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Kinetics of Colour Change of Double Concentrated Tomato Paste During Thermal Treatment

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ABSTRACT

The kinetics of the colour change of double concentrated tomato paste during heating was studied. The Hunter 'L', 'a', 'b' tristimulus values were measured to characterise the colour, and colour difference (ΔE); saturation index (SI) and 'a/b' ratio were calculated from those values. The kinetic study was performed using the capillary tube method with temperatures ranging from 70.0 to 100.0°C. The order of reaction and the constants E_a *and* k_0 *of the Arrhenius equation were determined. All the colour parameters followed an apparent first order kinetics, with the exception of ΔE, which showed a zero order behaviour. The degradation of the colour parameter 'L' followed two consecutive first order reactions, with* E_a *values of 11.5 and 5.73 kcal mol, and ln* k_0 *of 11.3 and 1.28* min^{-1} *for both phases, respectively. The parameter 'b'* ($E_a = 20.5$ *kcal mol;* $\ln k_0 = 22.2$ min^{-1}) *was more sensitive to temperature changes than the parameter 'a'* ($E_a = 9.79$ *kcal mol;* $\ln k_0 = 9.10$ min^{-1}), *and other colour parameters. The 'a/b' ratio showed an* $E_a = 6.86$ *kcal mol* ($\ln k_0 = 5.20$ min^{-1}), *smaller than that of all the other colour parameters, with the exception of 'L' (second phase). Thus, 'a/b' was less sensitive to changes during heating than most of the other parameters. Values of* E_a *and ln* k_0 *of 10.2 kcal mol and 12.9* min^{-1} *for ΔE, and 10.1 kcal mol and 9.28* min^{-1} *for SI were determined. The parameters obtained permit colour change prediction in double concentrated tomato paste during thermal processing.* © *1997 Elsevier Science Limited. All rights reserved*

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INTRODUCTION

The thermal processing of food is primarily intended to inactivate pathogens and other deteriorative microorganisms capable of making it unsuitable for consumption. At the same time, it has an inactivation effect on enzymatic systems, nutrients, and organoleptic properties, including texture and colour.

Colour is a very important quality factor in processed tomato products, particularly tomato concentrates, since it influences consumer acceptability. There are many reactions that can take place during thermal processing that affect colour. Among them, the most common are pigment degradation, especially carotenoids (lycopene, xanthophyll, etc.) and chlorophyll, and browning reactions such as the Maillard reaction and oxidation of ascorbic acid (Bontovits, 1981; Mauron, 1981). In tomato products, an important reaction is the degradation of the red pigment lycopene, originally in the *trans* form, that isomerises to the *cis* structure during heating, resulting in changes in colour. Additionally, the presence of residual chlorophyll that could be present in the juice as the result of grinding nonripe tomatoes for its manufacture, is converted to pheophytin of olive green colour (Narkiroj & Ranganna, 1977; Schwartz & Von Elbe, 1983; Schwartz & Lorenzo, 1990). The degradation of colour during heating of tomato products actually involves a series of complex reactions whose mechanisms and modes of action are presently not completely understood.

Colour is usually defined by three coordinates. There are various colour scales that can be used to characterise colour: CIE-X,Y,Z; the '*L*', '*a*', '*b*'; and the R_d, *a*, *b* scales. Similarly, colour indexes and differences can be calculated from these values. The '*L*', '*a*', '*b*' scale is recognised to show a better discrimination between small colour differences in the darker region of the colour space, providing good discrimination for saturated colours, as in the case of tomato products. For these reasons this scale is one of the most frequently used for food products (Francis, 1989), however, it is not so useful for light coloured samples (Anon., 1976).

The '*L*' value represents a nonlinear mathematical approximation of the white–black response of the eye, ranging from 100 for a perfect white to 0 for a perfect black, and measures the luminosity of the sample. A positive value of '*a*' indicates redness, and a negative value greenness. A plus value for '*b*' indicates yellowness and a minus value blueness. There are other parameters derived from the Hunter-'*L*', '*a*', '*b*' scale: the total colour difference (ΔE), the saturation index (SI) or chroma that indicates colour saturation and is proportional to its intensity, '*a*/*b*' ratio and the Hue angle among others (Francis & Clydesdale, 1975; Little, 1975; Anon., 1976; Abers & Wrolstad, 1979; Puppo-Ferreira, 1981; Francis, 1989). The '*a*/*b*' ratio has been used as a quality specification for tomato products. Values of 2.0 and above are indicative of an excellent colour in tomato paste, while a value below 1.80 is considered unacceptable (Goose & Binsted, 1973; Anon., 1976). The Hue angle ($\tan^{-1} b/a$) is another parameter frequently used to characterise colour in food products. An angle of 0 or 360° represents red Hue, while angles of 90, 180 and 270° represent yellow, green and blue Hue, respectively. It has been extensively used in the evaluation of colour parameters in green vegetables, fruits and meats.

Most of the quality factors, including colour, can be described by a degradation kinetics of zero or first order, with the effect of temperature in the velocity constant taken into account by the Arrhenius equation (Lund, 1975; Kessler & Fink, 1986; Wells & Singh, 1988; Rhim *et al.*, 1989a). Most of the kinetic analyses and studies

found in the literature for the change of colour by effect of heat in foods have been done in milk and some fruit juices (Rhim *et al.*, 1988a,b, 1989a,b; Pagliarini *et al.*, 1990). No kinetic studies related with the colour degradation of tomato concentrates during thermal processing were found in the literature. The objective of this research work was to study the kinetics of colour degradation by heat in double concentrated tomato paste in order to predict colour changes with temperature during thermal processing.

MATERIALS AND METHODS

Sample collection

Tomato samples of double concentrated tomato paste used in this study were collected during the tomato harvest season of 1992, in a commercial plant in Venezuela. The samples were drawn after the concentration process and before thermal processing for hot-filling. The plant used a cold-break rupture process. All samples used were from the same lot. About 15 l of tomato paste were sampled. The samples were kept in glass jars under refrigeration (about 4 to 7°C), protected from light with aluminium foil, and properly closed until used for the experiments.

Physical and chemical characteristics of tomato paste

The following analyses were done to characterise the double concentrated tomato paste used in this study: soluble solids (°Brix), corrected for temperature and acidity (NCA, 1968; Goose & Binsted, 1973), employing an Abbé refractometer (Bausch and Lomb, model 3-E); moisture: utilising a vacuum oven procedure at 70°C (AOAC, 1990); water activity (a_w) at 23°C using a psychometric equipment (Decagon CX-1); acidity: by electrometric titration (NCA, 1968; Goose & Binsted, 1973); pH: with a Ph meter (Bantex-300A) (NCA, 1968).

Colour analysis

After mixing the samples carefully, the determination of the colour parameters was done using a tristimulus colorimeter (Gardner XL-23) with the scales '*L*', '*a*', '*b*'. The equipment was calibrated against a red tile standard No. CG-6802 GCS-1 with the following colour parameters: '*L*' = 28.3; '*a*' = +49.6; '*b*' = +16.4. A 10 ml cell was used for the sample. The parameters '*L*', '*a*' and '*b*' of the sample were read and the values of ΔE, SI, '*a*/*b*', and Hue angle were calculated.

Colour difference ΔE was determined from the colour parameters of control nonheated samples and heated samples, and the values of SI and '*a*/*b*' from the parameters read using the equations presented by Francis and Clydesdale (1975), Anon. (1976), Abers and Wrolstad (1979), Puppo-Ferreira (1981) and Little (1975).

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

$$\text{SI} = \sqrt{a^2 + b^2} \quad (2)$$

$$\text{Hue angle} = \tan^{-1} b/a \quad (3)$$

where L_0, a_0 and b_0 denote the colour parameters for the control samples (unheated); L, a and b denote the colour parameters of the heated samples.

Kinetic studies for the change of colour during heating

To study the change of colour during heating, a modified capillary tube method similar to that described by Stumbo (1973) was followed. For this purpose and due to the difficulty of filling capillary tubes with tomato paste, Pyrex glass vials of about 9 ml in volume (total length: 59.3 mm; inner diameter: 13.5 mm; wall thickness: 2.0 mm), provided with bakelite screw caps were used. The vials were filled with 8.0 ml of sample, taking care to avoid incorporation of air bubbles and were closed. A total of 5 vials were used for each time period. All trials were done in triplicate. Therefore, a total of 15 vials were used for each time–temperature treatment.

For the heat treatment of the samples, time–temperature relationships applied included values in which the change of colour was visually evidenced. Temperatures of 70.0, 80.0, 90.0 and 100.0°C were selected. For the first three temperatures a thermostatic water bath (Blue-M-Magni Whirl, ± 0.1°C) was used, while for the temperature of 100.0°C a thermostatic bath using motor oil (Precision Scientific, ± 0.01°C) was employed. The heating time ranged from 5 to 90 min. After the heat treatment was applied the vials were removed and transferred to a cold water bath with crushed ice (about 2–3°C) in order to stop the heat treatment.

Due to the amount of product that had to be used, the samples were pre-heated in a microwave oven (Tappan 2450 MHz) to hasten heating to a temperature near to that of the experiment, and reduce the transient heat transfer process. In order to establish the heating time, a trial and error procedure was followed to determine the period to reach the temperature of the experiment within a range of about 2°C. The temperature was measured, after the samples were removed from the oven, introducing in the vials copper-constantan (Type T) thermocouples (Ellab, model 7667) using a Speedomax 2500 (Leeds and Northrup) equipment. In this way the error in the sample come-up time, after introduction in the bath, was minimised.

The heating time was measured from the moment the samples reached the processing temperature until they were introduced in the cold water bath, using a chronometer (Excelsior Park, ± 0.5 s). In order to determine the sample temperature a needle copper-constantan thermocouple, type T (Ellab, model 7667, 1 mm diameter), placed in the geometrical centre of the vials was used in junction with a time-temperature recorder (Leeds and Northrup, model Speedomax 2500, ± 1°F).

Once the samples (quintuplicates) were heated and cooled at the established time and temperature, they were carefully mixed in a composite sample, in order to have enough quantity to allow the measurement of the colour parameters in the colorimeter as indicated previously.

The apparent order of reaction for the colour parameters was determined by the adjustment of the experimental data to the integrated kinetic equations for orders 0, 1 and 2, using regression analysis. In each case the best fit was selected and the constant of velocity at each temperature determined from the slope of the straight line. The effect of temperature in the constant of velocity was determined from the linearised Arrhenius equation:

$$\ln k = \ln k_0 - E_a/RT \quad (4)$$

where k_0 denotes the pre-exponential factor (min^{-1}); E_a denotes the activation energy (kcal mol); R denotes the universal gas constant (kcal mol°K); and T the absolute temperature (°K). The value E_a was calculated from the slope and k_0 from the intercept of the straight lines given by eqn (4), using a linear regression program.

All the statistical analyses were done using the package STATGRAPHICS version 6.0.

RESULTS AND DISCUSSION

Physical and chemical characteristics

The physical–chemical characteristics obtained for double concentrated tomato paste used as sample for the determination of colour kinetics were ($\pm$ standard deviation): °Brix (20°C) = 28.1 $\pm$ 0.30; moisture (%) = 71.9 $\pm$ 0.03; a_w (24°C) = 0.96 $\pm$ 0.00; acidity (g citric acid/100 g sample) = 1.49 $\pm$ 0.36; pH = 3.90 $\pm$ 0.13. The values obtained are within the normal range reported in the literature for this product (Goose & Binsted, 1973; Serkat & Luh, 1976; Leoni & Bellucci, 1980; Madaiah *et al.*, 1986; Sandoval *et al.*, 1992).

Kinetics of colour change during heating

Colour parameter 'L'

The results obtained are presented in Fig. 1. This parameter tended to decrease faster during the first 20 min of the heating process, after which the reaction rate

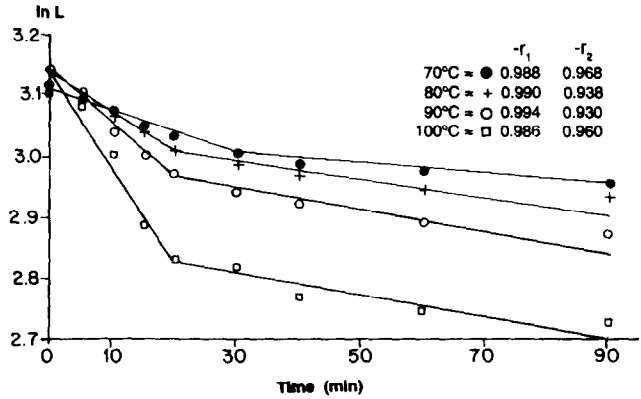

Fig. 1. Kinetics of the thermal degradation of the colour parameter '*L*' in double concentrated tomato paste.

became slower. The reaction took place according to two apparent consecutive first order reactions, with linear correlation coefficients (-r) between 0.986 and 0.994 for the first phase of the curve and between 0.938 and 0.968 for the second one. In all the cases the analysis of variance showed statistically significant linear regressions ($p < 0.05$).

This behaviour was possibly due to the presence of heat sensitive reactions in the first phase of the curve involving the degradation of thermolabile pigments, which in turn resulted in the formation of dark compounds that reduced luminosity, while in the second phase more thermostable pigments were involved. A similar behaviour was found by Palombo and Wijngaards (1990) in ground meat. The research works done in other products were indicative of simple first order reactions in: pear juice (Petropakis & Montgomery, 1984), sterilised juices (Tanchev, 1985), grape juice (Rhim *et al.*, 1989b), and green olives (Sanchez *et al.*, 1991). However, the above studies were done using relatively longer time intervals and relatively higher temperatures, so that the first phase could possibly not be evidenced in their experiments.

The change in colour during thermal processing of foods is postulated to take place by various mechanisms, including the degradation of pigments, oxidation of ascorbic acid, and the Maillard reaction (Serkat & Luh, 1976; Okitani *et al.*, 1983; Petropakis & Montgomery, 1984). These reactions can take place simultaneously but at a different reaction rate, depending on the colour factor involved, and may explain the two consecutive first order reactions found for this colour parameter. This reaction is actually an apparent first order reaction, since in the colour change there is probably more than one compound or colour factor involved and the reaction does not necessarily take place in one step through a single mechanism.

Figure 1 shows that the velocity constant (k) of the reaction, in any phase, increases with an increase in temperature. Values are greater in the first phase than in the second. The dependency of temperature on k was determined using the linearised Arrhenius eqn (3). The values obtained for the energy of activation E_a and the pre-exponential factor k_0 are presented in Table 1.

TABLE 1
Reaction Order and Arrhenius Constants for the Kinetics of Colour Change During Heating of Double Concentrated Tomato Paste

Colour parameter	*Reaction order*	E_a (*kcal mol*)	$\ln k_0$ (min^{-1})	(-r)
L (first phase)	1	11.5	11.3	0.996[b]
L (second phase)	1	5.73	1.28	0.979[a]
a	1	9.79	9.10	0.983[b]
b	1	20.5	22.2	0.952[a]
a/b	1	6.86	5.20	0.986[b]
ΔE	0	10.2	12.9	−0.977[a]
SI	1	10.1	9.28	0.989[b]
Hue angle	1	7.57	5.51	−0.993[b]

[a]Significant linear regression ($p < 0.05$).
[b]Highly significant linear regression ($p < 0.01$).

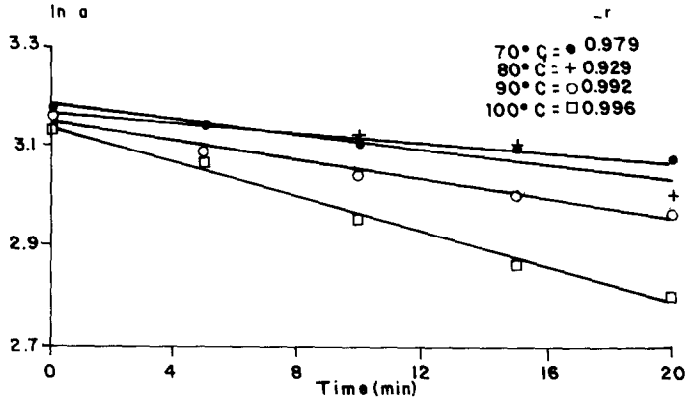

Fig. 2. Kinetics of the thermal degradation of the colour parameter '*a*' in double concentrated tomato paste.

*Colour parameter '**a**'*

The results for this parameter are shown in Fig. 2. A first order reaction was found. In all the cases a significant ($p < 0.05$) linear regression with correlation coefficients ($-r$) between 0.929 and 0.996 was obtained. A similar behaviour for this parameter was found by other authors in grape juice (Rhim *et al.*, 1989b), green olives (Sanchez *et al.*, 1991), dehydrated tomato juice (Lovric *et al.*, 1980), tomatoes (Bontovits, 1981) and spinach (Schwartz & Lorenzo, 1990, 1991). The results obtained for E_a and k_0 are shown in Table 1.

Various authors have pointed out that the change in the parameter '*a*' in tomato paste, depends basically on the effect of heat on two pigments: lycopene and xanthophyll, which are responsible for the red colour of this product. The colour of the paste depends also on the original chlorophyll content of the juice (Liu & Luh, 1977; Nagle *et al.*, 1979; Moressy & Liverotti, 1982). Red colour degradation with heat in tomato paste is due to the isomerisation of lycopene during heating (Boskovic, 1979), while the chlorophylls degrade to pheophytin, resulting in darkening of the paste (Schwartz & Lorenzo, 1990, 1991).

*Colour parameter '**b**'*

The results obtained for the parameter '*b*' (Fig. 3) followed a first order reaction. The analysis of variance showed highly significant linear adjustments ($p < 0.01$), with correlation coefficients ($-r$) from 0.902 to 0.995.

Similar results for the order of reaction were found by many authors in dairy products (Burton, 1984; Rhim *et al.*, 1988b) and in pickled green olives (Sanchez *et al.*, 1991). Rhim *et al.* (1988a) found a zero order kinetics in skim milk for this parameter.

The values calculated for E_a and k_0 are presented in Table 1.

*Colour parameter '**a**/**b**'*

As for the parameters '*a*' and '*b*', their ratio ('*a*/*b*') followed a first order kinetics (Fig. 4). The adjustment to this order showed statistically significant ($p < 0.05$) linear

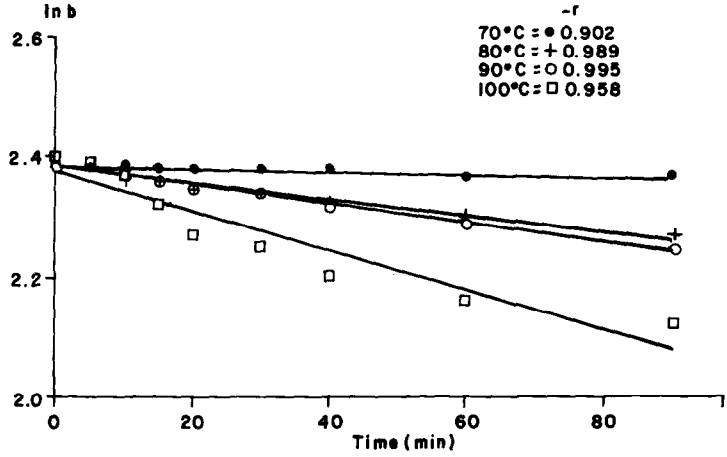

Fig. 3. Kinetics of the thermal degradation of the colour parameter '*b*' in double concentrated tomato paste.

regressions with correlation coefficients (*-r*) between 0.880 and 0.987. Likewise, this parameter could be also adjusted satisfactorily to a zero order kinetics as determined by Hayakawa (1977) in green vegetables. The values of E_a and k_0 obtained from the linearised Arrhenius equation are shown in Table 1.

Colour parameter 'ΔE'

This parameter was calculated from the primary colour parameters using eqn (1). The results obtained are presented in Fig. 5. A zero order reaction for the effect of

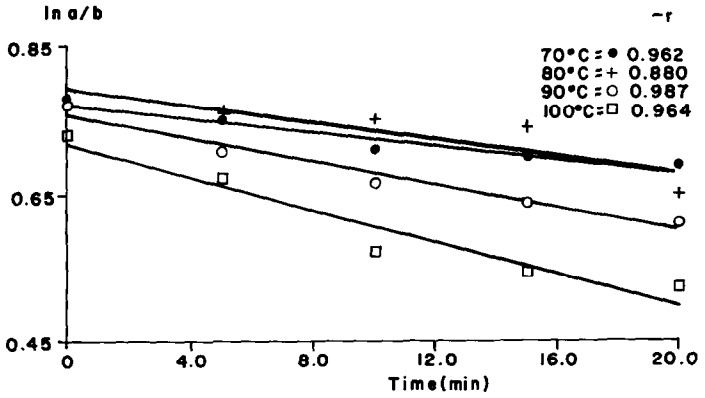

Fig. 4. Kinetics of the thermal degradation of the colour parameter '*a/b*' in double concentrated tomato paste.

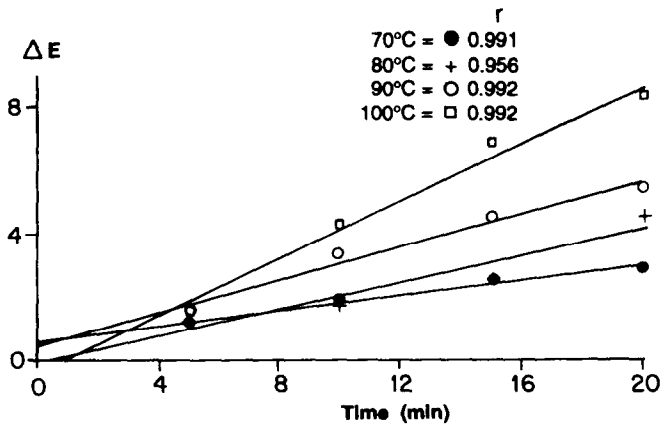

Fig. 5. Kinetics of the thermal degradation of the colour parameter 'ΔE' in double concentrated tomato paste.

heat in this parameter was determined. The same order of reaction was found by Flora (1976) and Rhim *et al.* (1989b) in grape juice, and by Pagliarini *et al.* (1990) in milk. Colour difference increased with temperature. A similar behaviour in other food products was reported by other authors: dairy products (Rhim *et al.*, 1988a; Pagliarini *et al.*, 1990), and grape juice (Rhim *et al.*, 1989b). The analysis of variance showed significant linear adjustments to this kinetic model ($p<0.05$) with linear correlation coefficients ($-r$) between 0.956 and 0.992. The E_a and k_0 values obtained from the Arrhenius equation are presented in Table 1.

Colour parameter 'SI'

The saturation index (SI) was calculated from values 'a' and 'b' using eqn (2). This parameter diminished as the temperature increased and showed a first order kinetics (Fig. 6). Similar behaviour was observed by Rhim *et al.* (1989b) in grape juice. The linear adjustment to this kinetic model showed significant ($p<0.05$) values in all cases, with linear correlation coefficients (r) between 0.936 and 0998. The values of E_a and k_0 are shown in Table 1.

Hue angle

The Hue angle followed a kinetics behaviour similar to that of the parameter 'a/b'. It increased with temperature and time, showing a first order kinetics (Fig. 7). The adjustment to this order showed statistically significant ($p<0.05$) linear regressions with correlation coefficients (r) between 0.890 and 0.980. As for 'a/b' this parameter could be also adjusted satisfactorily to a zero order kinetics. The values of E_a and k_0 obtained from the linearised Arrhenius equation are shown in Table 1.

From the data presented above for the colour parameters studied, it can be observed that the total colour difference ΔE showed the highest value for the rate constant (k), calculated with eqn (4) as compared with the other colour parameters, thus showing the greatest rate of change at a given temperature. According to the

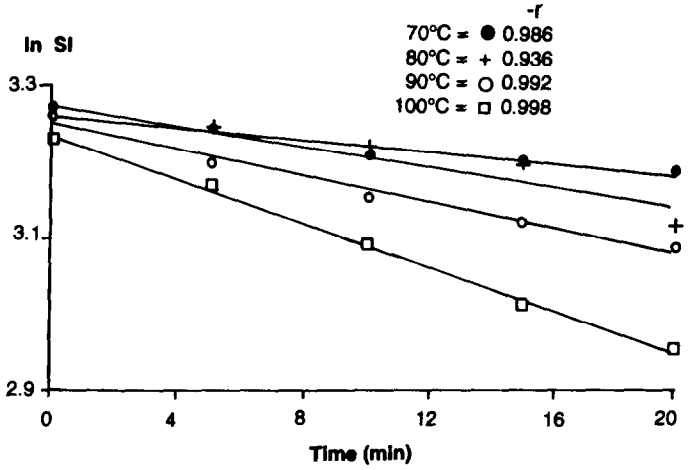

Fig. 6. Kinetics of the thermal degradation of the colour parameter 'SI' in double concentrated tomato paste.

experimental data obtained, the reaction rate for the colour parameters studied can be arranged in decreasing order at a given temperature. For example at 100°C the rate constant (k in min^{-1}) was: ΔE (0.458), 'a' (0.0172), L (first phase) (0.0150), SI (0.0144), 'a/b'(0.0110), Hue angle (0.00924), 'b' (0.00321) and L (second phase)

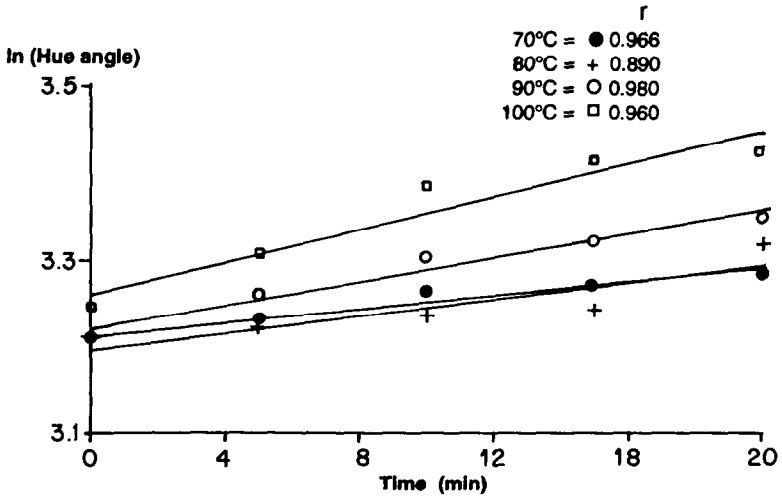

Fig. 7. Kinetics of the thermal degradation of the colour parameter 'Hue angle' in double concentrated tomato paste.

(0.00146); while at 70°C the order was: ΔE (0.122), '*a*'(0.00500), '*a*/*b*'(0.00460), SI (0.00406), Hue angle (0.00384), *L* (first phase) (0.00360), *L* (second phase) (0.000762) and '*b*' (0.000240). These differences between the relative magnitudes of the reaction rate of the colour parameters studied at lower and higher temperatures can be explained by the sensitivity of the reaction constant to changes in temperature given by the term E_a in eqn (4). From all the parameters studied, '*b*' showed the highest E_a (20.5 kcal mol), and therefore was the most sensitive to changes in temperature, although its rate constant was among the lower. *L* (second phase) and '*a*/*b*' showed E_a values of 5.73 and 6.86 kcal mol, among the less sensitive to changes in temperature.

CONCLUSIONS

The kinetic study of the colour change of double concentrated tomato paste during heating at temperatures between 70.0 and 100.0°C, showed that all the colour parameters investigated ('*L*', '*a*', '*b*', '*a*/*b*', SI and Hue angle) followed a first order reaction kinetics during heating, with the exception of the parameter ΔE that followed a zero order kinetics. The colour parameter '*L*' changed according to two consecutive apparent first order reactions with values of E_a of 11.5 and 5.73 kcal mol and $\ln k_0$ of 11.3 and 1.28 min^{-1} for both phases, respectively, and a breaking point after 20 min of heating. The '*a*/*b*' ratio and the Hue angle could also be adjusted to a zero order kinetics.

The values of E_a and k_0 obtained for the colour parameters studied in this research work (Table 1), allows the prediction of colour changes that take place in double concentrated tomato paste during thermal processing.

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