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RESEARCH ARTICLE

MODELLING THERMAL DEGRADATION KINETICS OF COLOUR IN CURRY LEAF PASTE

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ABSTRACT

Curry leaf, has been used as a food flavourant in Asian-Indian cuisine since ancient times. The colour degradation kinetics of curry leaf paste was studied using fraction conversion technique during thermal treatment at 50, 65, 80 and 95 °C for up to 60 min. Blanched, comminuted curry leaf paste was subjected to heat treatment at selected temperatures in awater 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. Curry leaf paste colour was expressed in terms of tristimulus colour value *a* and combination (*L,a,b*). First order reaction kinetics adequately described the changes in colour values during thermal treatment of puree. The process activation energies were 29.3 and 22.1 kJ/mol, respectively, for *a* and (*L,a,b*) during thermal treatment. The *Lab* and TCD translate the real behavior of heat treated curry leaf puree. The retention of total colour of curry leaf paste may be used as a quality indicator for thermal processing conditions.

INTRODUCTION

Curry leaf (*Murraya koenigii*), is a popular leafy-spice native to India, which has been spread by Indians who have migrated around the world. It is used by Asian Americans originating from South Asia almost daily in its fresh form when available and is preserved as dried or frozen for long-term storage. Parts of the plant have been used as raw material for traditional medicine formulation in India to treat various ailments (Khan *et al.,* 1997). *M. koenigii* leaves and roots can be used to cure piles and allay heat of the body, thirst, inflammation and itching. The aromatic leaves, which retains their flavour and other qualities even after drying, are slightly bitter, acrid, cooling, weakly acidic in tastes and are considered as tonic, anthelminthic, analgesic, digestive, appetizing agents (Khanum *et al.,* 2000). They are widely used in Asian-Indian cuisine, in India they are fried in combination with other spices and used in a wide range of curries giving a delicious flavour and aroma and have created a ready market and greater demand for this spice (Palaniswamy 2001). Thermal process optimization relies on degradation kinetic models for food safety and quality. Therefore, the objective of this research study was the mathematical modelling kinetics of thermal degradation of colour in curry leaf paste, in order to apply it as an indicator for sterilisation thermal processing conditions impact on quality.

MATERIALS AND METHODS

Raw material: Fresh curry leaves were procured from a local firm (Nilgris) of Thanjavur in the state of Tamil Nadu, India.

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Preparation of puree

Curry leaves were separated from stems, washed in running tap water and blanched for 2min in hot water at 90°C. The blanching time was based on preliminary trials that resulted in complete inactivation of peroxidase enzyme in curry leaves. Curry 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°C, immediately, until further use.

Thermal treatment

Thermal degradation kinetics of curry leaf puree was studied by isothermal heating at selected temperatures (50, 65, 80, 95 and 110°C) for a residence time of 0 to 60 min. The samples (50g each) were sealed in vials and immersed in thermos tatted water-bath for times (0, 15, 30, 45 and 60 min) following the method described by Weemaes *et al.,* (1999) and Ahmed *et al.,* (2002a). The desired temperature was considered to have been achieved when the correct temperature of the water bath was reached. The samples were transferred to an ice water bath immediately after the thermal treatment.

Measurement of visual colour

Visual colour was measured with a Hunter colour lab 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 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.

Nomenclature	
C	measured Hunter colour value(s) (L, a, b) or a combination of these at time t (dimensionless)
C_o	measured colour value at zero time (dimensionless)
C_∞	measured colour value(s) at infinite time (dimensionless)
E_a	activation energy for colour degradation (kJ/mol)
k	rate constant (min^{-1})
k_o	frequency factor (min^{-1})
R	universal gas constant [8.314 J/(mol K)]
T	absolute temperature (K)
t	heating time (min)

Kinetics model considerations

A number of research studies apply zero- (Eq. (1)) or first-order (Eq. (2)) models to describe the thermal degradation of colour in food products:

$$C = C_o - kt \text{ ,} \quad (1)$$

$$C = C_o \exp(-kt) \quad (2)$$

The fractional conversion model can also be used to describe the thermal colour degradation kinetics (Levenspiel, 1972). It is a convenient variable, often used in place of concentration (Levenspiel, 1974), and has been reported to increase the accuracy of the calculation (Gunawan & Barringer, 2000). 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,

$$f = [(C_o - C)/ (C_o - C_\infty)] \quad (3)$$

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

$$ln(1-f)= -k .t \quad (4)$$

Colour concentration can be translated using the *L*, *a* and *b* parameters of the colour system, or by a combination of these three values, such as *La/b* and *TCD*:

$$TCD=\sqrt{(\Delta L^2 + \Delta b^2 + \Delta a^2)}. \quad (5)$$

La/b and TCD express a total colour change of the food product (Shin & Bhowmik, 1995). The Arrhenius equation is usually applied to evaluate the dependence of the degradation rate constant on temperature:

$$k = k_o \exp (-E_a/RT) \quad (6)$$

The infinite value of visual colour (C_∞) was determined by the methods of Steet and Tong (1996), Weemaes *et al.,* (1999) and Ahmed *et al.,* (2002b). Curry leaf paste 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).

Statistical Analysis

Experimental data for the different parameters were fitted to kinetic models and processed by using SPSS version 13.0 software. Correlation coefficient value was used as the basis to select the best fitting for estimation of the parameters of the models. Statistical analysis was carried out by the method described by Gacula and Singh (1994). When comparing means, trends were considered significant at $P < 0.05$.

RESULTS AND DISCUSSION

Kinetics of thermal degradation

The hunter colour values of fresh curry leaf paste were *L*=30.21, *a*=- 1.45 and *b*=7.97. As heating temperature and time increases the curry leaf becomes darker. The *L* and *b* values decreased (Figure 1 & 3) whereas *a* value increased (Figure 2) during thermal treatment. The post-process values *l, a,* and *b* ranged from 29.47 to 28.46, -0.41 to 1.54 and 7.43 to 6.32, respectively, after 1 h. The equilibrium Hunter L_∞, a_∞ and b_∞ values were 17.45, 2.56 and 2.23 respectively, when the paste was acidified in concentrated hydrochloric acid and thermally processed for a prolonged time (24 h). Curry leaf is green in colour, so Hunter -*a* value was considered as the physical parameters to describe the visual colour degradation during thermal processing. Several researchers (Ahmed et al., 2002a, 2002b; Steet & Tong, 1996) have used the Hunter colour -*a* value while studying the colour degradation kinetics of green vegetables. Eq. (3) can be written for Hunter -*a* value using fraction conversion technique as:

$$\ln[(-a)-(-a_\infty)]/[(-a_o)-(-a_\infty)] = -k.t \quad (7)$$

The rate of degradation of colour of curry leaf puree was determined by linear regression [Eq. (7)] The R^2 values were greater than 0.961 while the standard error were less than 0.0017for the entire range (Figure 4). Solid lines in Fig. 1 represent the model values. It is obvious from Figure 4 that degradation of visual colour represented by -*a* value of curry 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 al.,* 2002a; Shin & Bhowmik, 1994). In the present study, both L and b decreased with increase in -*a* value. Therefore the combination *Lab* was selected to represent the colour change. The Eq. (5) takes the following form in terms of *Lab* colour combination:

$$\ln[(Lab)-(L_\infty a_\infty b_\infty)]/ [[(L_o a_o b_o)-(L_\infty a_\infty b_\infty)] = -k.t \quad (8)$$

The rate of degradation of total colour was determined by linear regression [Eq. (8)]. The R^2 values were greater than 0.951 while the standard error were less than 0.0025 for the entire range (Figure 5). It is obvious from Fig. 2 that degradation of visual total colour as represented by (*Lab*) of curry leaf puree, followed first-order reaction kinetics. The

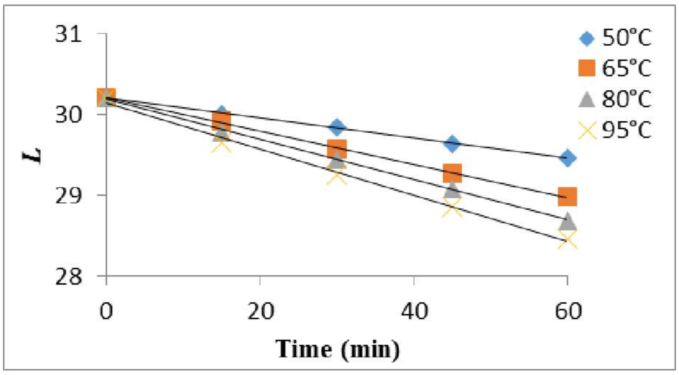

Figure 1. Thermal degradation of the *L*-value colour parameter as a function of time and temperature (experimental and model predicted values)

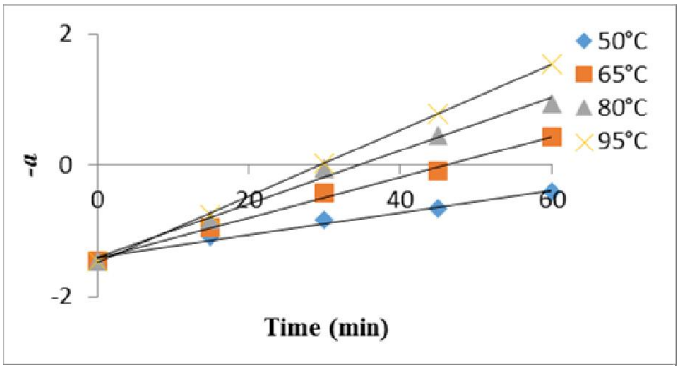

Figure 2. Thermal degradation of the (*-a)* value colour parameter as a function of time and temperature (experimental and model predicted values)

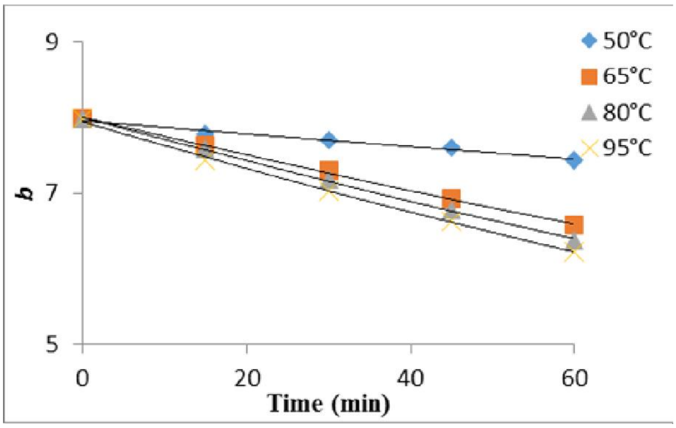

Figure 3. Thermal degradation of the *b*-value colour parameter as a function of time and temperature (experimental and model predicted values)

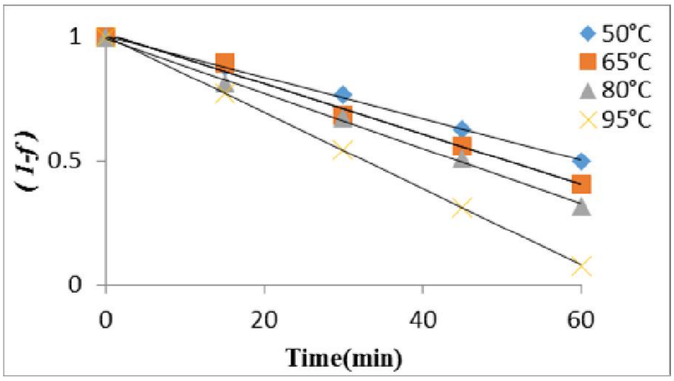

Figure 4. First-order colour (Hunter *-a* value) degradation kinetics of curry leaf puree at selected temperatures

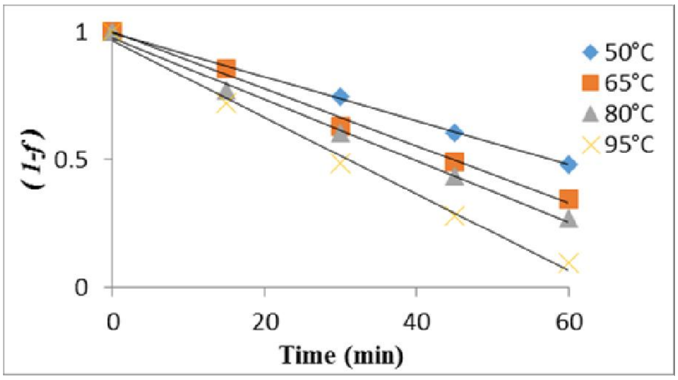

Figure 5. First-order colour (Hunter *lab* value) degradation kinetics of curry leaf puree at selected temperatures

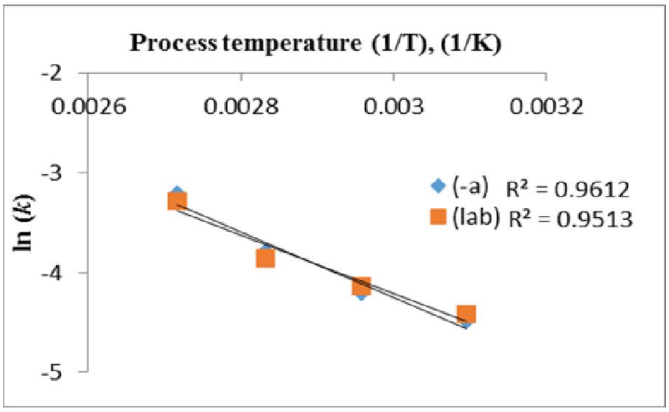

Figure 6. The Arrhenius plot relating rate constant to process temperature. Eqn [6]

Arrhenius plot (Figure 6) indicated that the dependence of rate constants for both *–a* and (*Lab*) on temperature followed the Arrhenius relationship (R^2>0.951). The activation energies for degradation of *-a* and (*Lab*) were 29.30 and 22.13 kJ/mol respectively. High activation energy signifies greater heat sensitivity of visual colour degradation during thermal treatment. It may therefore be inferred that the Hunter colour -*a* value should preferably be used to represent colour degradation during thermal processing of curry leaf paste.

Conclusion

The real behavior of curry leaf paste when subjected to heat treatment is translated using the *L, a,* and *b* system of objective colour measurement. The *L, -a, b* and *lab* values proved to be good indicators of the colour change during heating of curry leaf paste. The dependence of the reaction rate constant for all the colour parameters considered were described by Arrhenius model. Thermal degradation of colour followed a first-order reaction kinetics for the *L* and *b* values with activation energies of 18.44 ± 3.2 and 17.203 ± 5.9 KJ/mol, respectively. The modelling of *-a* and *lab* parameters were done using the fractional conversion equation. The activation energies obtained were 25.7 ± 1.7 and 23.53 ± 2.5 KJ/mol. Data reported in the experiment should be useful for the curry leaf processing industry and it is revealed that Hunter both *–a* and *lab* could be used as colour degradation parameter for online quality measurement.

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