed faster colour degradation due to heat-induced browning. The decrease in colour values during storage is also likely due to oxidation of pigments.

The fractional conversion technique was applied to represent colour degradation of paste during storage, using infinite values, as mentioned earlier for puree. The [Eqs. \(5\) and \(6\)](#page-3-0) represented degradation of colour of paste during storage period. The colour degradation rate constants followed the Arrhenius relationship  $(R^2 = 0.98)$  (shown in [Fig. 3](#page-4-0)). The process activation energies for colour degradation during storage were 12.7 and 12.2 kJ/mol, respectively, for a and  $L \times a \times b$ . Expectedly, these values are significantly lower  $(P<0.05)$  than the range for colour degradation of puree during thermal processing at  $50-110$  °C, as previously discussed. This implies that the colour degrada-



Fig. 5. Variation of the consistency index and apparent viscosity at  $100 s^{-1}$  with temperature.

Table 2 Microbiological data on coriander leaf paste during 6 months of storage

Parameter	$5^{\circ}C$			$25 \text{ °C}$			$37^{\circ}$ C		
Months								2 4 6 2 4 6 2 4 6	
TPC $(CFU/g)$ 300 400 600 450 600 750 700 900 950									

tion of puree and paste followed first order reaction kinetics, irrespective of addition of salt or acid and over a wide temperature range (5–110 °C). Further, higher E values indicate sensitivity of colour during thermal treatment (50–110  $\degree$ C) compared to that at storage temperatures.

# 3.4. Microbiological qualities of paste during storage

The total plate counts (TPC) and lactobacillus counts of the paste before thermal treatment were  $3\times10^4$  and  $3.7 \times 10^{3}/g$ , respectively, while the coliform and yeast and mold counts were below 10 and 100, respectively. Thermal processing of paste reduced TPC to 100 while lactobacillus, coliforms and yeast and molds were found to be negative. The microbiological counts of the paste during storage are listed in Table 2. The TPC values increased from 100 to 950 CFU/g, while coliforms were absent and lactobacillus and yeast and mold counts increased from nil to less than 100 during 6 months of storage. The presence of bacteria of public health significance was below the prescribed limit [\(Pimm, 1994\)](#page-6-0). It can therefore be concluded that prepared paste stored up to 6 months was microbiologically safe.

## 4. Conclusions

In the present study, colour degradation kinetics of coriander leaf puree were studied using the fractional conversion technique. It was found that the degradation of colour of coriander leaf puree, during thermal processing and storage of paste at selected temperatures followed first-order reaction kinetics. The process activation energy ranged between 12.2 and 29.3 kJ/mol. The puree showed pseudoplastic behaviour with yield stress and the Herschel–Bulkley model adequately described the flow behaviour. The flow activation energy was found to be 17.2–17.9 kJ/mol. Data reported in the experiment should be useful for the coriander leaf puree/paste processing industry and it was revealed that both Hunter  $-a$  and  $(L \times a \times b)$  could be used as colour degradation parameter for on-line quality measurement. The paste processed at 80  $\degree$ C for 30 min was shelf stable with insignificant  $(P>0.05)$  change in physicochemical properties during storage and was microbiologically safe for human consumption.

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