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Kinetic models of quality changes and shelf-life prediction of sukiyaki sauce during storage

[1]Yuenyongputtakal, W., [2]Srisuk, N., [1]Wongyai, S. and [1]*Jirasatid, S.

[1]*Department of Food Science, Faculty of Science, Burapha University, Chonburi, 20130, Thailand*
[2] *Biological Science Program, Faculty of Science, Rajabhat Rajannagarindra University, Chachoengsao, 24000, Thailand*

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Abstract

The shelf-life models based on various temperatures were developed for Thai-style sukiyaki sauce. The sauces were stored at 4°C for 224 days, 30°C for 140 days, and 40°C for 30 days. Quality changes were measured during storage, and kinetic models for selected quality indices were developed to estimate changes and predict shelf life. The results indicated that sukiyaki sauces experienced changes in water activity, pH, and colour parameters (L^*, a^*, b^*, hue, total colour difference (TCD), and browning index (BI)) along with a decline in sensory attributes (appearance, colour, odour, taste, and overall acceptability) during storage, particularly at high temperatures (40°C). However, low levels of total viable count, yeast and mould count, and *Staphylococcus aureus* count remained constant throughout the storage period at various temperatures. Sensory attributes, TCD, and BI were selected as critical parameters for predicting shelf life due to their major deterioration in sukiyaki sauce. The decrease in sensory attributes and the increase in BI followed a second-order kinetic reaction, while the increase in TCD followed a zero-order kinetic reaction. The rate constant variations with temperature were well described by the Arrhenius and Ball models. Shelf-life models derived from multiple quality indices associated with the Arrhenius (R^2 = 0.999) and Ball (R^2 = 1.0) models, indicated approximately 6.7 and 6.5 months, respectively, for the shelf life of sukiyaki sauce stored at 30°C.

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Introduction

Thai-style sukiyaki sauce is a spicy dipping sauce. It is an acidified food (pH < 4.6), allowing it to undergo pasteurisation to destroy foodborne pathogens and microbial spoilage before being hot filled into glass bottles and stored at room temperature (McGlynn, 2016; Purohit *et al*., 2023).

The quality deterioration of sauces including browning and the development of off-flavours occurs during storage, negatively impacting consumer acceptance (Lee *et al*., 2015; Purohit *et al*., 2023). In addition, the proliferation of microorganisms such as microbial spoilage and pathogens in sauce can result in spoilage and foodborne diseases, respectively (Ray and Bhunia, 2014). Microbial activities also play a role in altering the physicochemical properties of sauces such as water activity (a_w) and pH value (Yun *et al*., 2007; Lee *et al*., 2015; Purohit *et al*., 2023).

Maintaining food safety and acceptable sensory characteristics are essential throughout its shelf life. Various factors such as temperature, storage duration, pH, and a_w significantly influence quality deterioration, ultimately limiting the shelf life of sauces (Yun *et al*., 2007; Purohit *et al*., 2023). Yun *et al.* (2007) reported notable changes in the pH, thiobarbituric acid levels, microbial counts, and colour values of Korean savoury sauce over 210 days of storage, with more pronounced changes at high temperatures (35°C) compared to low temperatures (5°C). Changes in food quality during storage, including colour deterioration, off-flavour development, and microbial growth can be evaluated using kinetic models such as the Arrhenius and Ball

*Corresponding author.
Email: sani@go.buu.ac.th

models, which enable accurate prediction of shelf life based on quality degradation rates (Shukla *et al*., 2020; Zhang *et al*., 2021; Kim *et al.,* 2022). Zhang *et al*. (2021) applied the Ball model to predict the shelf life of kiwifruits, demonstrating that shelf life based on quality indices can be accurately estimated under different storage temperatures.

Knowledge of the effect of storage temperature on sukiyaki sauce helps evaluate the quality changes and then predict its shelf life. This can be accomplished through empirical approaches using kinetic models based on reaction orders such as zero-, first-, and second-order reactions (Shukla *et al*., 2020; Zhang *et al*., 2021; Kim *et al*., 2022). Hence, the objectives of the present work were to (1) investigate the quality changes including physicochemical, microbiological, and sensorial properties of sukiyaki sauce during storage at different temperatures and durations, and (2) develop kinetic models to determine the quality changes and subsequently predict the product's shelf life under different storage temperatures.

Materials and methods

Sample preparation

Thai-style sukiyaki sauce, a local product, was acquired from a local manufacturer in Chonburi province, Thailand. The ingredients included pickled garlic, pickled red chili, white sesame seeds, sesame oil, vinegar, sugar, salt, soy sauce, chili sauce, water, and a preservative like sodium benzoate. The pickled garlic, pickled red chili, chili sauce, and water were homogeneously blended. Afterword, other ingredients were added, mixed well, and packed in glass bottles. The proportions of the ingredients and specifics of the manufacturing process are unavailable because they are kept confidential by the company.

Heating treatment

The sauce was hot-fill-hold processed for microbial safety. Heating (1,000 mL) at 90°C for 15 min in a water bath reduced the total viable count (TVC) to 1.8 log CFU/g and the yeast and mould count (YMC) to < 1 log CFU/g, meeting Thai Community Product Standard (TCPS No. 466/2556) limits of < 4 and < 2 log CFU/g, respectively (TCPS, 2013). Following heat treatment, samples were immediately removed from the water bath. Samples (200 mL) were then hot filled at 85°C into the glass bottles, screwed with caps, inverted, and held for 2 min to sterilise the inner surfaces before being cooled to 35°C by water spraying (McGlynn, 2016; Purohit *et al*., 2023).

Storage condition

Samples were stored at three different temperatures (4, 30, and 40°C). As the rate of sukiyaki sauce deterioration increased with temperature, the total storage duration and sampling frequency were adjusted accordingly. Sampling (two bottles) was conducted every 28 d for up to 224 d at 4°C, every 14 d for up to 140 d at 30°C, and every 3 d for up to 30 d at 40°C. Qualitative analysis was conducted through sensory evaluation and physicochemical (a_w, pH, and colour parameters) and microbiological measurements (TVC, YMC, and *Staphylococcus aureus*).

Water activity and pH measurement

The water activity (a_w) was determined using a digital water activity meter (EZ-200, Japan) (Yun *et al*., 2007). For the pH measurement, 30 mL of sample was transferred to a 100 mL beaker, and the pH was measured using a calibrated pH meter (Lab 850, Schott, Germany) (Purohit *et al*., 2023).

Colour measurement

The colour of sukiyaki sauce was assessed based on the CIE colour system using a Hunter colorimeter (CR-400, Konika Minota, Japan). The instrument was calibrated with black and white plates. Sample (2 mL) was poured into a transparent plastic dish and the colour parameters including L^* (lightness), a^* (redness/greenness), and b^* (yellowness/blueness) values were measured in dark conditions (Purohit *et al*., 2023).

The total colour difference (ΔE; TCD) between initial and stored samples was computed using the Hunter-Scottfield equation (Eq. 1):

$$\text{TCD} = \sqrt{(L_0^* - L^*)^2 + (a_0^* - a^*)^2 + (b_0^* - b^*)^2} \quad \text{(Eq. 1)}$$

where, L_0^*, a_0^*, and b_0^* = colour values at the initial time; L^*, a^*, and b^* = colour values at any given time during storage, respectively (Purohit *et al*., 2023).

The browning index (BI) corresponds to the brown colour of the samples, resulting from enzymatic and/or non-enzymatic browning reactions (Choosuk *et al*., 2022). The BI was calculated using Eqs. 2 and 3:

$$\mathrm{BI} = \frac{100(x-0.31)}{0.172} \quad \text{(Eq. 2)}$$

$$x = \frac{(a^* + 1.75L^*)}{(5.645L^* + a^* - 3.012b^*)} \quad \text{(Eq. 3)}$$

The hue angle was computed as follows: hue angle = arctan (b^*/a^*) for the first quadrant ($+a^*$, $+b^*$) (McLellan *et al*., 1995).

Microbiological analysis

Samples (25 g) were homogenised with 0.1% sterile peptone (225 mL) and then subjected to a 10-fold serial dilution. TVC was enumerated after incubation at 37°C for 48 h on compact dry total count (Nissui Pharmaceutical Co., Ltd, Japan). The mesophilic bacterial colonies were coloured red (Yousef and Carlstrom, 2003). For YMC, appropriate dilutions of samples (10^{-1}) were incubated at 30°C for 4 - 5 d on compact dry yeast and mould plates (Nissui Pharmaceutical Co., Ltd, Japan). Yeasts appeared as blue colonies, while moulds formed cottony-like colonies with characteristic colours. The TVC and YMC were reported as log colony-forming units per mL (log CFU/g) (Yousef and Carlstrom, 2003).

For the detection of *Staphylococcus aureus*, a sample (25 g) was first diluted in 0.1% peptone water (225 mL) and thoroughly mixed. Then, the diluted sample (1 mL) was transferred into 10 mL of tryptic soy broth (TSB) containing 10% NaCl and incubated at 35°C for 48 h. For the isolation step, the enriched sample was streaked onto Baird-Parker agar medium and incubated at 35°C for 48 h. *S. aureus* colonies on the Baird-Parker agar appeared jet-black to grey-black, surrounded with an opaque halo and a clear zone. To confirm the presence of *S. aureus*, a coagulase test was performed using rabbit plasma with ethylenediaminetetraacetic acid (EDTA). Two to three colonies from the Baird-Parker agar were transferred into the plasma, and if the plasma clotted within 6 h of incubation at 35°C, they were considered coagulase-positive. The result was reported as *S. aureus* detected in 0.1 g of food (Yousef and Carlstrom, 2003; DMSc, 2017).

Sensory evaluation

Sensory evaluation of sukiyaki sauce was conducted by 30 untrained panellists, all of whom were over 18 years of age. The tasting panel consisted of students and staff from the Department of Food Science, Burapha University, Thailand. Prior to the test, all panellists received information regarding sensory evaluation. The sample was evaluated in a tasting room equipped with separate booths featuring temperature and lighting controls. Sukiyaki sauce (20 mL) was served at room temperature in white plastic cup coded with a random three-digit number following the sensory evaluation standard procedure. Panellists were instructed to use a 9-point hedonic scale to rate their liking for appearance, colour, odour, taste, and overall acceptability (1 = dislike extremely, 5 = neither like nor dislike, 9 = like extremely) (Lee *et al*., 2015).

Kinetics models

The reaction rates of changes in quality indices such as sensory characteristics, TCD, and BI during storage of sukiyaki sauces were described using simple kinetic models based on zero-, first-, and second-order kinetics as presented in Eq. 4:

$$\frac{dC}{dt} = \pm kC^n \quad \text{(Eq. 4)}$$

where, k = rate constant, n = order of the reaction, and C = quantitative value of sensory characteristics, TCD, and BI (Van Boekel, 2009; Ling *et al*., 2015).

In Eq. 4, a negative sign indicates a decrease in quantitative values such as a loss in sensory scores with time. Conversely, a positive sign indicates an increase in quantities over time such as with TCD and BI (Singh *et al*., 2009). A kinetic model was proposed and interpreted into a mathematical model by deriving a differential equation using zero-, first-, and second-order kinetics (n = 0, 1, and 2, respectively). The differential equation was solved using numerical integration and fitted to the experimental data through non-linear regression. The reaction order was determined from the best fit line using the coefficient of determination (R^2) and root mean square error (RMSE) (Remini *et al*., 2015; Batu and Kadakal, 2021).

Arrhenius model (Eq. 5) was used to simulate the temperature dependence of the reactions:

$$k = k_0 exp\left[-\frac{E_a}{RT}\right] \quad \text{(Eq. 5)}$$

where, k = rate constant, k_0 = Arrhenius constant, T = absolute temperature (K), R = ideal gas constant (8.3145 J/mol·K), and E_a = activation energy (J/mol), which is calculated by multiplying R by the slope in the plot of ln k versus $1/T$ (Van Boekel, 2009; Kim *et al*., 2022).

The Ball model (Eqs. 6 and 7) is widely used in food processing for microbial destruction. It is employed to estimate the decimal reduction time (D value), which is related to temperature *via* a Z value. The Z value was calculated as the reciprocal of the slope from the plot of $\log_{10} D$ against temperature (T) using regression analysis (Van Boekel, 2009; Ling *et al*., 2015).

$$D = \frac{\ln 10}{k} \quad \text{(Eq. 6)}$$

$$D = D_0 10^{\frac{-T}{Z}} \quad \text{(Eq. 7)}$$

where, D = storage time required to reduce the quantitative values by 90% (days), D_0 = D value at T = 0°C (days), T = temperature (°C), and Z = temperature required for one $\log_{10} D$ value reduction or a factor of 10 (°C) (Ling *et al*., 2015; Kim *et al*., 2022).

Shelf-life prediction

The key parameters were applied to predict the shelf life of products using Eqs. 8 to 10 for zero-, first-, and second-order reactions, respectively, according to Eq. 4:

Zero-order reaction; $C_t - C_0 = \pm k t_s$ (Eq. 8)

First-order reaction; $\ln\left(\frac{C_t}{C_0}\right) = \pm k t_s$ (Eq. 9)

Second-order reaction; $\frac{1}{C_0} - \frac{1}{C_t} = \pm k t_s$ (Eq. 10)

where, C_0 and C_t = quantitative value at initial and any storage time, respectively, and t_s = predicted shelf life.

In Eqs. 8 to 10, a negative sign of the k value indicates a decrease in quantitative values like sensory characteristics, while a positive sign of the k value indicates an increase in quantities over time like TCD and BI (Khalil, 2023).

Model evaluation

The goodness of model fitting to the experimental data was evaluated by the values of R^2 (Eq. 11) and RMSE (Eq. 12), acquired from the kinetic plots with n = 0, 1, and 2, respectively. The best fit of the model to the experimental data was considered from the highest of R^2 and the lowest of RMSE (Remini *et al*., 2015; Batu and Kadakal, 2021). R^2 values close to 1 show that the curve closely follows the data.

$$R^2 = 1 - \frac{\sum_{i=1}^{n}(P_{obs} - P_{pred})^2}{\sum_{i=1}^{n}(P_{obs} - P_{mean})^2} \quad \text{(Eq. 11)}$$

$$\text{RMSE} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(P_{obs} - P_{pred})^2} \quad \text{(Eq. 12)}$$

where, P_{obs} = observed value, P_{pred} = predicted value, P_{mean} = average value from all observed values, and n = number of the data (Batu and Kadakal, 2021; Hodson, 2022).

Statistical analysis

Each analytical determination was performed in triplicate. The results were expressed as mean ± standard deviation. All experiments were conducted using a completely randomised design (CRD), except that the sensory evaluation was designed using a randomised complete block design (RCBD). To determine the significant effect of storage time on sauces, One-way ANOVA was deployed with Tukey's range test at a 95% confidence level ($p \leq 0.05$). All statistical analyses were conducted using Minitab software version 18 (Minitab Pty. Ltd., Sydney, NSW, Australia). All data obtained by the Arrhenius and Ball models were analysed by Microsoft Excel.

Results and discussion

Changes in water activity and pH values

As seen in Figure 1a, the initial a_w values of all sauces were 0.96 - 0.97. For sauces stored at 4°C, the a_w decreased continuously during the first 56 d, followed by a slight increase between 112 and 224 d. Significant differences in a_w values, ranging from 0.838 to 0.983, were observed over 224 d of storage period at 4°C ($p \leq 0.05$), possibly due to microbial metabolic activity. However, there were no significant changes in the a_w of sukiyaki sauces stored at 30°C for 140 d, with values initially at 0.968 and decreasing to 0.886 by the end of storage ($p > 0.05$). In addition, the a_w of sauce stored at 40°C varied within the range of 0.96 - 0.99 over 30 d ($p \leq 0.05$). This suggested that all samples maintained $a_w > 0.83$ throughout the storage time, thereby increasing the probability of deterioration from lipid oxidation, non-enzymatic browning reactions, and microbial growth (Babosa-Canovas *et al*., 2007).

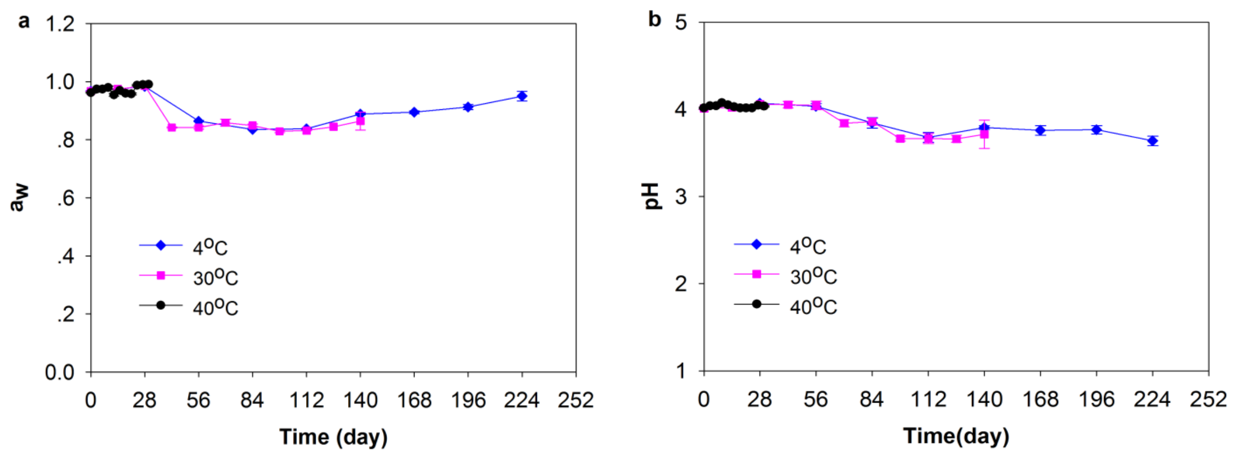

Figure 1. Changes in physicochemical of sukiyaki sauces stored at different storage temperatures: **(a)** a_w and **(b)** pH values.

The initial pH of the sauces was 4.01 - 4.02, and it decreased continuously to 3.64 after 224 d of storage at 4°C ($p \leq 0.05$) and to 3.72 after 140 d at 30°C ($p \leq 0.05$). The minimal changes in pH were observed during storage at 40°C for 30 d, with values ranging between 4.02 - 4.04 ($p \leq 0.05$) (Figure 1b). The decrease in pH value may result from both chemical reactions and the metabolic activities of microorganisms. Chemical reactions including lipolysis and lipid oxidation of oil components within sukiyaki sauce could occur, resulting in a drop in pH and the development of a rancid flavour. Simultaneously, microbial growth in the product could lead to the accumulation of organic acids and/or the hydrolysis of lipid components, resulting in the formation of fatty acids and consequently a decrease in pH value (Yun *et al.*, 2007; Lee *et al.*, 2015; Purohit *et al.*, 2023). The alterations observed in the a_w and pH values of sukiyaki sauces in the present work agreed with the findings from other studies examining the storage of various sauces (Yun *et al.*, 2007; Ji *et al.*, 2024).

Colour changes

Throughout the storage period, the values of L^*, a^*, and b^* of all sukiyaki sauces showed no significant changes at various temperatures ($p > 0.05$) (Figures 2a - 2c). The sukiyaki sauces exhibited a predominately orange visual, as indicated by the hue-angle fluctuating from 44.69 to 44.63, 44.71 to 47.80, and 46.40 to 46.86° of sauces stored at 4°C for 224 d, 30°C for 140 d, and 40°C for 30 d, respectively (Figure 2d) (McLellan *et al.*, 1995).

As shown in Figure 2e, TCD of all sample increased significantly over storage time ($p \leq 0.05$), resulting from changes in L^*, a^*, and b^* values. TCD of 3.02, 4.33, and 3.81 units were recorded after storage at 4°C for 224 d, 30°C for 140 d, and 40°C for 30 d, respectively. This indicated that the colour differences were perceivable to the human eye due to TCD > 3 units (Urbina *et al.*, 2021).

A stable BI value range of 284.93 to 281.02 was observed at 4°C for 224 d ($p > 0.05$), indicating the absence of browning pigment formation under refrigerated conditions (Figure 2f). At higher storage temperatures (30 and 40°C), the index increased insignificantly over storage time ($p > 0.05$), with values approximately 1.1 times higher after 140 d at 30°C and about 1.18 times higher after 30 d at 40°C.

These results suggested that the majority of colour changes in sukiyaki sauces during storage were apparently due to the formation of browning pigments and possibly the decomposition of carotenoid pigments. A non-enzymatic browning reaction such as Maillard is reported to be responsible for the formation of brown pigments during the storage of food products and these reactions can occur through a variety of mechanisms (Shukla *et al.*, 2020). Additionally, browning products may form due to the degradation of vitamin C (Bharate and Bharate, 2014). It can be summarised that the temperature impacted colour parameters, particularly TCD and BI. Nevertheless, the colour parameters (L^*, a^*, b^*, hue, and BI) remained stable at 4°C, as evidenced by the results earlier described. These observations agreed with the findings of Purohit *et al.* (2023), who reported slight decreases in L^* values, stability in hue-angle, and significant increases in TCD of various chicken-wing sauces during storage at ambient temperature (18.35°C) for 12 months.

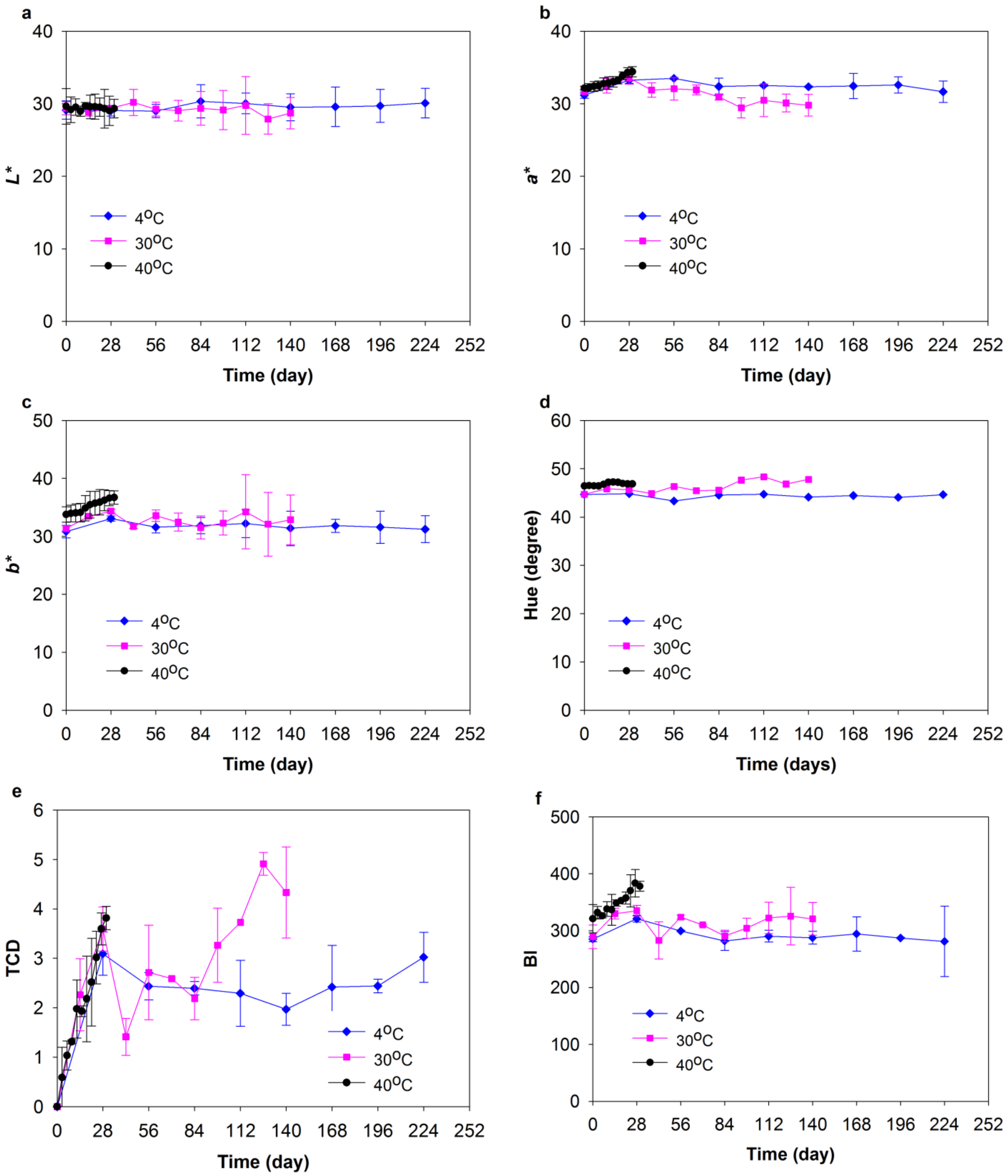

Figure 2. Colour changes of sukiyaki sauces during storage at different temperatures: **(a)** *L**, **(b)** *a**, **(c)** *b**, **(d)** hue-angle, **(e)** total colour difference (TCD), and **(f)** browning index (BI).

Microbial changes

The overall microbial load was low in sukiyaki sauces during storage (Table 1). The TVC tended to be stable throughout the storage period, ranging from 1.64 to < 1 and 1.35 to < 1 log CFU/g for samples stored at 4°C for 224 d and 30°C for 140 d, respectively. In contrast, an increase in TVC, ranging from 1.18 to 1.60 log CFU/g, was detected in samples stored at 40°C for 30 d ($p \leq 0.05$). Additionally, YMC remained < 1 log CFU/g throughout the storage period in all sauces. *S. aureus*, pathogenic bacteria, was not detected in 0.1 g of sauces throughout the storage period. These results confirmed the effectiveness of heating and hot-filling processes of sukiyaki sauces in reducing the initial microbial loads. Besides, the product had sufficient acidity (pH < 4.5), which could be a hurdle for controlling microbial proliferation during storage. Nevertheless,

Table 1. Microbiological properties of sukiyaki sauces during storage at different temperatures.

Test	Temperature	Storage time (d)																								
	(°C)	0	3	6	7	9	12	14	15	18	21	24	27	28	30	42	56	70	84	98	112	126	140	168	196	224
TVC	4	1.64												1.54			< 1		1.0		< 1		1.48	< 1	1.30	1.0
(log CFU/g)	30	1.35						1.40						1.54		1.18	1.0	< 1	1.18	1.18	< 1	< 1	< 1			
	40	1.30	1.18	1.18		1.18	1.35		1.47	1.51	1.40	1.57	1.57		1.60											
YMC	4	< 1												< 1			< 1		< 1		< 1		< 1	< 1	< 1	< 1
(log CFU/g)	30	< 1						< 1						< 1		< 1	< 1	< 1	< 1	< 1	< 1	< 1	< 1			
	40	< 1	< 1	< 1		< 1	< 1		< 1	< 1	< 1	< 1	< 1		< 1											
	4	nd												nd			nd		nd		nd		nd	nd	nd	nd
S. aureus	30	nd						nd						nd		nd	nd	nd	nd	nd	nd	nd	nd			
	40	nd	nd	nd		nd	nd		nd	nd	nd	nd	nd		nd											

nd: not detected in 0.1 g of sauce; TVC: total viable count; and YMC: yeast and mould count.

certain spoilage microorganisms, particularly lactic acid bacteria, and some yeasts such as *Saccharomyces cerevisiae*, are capable of growing in low-pH products.

The results concluded that the TVC, YMC, and *S. aureus* in sukiyaki sauces conformed to the TCPS (2013) and the notification of the Ministry of Public Health of Thailand (FDA Thai, 2020), thus confirming their safety for human consumption. This agreed with a study conducted by Purohit *et al.* (2023), where five sauces, subjected to heating at 90.55°C for 45 S and hot-filled, showed stability in aerobic plate count (< 1 log CFU/g) and YMC (< 1 log CFU/g), and no detection of *S. aureus* (in 25 g of sample) over a 12-month storage period. In addition, Yun *et al.* (2007) observed a slight increase in microbial counts in Korean savoury sauce stored at 5 - 25°C for 250 d, while pronounced microbial growth occurred at 35°C.

Sensory evaluation

The average initial scores (0 day) for sukiyaki sauces were 7.77, 7.76, 7.60, 7.52, and 7.64 for appearance, colour, odour, taste, and overall acceptability, respectively. These scores corresponded to the level of 'like moderately' to 'like very much,' showing sensorial acceptance by the panellists. The sukiyaki sauces experienced great decreases in all sensory attributes as the storage period ($p \leq 0.05$) and temperature increased, as seen in Figure 3. Panellists noted a displeasing dark colour and rancid flavour in the sukiyaki sauces, likely resulting from the formation of browning pigments and lipid oxidation, which were accelerated at high temperatures (40°C). Nevertheless, the good sensory attributes were maintained until the end of the storage time, as indicated by scores of > 5 for each sensory attribute. This suggested that the loss of sensory attributes induced by an increase in temperature could be attributed to the increases in TCD and BI of sukiyaki sauces, as observed in our results. A similar result was shown by Lee *et al*. (2015), who observed a decrease in the sensory characteristics of tomato-based and Korean traditional fermented food-based sauces with temperature (25 - 45°C) and time (180 d) of storage, resulting from rancidity and dark colour in the sauces. Furthermore, according to Purohit *et al.* (2023), minimal changes were observed in sensory attributes such as colour, flavour, and odour in different chicken-wing sauces during the first 6-month storage at ambient temperature (18.35°C). However, after 7-month storage, colour changes and a slight rancid flavour were noted in chicken-wing sauces like lemon pepper.

Kinetics models and shelf-life prediction

The shelf life of food products refers to the duration during which foods remain safe to consume and maintain acceptable qualities including sensory, nutritional, and physicochemical properties (Man and Jones, 2000). In the present work, although the microbiological properties of sukiyaki sauces remained stable, major changes were observed in sensory attributes, TCD, and BI over the storage period, especially at high temperatures (40°C). Consequently, sensory attributes, TCD and BI emerged as key parameters for determining the shelf life of sukiyaki sauce.

Kinetic models for predicting shelf life can be estimated using zero-, first-, and second-order reactions (Khalil, 2023). The reaction order of quality indices was determined based on the R^2 and RMSE obtained from changes in each quality index over the storage period at all temperatures (Remini *et al*., 2015). Table 2 presents the kinetic parameters of quality indices during the storage of sukiyaki sauces. The results indicated that all sensory attributes including appearance, colour, odour, taste, and overall acceptability showed a better fit with the second-order reaction model. This was evidenced by higher R^2 values and lower RMSE values as compared to the first- and zero-order models. However, all sensory attributes of sukiyaki sauces stored at 4°C exhibited relatively poor fit with zero-, first-, and second-order reactions due to an indirectly observable trend of sensory scores during storage. The second-order reaction model has been used in previous studies to predict shelf life and describe food quality deterioration (Weiss *et al*., 2018; Khalil, 2023). In the present work, the reaction rate constant (k) for the decrease of all sensory attributes increased greatly with storage temperature from 4 to 40°C, indicating that temperature was a crucial parameter for the storage of sukiyaki sauces (Table 2). This agreed with the results obtained in previous studies, which reported higher k values for sensory score deterioration at higher storage temperatures (Shukla *et al*., 2020; Kim *et al*., 2022).

Considering the physicochemical qualities, non-regression analysis revealed that the increase in

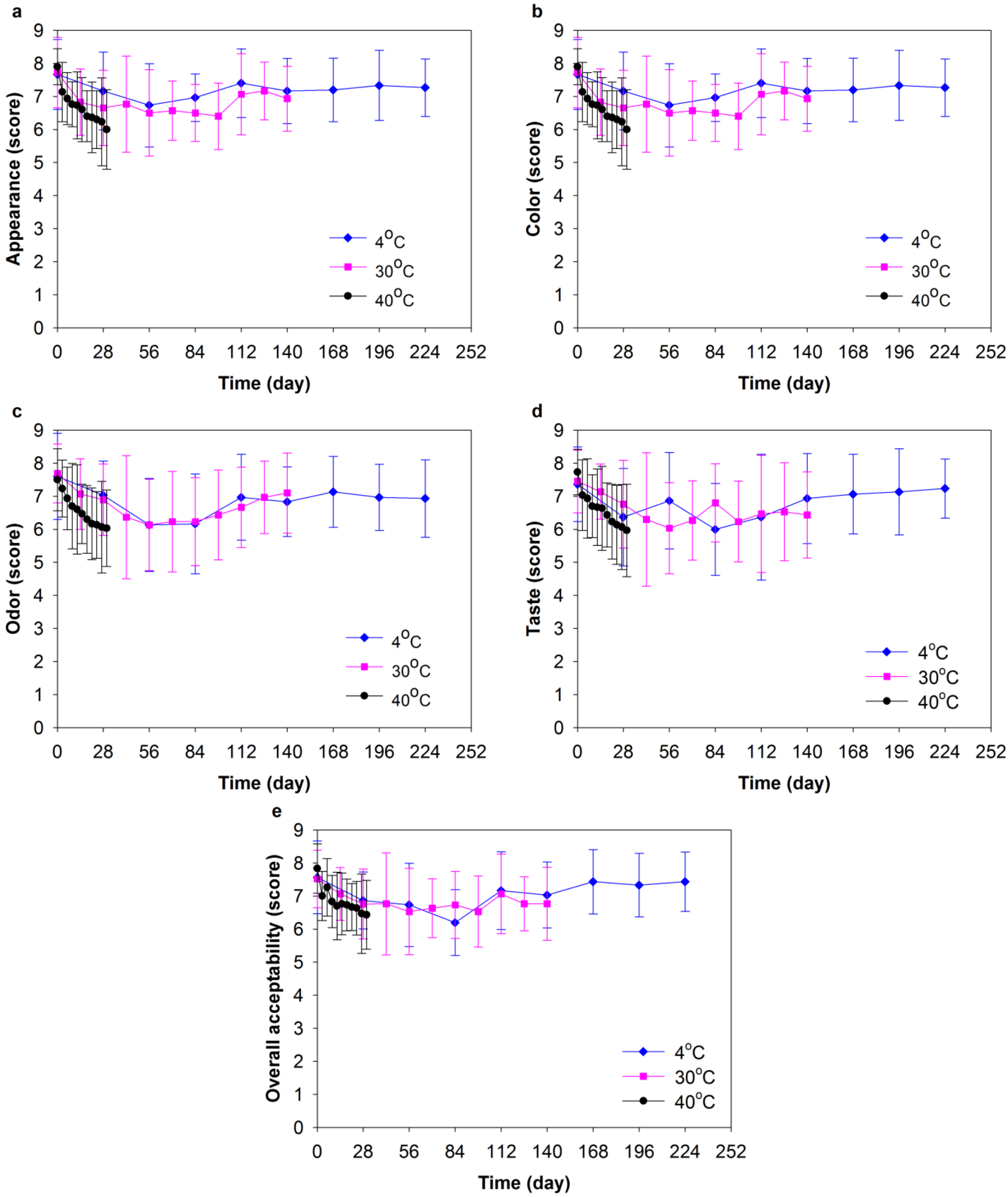

Figure 3. Average scores for sensory attributes of sukiyaki sauces during storage at different temperatures: **(a)** appearance, **(b)** colour, **(c)** odour, **(d)** taste, and **(e)** overall acceptability.

Table 2. Kinetic parameters of quality indices during storage of sukiyaki sauces at different temperatures obtained by Arrhenius and Ball models.

Parameter	Kinetic order	Temperature (°C)	k	R^2	RMSE	Arrhenius model			Ball model		
						E_a (kJ/mol)	k_0	R^2	Z (°C)	D_0 (day)	R^2
Sensory attribute											
Overall acceptability	Second	4	4.893×10^{-5}	0.095	0.533						
		30	1.611×10^{-4}	0.229	0.343	55.71	1.291×10^{6}	0.839	29.3	7.943×10^{5}	0.857
		40	1.137×10^{-3}	0.760	0.283						
Appearance	Second	4	9.805×10^{-5}	0.103	0.463						
		30	1.680×10^{-4}	0.010	0.570	32.19	99.963	0.756	50.5	3.243×10^{5}	0.778
		40	6.328×10^{-4}	0.814	0.153						
Colour	Second	4	5.383×10^{-5}	0.000	0.347						
		30	1.940×10^{-4}	0.043	0.531	59.60	7.602×10^{6}	0.841	27.4	7.288×10^{5}	0.859
		40	1.520×10^{-3}	0.896	0.262						
Odour	Second	4	9.536×10^{-5}	0.002	0.570						
		30	2.283×10^{-4}	0.088	0.584	45.69	3.299×10^{5}	0.799	35.7	3.715×10^{5}	0.819
		40	1.290×10^{-3}	0.961	0.122						
Taste	Second	4	6.172×10^{-5}	0.080	0.579						
		30	2.269×10^{-4}	0.349	0.403	56.98	2.859×10^{6}	0.863	28.7	6.101×10^{5}	0.881
		40	1.460×10^{-3}	0.928	0.208						
Physicochemical property											
TCD	Zero	4	0.016	0.237	1.138						
		30	0.035	0.616	1.032	37.61	1.709×10^{5}	0.842	43.5	2.046×10^{2}	0.860
		40	0.129	0.981	0.189						
BI	Second	4	2.716×10^{-7}	0.204	13.221						
		30	2.740×10^{-6}	0.037	19.473	77.80	1.110×10^{8}	0.958	21.1	1.470×10^{8}	0.968
		40	1.656×10^{-5}	0.952	4.410						

k: reaction rate constant (1/score·day for sensory attributes degradation, 1/unit·day for change in BI, unit/day for change in TCD); R^2: coefficient of determination; RMSE: root mean square error; E_a: activation energy; k_0: Arrhenius constant; Z: the required temperature for one $\log_{10} D$ value reduction; and D_0: D value at $T = 0$°C.

TCD of sukiyaki sauces followed a zero-order reaction, while the increase in BI values followed a second-order reaction. This finding was indicated by higher R^2 and lower RMSE as compared to the other reaction orders. The rates of TCD and BI in sukiyaki sauces increased with higher storage temperatures, as evidenced by the higher k values observed at elevated temperatures (Table 2). The zero-order kinetics for the increase in TCD during storage is consistent with a study on the storage of homogenised apple-based beikost (Palazon *et al*., 2009).

The Arrhenius and Ball models are widely used in the existing studies and have been successfully applied to estimate the deterioration of quality in food products (Zhang *et al*., 2021; Kim *et al*., 2022). The temperature dependence of k values for each quality index was further modelled by the Arrhenius and Ball equations (Eqs. 5 and 7). The kinetic parameters obtained by the Arrhenius and Ball models are illustrated in Table 2. Activation energy (E_a) represents the minimum amount of energy required for a chemical reaction to occur among the compounds involved, whereas the Z value indicates the temperature required to reduce the D value by tenfold (Kim *et al*., 2022). The results showed that the Arrhenius model could be used to describe changes in sensory attributes, namely appearance, colour, odour, taste, and overall acceptability, as indicated by R^2 value ranges of 0.756 - 0.863 (Figure 4a). Activation energies ranged from 32.19 to 59.60 kJ/mol and the Arrhenius constant (k_0) varied between 99.963 to 7.603×10^6 score^{-1}·day^{-1}. Besides, the activation energies for the increases of TCD and BI in sukiyaki sauce during storage at 4 - 40°C were 37.61 (R^2 = 0.842) and 77.80 kJ/mol (R^2 = 0.958), respectively (Figure 4b). The activation energies followed the order: $E_{a\ (BI)} > E_{a\ (color)} > E_{a\ (taste)} > E_{a(overall\ acceptability)} > E_{a\ (odour)} > E_{a\ (TCD)} > E_{a\ (appearance)}$. This indicated that the BI of sukiyaki sauce was more susceptible to changes in temperature than the other quality indices. Conversely, appearance preference was the least affected by an increase in temperature (Shukla *et al.,* 2020; Zhang *et al*., 2021).

The plotting of the Ball model demonstrated its applicability in illustrating the deterioration in sensory attributes, such as appearance, colour, odour, taste, and overall acceptability, as indicated by R^2 values ranging from 0.778 to 0.859 (Figure 4c). The Z values for the loss of sensory scores of sukiyaki sauce stored at 4 - 40°C ranged from 27.4 to 50.5°C. Additionally, the Ball model fitted well with the temperature dependence of D values for increases in TCD (R^2 = 0.860) and BI (R^2 = 0.968) of sukiyaki sauce during storage and the Z values were determined to be 43.5 and 21.1°C, respectively (Figure 4d). As observed, BI displayed a lower Z value and a higher E_a. This implied strong temperature dependence in the reactions, indicating that an increase in BI will occur slowly at lower temperatures and relatively faster at higher temperatures (Van Boekel, 2009; Kim *et al*., 2022). These results suggested that the Ball model exhibited better performance than the Arrhenius model, as evidenced by higher R^2 values. This agreed with the results reported in a previous study, which indicated better performance of the Ball model as compared to the Arrhenius model for describing the quality changes in kiwifruits during storage (Zhang *et al*., 2021).

The end of the shelf life of sukiyaki sauce was estimated using a threshold value of quality indices. For sensory characteristics, a hedonic score of 5 was used as the lowest limit of consumer acceptance based on responses by panellists (Tsironi and Taoukis, 2010; Shukla *et al.,* 2020). However, there were no specific threshold values for TCD and BI because these values vary depending on various factors such as storage temperatures, food products, and packaging conditions (Zhang *et al*., 2021).

Pearson's correlation analysis revealed that TCD showed a moderate negative correlation with overall acceptability (r = -0.561, $p < 0.01$) and colour scores (r = -0.493, $p < 0.01$). Likewise, BI exhibited a moderate negative correlation with overall acceptability (r = -0.426, $p \geq 0.01$) and colour scores (r = -0.635, $p < 0.01$) (Schober *et al*., 2018). These results indicated that TCD correlated more strongly with overall acceptability, whereas BI correlated more strongly with hedonic colour. Hence, using a threshold score of 5 for overall acceptability, the corresponding TCD threshold was 13.24 units. Likewise, with a colour score threshold of 5, the BI threshold was 396.58 units.

Table 3 presents the predicted shelf life of sukiyaki sauce based on protocol thresholds for quality indices. The models for shelf-life prediction of sensory characteristics and BI of sukiyaki sauce followed a second-order reaction, while TCD followed a zero-order reaction. The predicted shelf life based on each sensory attribute was almost

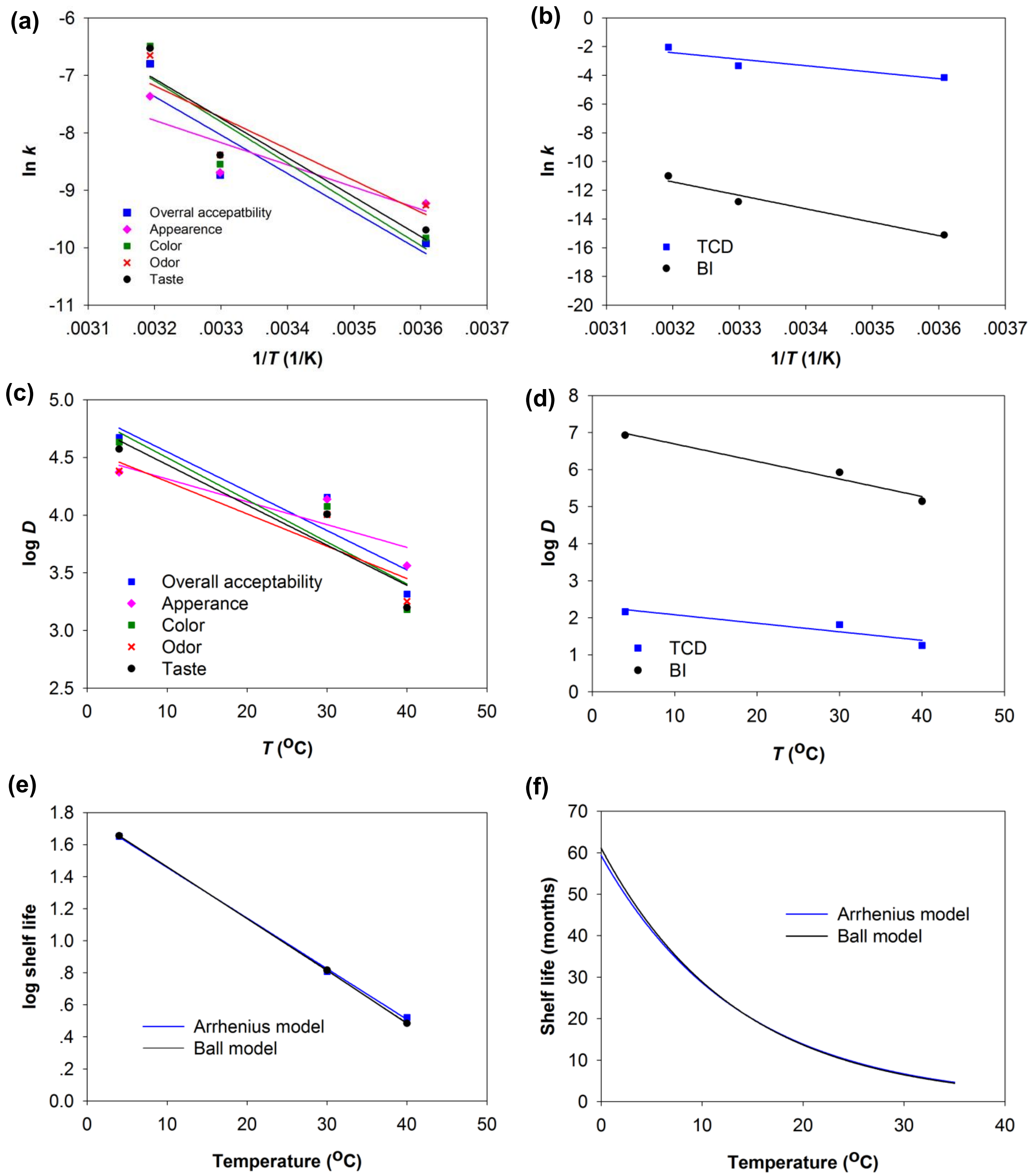

Figure 4. Arrhenius plot for sensory characteristics **(a)** and colour properties **(b)** of sukiyaki sauces as function of storage temperature. Ball plot for sensory characteristics **(c)** and colour properties **(d)** of sukiyaki sauces as function of storage temperature. Shelf-life plot of sukiyaki sauce stored at 4, 30, and 40°C **(e)**. Predicted shelf life of sukiyaki sauce based on multiple quality indices, associated with the Arrhenius and Ball models at different storage temperatures **(f)**.

Table 3. Shelf lives and predictive equations for quality indices of sukiyaki sauces stored at different temperatures.

Property	Parameter	Temperature (°C)	Arrhenius model: Predictive equation	Arrhenius model: Shelf life (months)	Ball model: Predictive equation	Ball model: Shelf life (months)
Sensory attribute	Overall acceptability	4	$\frac{\frac{1}{S_1}-\frac{1}{S_0}}{k_0 \exp\left[\frac{-E_a}{RT}\right]} = t_s$ where S_1 = 5; S_0 = initial score of each sensory attribute; T = temperature (K); E_a = activation energy (J/mol); t_s = shelf life (day).	56.3	$\frac{1}{S_1}-\frac{1}{S_0}\left(\frac{D_0 \times 10^{(-T/Z)}}{ln\,10}\right) = t_s$ where T = temperature (°C); D_0: D value at T = 0°C; t_s = shelf life (day).	58.0
		30		7.1		7.5
		40		3.5		3.4
	Appearance	4		27.7		27.9
		30		8.4		8.5
		40		5.6		5.4
	Colour	4		53.5		53.7
		30		5.8		6.0
		40		2.7		2.6
	Odour	4		28.3		28.4
		30		5.2		5.3
		40		2.9		2.8
	Taste	4		42.7		42.9
		30		5.1		5.3
		40		2.5		2.4
Physicochemical property	TCD	4	$\frac{TCD_1 - TCD_0}{k_0 \exp\left[\frac{-E_a}{RT}\right]} = t_s$ where TCD_1 = reject TCD (13.24), which is TCD at overall acceptability score of 5; TCD_0 = initial TCD (0).	31.6	$TCD_1 - TCD_0\left(\frac{D_0 \times 10^{(-T/Z)}}{ln\,10}\right) = t_s$	31.7
		30		7.8		7.1
		40		4.8		3.5
	BI	4	$\frac{\frac{1}{BI_1}-\frac{1}{BI_0}}{k_0 \exp\left[\frac{-E_a}{RT}\right]} = t_s$ where BI_1 = reject BI (396.58), which is BI value at colour score of 5; BI_0 = initial BI (298.4).	114	$\frac{1}{BI_1}-\frac{1}{BI_0}\left(\frac{D_0 \times 10^{(-T/Z)}}{ln\,10}\right) = t_s$	114.7
		30		6.3		6.7
		40		2.4		2.2

S: sensory score; TCD: total colour difference; and BI: browning index.

similar but slightly different from the predicted shelf life based on physicochemical properties such as TCD and BI. This was because the reaction rate of changes in these quality indices during storage differed (Zhang *et al*., 2021).

All quality indices served as crucial parameters for predicting the shelf life of sukiyaki sauce. Particularly, the appearance, colour, and browning of the product significantly influence consumers' purchasing decisions. In the present work, the multiple quality indices were further applied to estimate the shelf life of sukiyaki sauce. The average shelf-life value at storage temperatures of 4, 30, and 40°C for each quality index was calculated. A regression of the logarithm of shelf life obtained from the Arrhenius or Ball models versus temperature (°C) was plotted to derive the shelf-life models, as described by Eqs. 13 and 14, respectively (Figure 4e) (Palazon *et al*., 2009; Choosuk *et al*., 2022).

Arrhenius;

$$\log t_s = -0.0316T + 1.7732 \; (R^2 = 0.999) \quad \text{(Eq. 13)}$$

Ball;

$$\log t_s = -0.0325T + 1.7859 \; (R^2 = 1.000) \quad \text{(Eq. 14)}$$

where, T = storage temperature (°C), t_s = shelf life (in months) of sukiyaki sauce, with one month was equivalent to 30 days.

This suggested that the shelf-life plots obtained from both the Arrhenius and Ball models were suitable for predicting shelf life, as shown by the high value of R^2. From these equations, they were able to estimate the shelf life of the product at different storage temperatures (Figure 4f). For instance, the shelf life of this product at refrigerated (4°C) and room temperature (30°C) using the Arrhenius equation would be 44.3 and 6.7 months, respectively, while using the Ball equation, it would be 45.3 and 6.5 months, respectively. The results showed that the shelf life of sukiyaki sauce increased at lower storage temperatures due to slower deterioration in colour and browning, which affected sensory perception. This result agreed with previous reports that low-temperature storage could extend sauce shelf life. The duration varied from several months to one to three years, depending on their formulation, processing, and storage conditions (Yun *et al*., 2007; Lee *et al*., 2015; Zardetto and Barbanti, 2020).

In summary, the result suggested that sukiyaki sauce could be stored at 30°C for approximately 6.5 - 6.7 months, during which the product remained sensorially acceptable and maintained physicochemical properties such as TCD and BI throughout the storage duration. Nevertheless, the shelf life of a product depends on temperature variations in real-life situations. These variations occur during the manufacturing process in areas such as the packaging facility and warehouse, throughout the supply chain, within shopping facilities and in customer storehouses (Shukla *et al*., 2020).

Model validation

The accuracy of the kinetic models in predicting experimental data for changes in scores of all sensory attributes and values of TCD and BI was validated. The results showed that the Arrhenius model reasonably fitted the experimental data for sensory attributes, including overall acceptability, appearance, colour, odour, and taste, as well as TCD and BI, with low RMSE values ranging from 0.475 - 0.599, 0.239 - 0.693, 0.353 - 0.858, 0.379 - 0.859, 0.458 - 0.699, 0.705 - 2.038, and 12.010 - 24.283, respectively. Additionally, the fitting of the Ball model with the experimental results of sensory attributes, including overall acceptability, appearance, colour, odour, and taste, as well as TCD and BI, gave low RMSE intervals of 0.467 - 0.571, 0.229 - 0.683, 0.353 - 0.822, 0.358 - 0.834, 0.430 - 0.688, 0.664 - 1.932, and 10.786 - 23.059, respectively.

Conclusion

Overall, the results indicated that variations in the TCD, BI, and sensory characteristics of sukiyaki sauce during storage were strongly dependent on storage conditions. Higher temperatures led to progressively negative impacts on these quality indices. The Arrhenius and Ball models were found to adequately describe the kinetic constants as a function of temperature. When combined with a second-order kinetic reaction, the Arrhenius and Ball models could be used to predict the shelf life of sukiyaki sauce based on sensory attributes and BI, while a zero-order kinetic reaction was applied for shelf-life prediction based on TCD. According to the shelf-life models based on multiple quality indices, the shelf life of sukiyaki sauce at room temperature (30°C) was estimated to be approximately 6.5 - 6.7 months. This finding can assist distributors in deciding stock and sales strategies as well as consumers in determining the optimum edible duration.

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