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Development of Respiration Models for Modified Atmosphere Packaging of Horticultural Commodities

Yoshio MAKINO*

Department of Postharvest Technology, Kagawa Prefectural Food Research Institute
(Goto, Takamatsu, Kagawa, 761-8031 Japan)

Abstract

Oxygen uptake models for horticultural commodities were constructed based on the Langmuir adsorption theory, mechanism of depression of respiration caused by CO_2 and transition state theory in order to design modified atmosphere packaging (MAP) systems. To examine the usefulness of the models, O_2 uptake rate data fro the commodities were applied to the models. The models were found to be suitable for describing the respiration of several kinds of commodities. Mathematical analysis of MAP systems for shredded lettuce and shredded cabbage was conducted using the proposed rate equations and mass balance equations. The simulated results were in agreement with the experimental date. The O_2 uptake models were found to be suitable for the design of the MAP systems.

Discipline: Postharvest technology
Additional key words: lettuce, cabbage, mathematical model, MAP

Introduction

In the storage and/or the transportation process of horticultural commodities, respiration control plays an important role in prolonging the postharvest life of the commodities. Decrease of O_2 and increase of CO_2 concentrations in the environment surrounding the commodities are effective for the depression of respiration[11]. A modified atmosphere packaging (MAP) system is generally adopted for controlling the gas environment[26]. The system creates an optimum composition of O_2 and CO_2 suitable for the storage of the commodities by controlling the respiration and film permeability.

Investigations on the prediction of the dynamics of atmosphere in various MAP systems have been undertaken since the 1960s[10]. Respiration models of the commodities used for the prediction were constructed by empirical approaches except for a model by Lee et al.[15]. It is thus necessary to develop a respiration model on a theoretical basis.

In the present study, theoretical models were constructed for representing the relationship between O_2 uptake rate in the commodities and O_2, CO_2 and temperature. Suitability of the derived models was evaluated using respiration data for various kinds of commodities. Mathematical analysis of the MAP systems for shredded lettuce and shredded cabbage was conducted to analyze the suitability of the models.

Mathematical models

1) Respiration models

Respiration is controlled by an elaborate interlocking system of feedback control that coordinates the rates of glycolysis, fatty acid breakdown, the citric cycle and electron transport[1]. In the biochemical reaction of O_2 uptake in a unit cell, it is assumed that one molecule of O_2 is adsorbed on an active site of the cytochrome oxidase complex buried in the inner membrane of mitochondria, and that the O_2 molecule is desorbed from the site when it accepts 4 electrons, which react with the O_2 molecule, and is transformed to 2 molecules of H_2O[25]. The

Present address:
* Kagawa Prefectural Fermentation and Food Experimental Station (Nohma, Uchinomi, Shohzu, Kagawa, 761-4421 Japan)

cytochrome oxidase reaction is estimated to account for 90% of the total O_2 uptake in most cells[1]. To analyze these biochemical reactions mathematically, a very complicated model is required. From a practical point of view, a simplified expression is desirable for predicting the atmospheric conditions within the package. An organism, for example a horticultural commodity, takes up an O_2 molecule by a chemical adsorptive reaction on an active enzyme site[25]. Cytochrome oxidase complex adsorbs one molecule of O_2 per active site[1]. I therefore attempted to construct a practical model for O_2 uptake in horticultural commodities based on the adsorption theory proposed by Langmuir[14].

$$J_o = \frac{abp_o}{1 + ap_o} \qquad (1)$$

It is generally assumed that in the respiratory depression by CO_2, the CO_2 molecule induces the metabolic inhibition of organic acids in the TCA cycle which is one of the most important steps in the respiratory process[4,8,19]. The inhibition decreases the formation of NADH and $FADH_2$ which are produced from NAD+ and FAD. As a result, the desorption of the O_2 molecule in the final step of the electron transport chain is indirectly reduced[2]. Eq. 1 is modified to rate Eq. 2 for O_2 uptake[17] as follows:

$$J_o = \frac{abp_o}{1 + ap_o + aip_o p_c} \qquad (2)$$

On the basis of the transition state theory[7,18], the maximum O_2 uptake rate b in Eq. 1 or 2 is expressed as follows:

$$b = \frac{NkT}{Lh} e^{-\frac{\Delta G}{RT}} \qquad (3)$$

When the temperature dependence of the rate parameters a and i is assumed to be much lower than the dependence of b, the O_2 uptake rate of a horticultural commodity for storage is calculated using Eqs. 1 (or 2) and 3.

2) Respiration rate parameters

Eq. 1 is modified to a linear form given as follows:

$$\frac{p_o}{J_o} = \frac{1}{ab} + \frac{1}{b} p_o \qquad (4)$$

The values of a and b are obtained from the slope b^{-1} and the intercept $(ab)^{-1}$ of $p_o \cdot J_o^{-1}$ vs p_o plots.

Eq. 2 can be modified to a linear form as given below:

$$\frac{1}{J_o} = \frac{1}{b} + \frac{1}{abp_o} + \frac{i}{b} p_c \qquad (5)$$

Equations for the calculation of the parameters a, b and i are derived from Eq. 5 as follows[17]:

$$a = \frac{C_1 p_1 - C_2 p_2}{p_1 p_2 (C_2 - C_1)} \qquad (6)$$

$$b = \frac{p_1 - p_2}{C_1 p_1 - C_2 p_2} \qquad (7)$$

$$i = \frac{B(p_1 - p_2)}{C_1 p_1 - C_2 p_2} \qquad (8)$$

A linear form of Eq. 3 is given as follows:

$$\ln\left(\frac{b}{T}\right) = -\frac{\Delta G}{RT} + \ln\left(\frac{NK}{Lh}\right) \qquad (9)$$

3) Changes in atmosphere in a MAP system

Mass balance equations of O_2, CO_2 and N_2 gases in a MAP system are expressed by the following equations[10]:

$$\frac{dv_o}{dt} = \frac{P_o A}{X}(q_o - p_o) - J_o W \qquad (10)$$

$$\frac{dv_c}{dt} = \frac{P_c A}{X}(q_c - p_c) + J_c W \qquad (11)$$

$$\frac{dv_N}{dt} = \frac{P_N A}{X}(q_N - p_N) \qquad (12)$$

Temperature dependence of a gas permeability coefficient of a polymeric film is expressed by the Arrhenius equation as follows[22]:

$$P_i = F_i e^{-\frac{E_i}{RT}} \qquad (13)$$

Respiratory quotient (RQ) is defined as follows:

$$Q = \frac{J_c}{J_o} \qquad (14)$$

Atmospheric change with time in a MAP system included in a practical temperature range for storage of horticultural commodities is simulated by solving Eqs. 1 (or 2), 3 and 10–14 simultaneously by the Runge-Kutta method using a microcomputer.

Materials and methods

1) Horticultural commodities

Head cabbage (variety YR-Aoba), head lettuce (variety Cisco), tomatoes (variety Momotaro) and broccoli (variety Naomidori) were purchased from a wholesale market in Takamatsu (Kagawa Prefecture, Japan).

Head cabbage was shredded into 1 mm wide slices with a cooking cutter (CQ-34R, Toshiba, Inc., Tokyo, Japan). The shredded cabbage was washed twice in a stainless-steel pan and rinsed for 1 min using a water sprinkler. The water adhering to the surface of the cabbage was removed by centrifugation (64.1 m s^{-2} for 30 s). Head lettuce was shredded into 30 × 30 mm (900 mm^2) fragments with a kitchen knife. The shredded lettuce was treated by the same method as the shredded cabbage after shredding.

2) Measurement of O_2 uptake and CO_2 production rates

Oxygen uptake and CO_2 production rates of horticultural commodities were measured by the method of Jurin & Karel[10] under gaseous environments including the combination of 5 levels of O_2 [2, 5, 10, 15 and 21% (partial pressures of 2.03, 5.07, 10.1, 15.2 and 21.3 kPa under 101.325 kPa)] and 3 levels of CO_2 [0, 3, and 9% (partial pressures of 0, 3.04 and 9.12 kPa under 101.325 kPa)] balanced with N_2.

3) Measurement of O_2 and CO_2 concentrations in a MAP system

A low density polyethylene (LDPE) pouch (thickness 0.025 mm, effective area 0.072 m^2) was used for the MAP test with shredded cabbage or shredded lettuce. Oxygen, CO_2 and N_2 permeability coefficients of LDPE were determined by the method of Makino & Hirata[16] at 5, 15, 20 or 30°C. The data of the permeability coefficients were analyzed by linear regression using the linear form of Eq. 13 [$\ln(P_i) = -E_i \cdot (RT)^{-1} + \ln(F_i)$].

The prepared shredded cabbage (0.06 kg) and shredded lettuce (0.08 kg) were enclosed in LDPE pouches. When shredded lettuce was enclosed, a small package of CO_2 scrubber (Ageless C®, Mitsubishi Gas Kagaku, Inc., Tokyo, Japan) was also enclosed in the pouch because it is desirable that lettuce be stored under 0% CO_2[5]. The pouch was stored for 3 d (shredded cabbage) or 6 d (shredded lettuce) at 10°C. The void volume of the pouch was determined based on water displacement. Oxygen and CO_2 concentrations within the pouch were periodically measured during storage by gas chromatography.

Data for the changes in the gas concentrations in the LDPE pouch over time were compared with the simulated values to estimate the practical effectiveness of the model equations proposed in the present study.

Results and discussion

1) Suitability of the O_2 uptake model for respiration in horticultural commodities

Fig. 1 shows the relationship between $p_o \cdot J_o^{-1}$

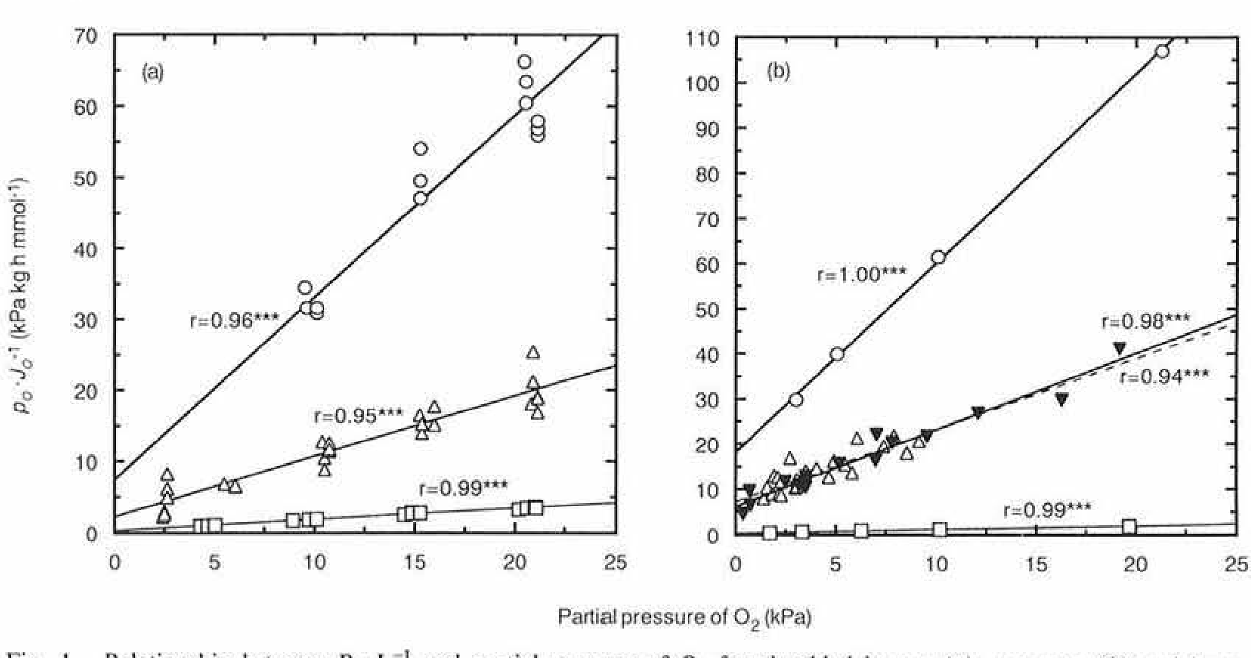

Fig. 1. Relationship between $P_o \cdot J_o^{-1}$ and partial pressure of O_2 for shredded lettuce (Δ), tomatoes (○) and broccoli (□) based on the experimental data obtained in this study (a) and for apples (○), broccoli (□), bananas (▼) and blueberries (△) based on the published data (b)

Solid lines denote the linear regression lines. A dashed line denotes the linear regression line for blueberries.

and p_o of the experimental data of O_2 uptake for shredded lettuce, tomatoes and broccoli, and of the published data for apples[6], broccoli[15], bananas[13], and blueberries[3] with linear regression lines. The correlation coefficients were found to be in the range of 0.94 to 1.00, which were significant at 99.9% level of Fisher's z-transformation method, suggesting that Eq. 1 may be applied for the prediction of the O_2 uptake rate of many kinds of commodities. Eq. 1 is a simplified mathematical form of the enzyme kinetic model proposed by Lee et al.[15]. The validity of this mathematical form has already been demonstrated by Lee et al.[15]. In this study, I observed that the same mathematical equation form can be derived using either the enzyme kinetic theory or the adsorption theory, as the control mechanism.

Fig. 2 shows the relationship between J_o^{-1} and p_c in the experimental data of O_2 uptake for shredded cabbage, tomatoes, and broccoli in this study, and those in the published data for broccoli[15]. The value of J_o^{-1} for broccoli obtained in this study was larger than that obtained in the literature[15], presumably due to the difference in the experimental temperature: 16°C in this experiment, and 24°C in the literature. Kader et al.[11] reported that temperature and/or cutting treatment affect(s) the respiration rate of horticultural commodities. The correlation coefficients between the experimental data and the straight fit lines were found to be in the range of 0.587 to 0.902, values significant at 95% level of the Fisher's z-transformation except for one coefficient. These findings suggest that Eq. 2 may be applied for the prediction of the O_2 uptake rates of the 3 kinds of horticultural commodities. The O_2 uptake data from the literature[15] for broccoli at 12.7 kPa of O_2 partial pressure resulted in a correlation coefficient which was not significant at the 95% level. However, the experimental data substituted in Eq. 5 were obviously linear as shown in Fig. 2.

The validity of Eq. 2 as a rate equation was demonstrated in the present study as shown in Fig. 2. Eq. 2 is a simplified mathematical form of the enzyme kinetic model with noncompetitive inhibition. The validity of this mathematical form has already been demonstrated by Lee et al.[15] for the aerobic respiration of horticultural commodities. In this study, I observed that the same mathematical form equation describing the control mechanism can be derived using either the enzyme kinetic theory or the adsorption theory. The noncompetitive inhibition is defined as the direct inhibition of the enzymic activity where an inhibitor binds to the enzyme-substrate complex but does not bind to the free enzyme[24]. Lee et al.[15] approximated the respiration rate of horticultural commodities with an noncompetitive inhibition model. However, many authors concluded that the mechanism for CO_2 inhibition of O_2 uptake in some way interferes with organic acid metabolism,

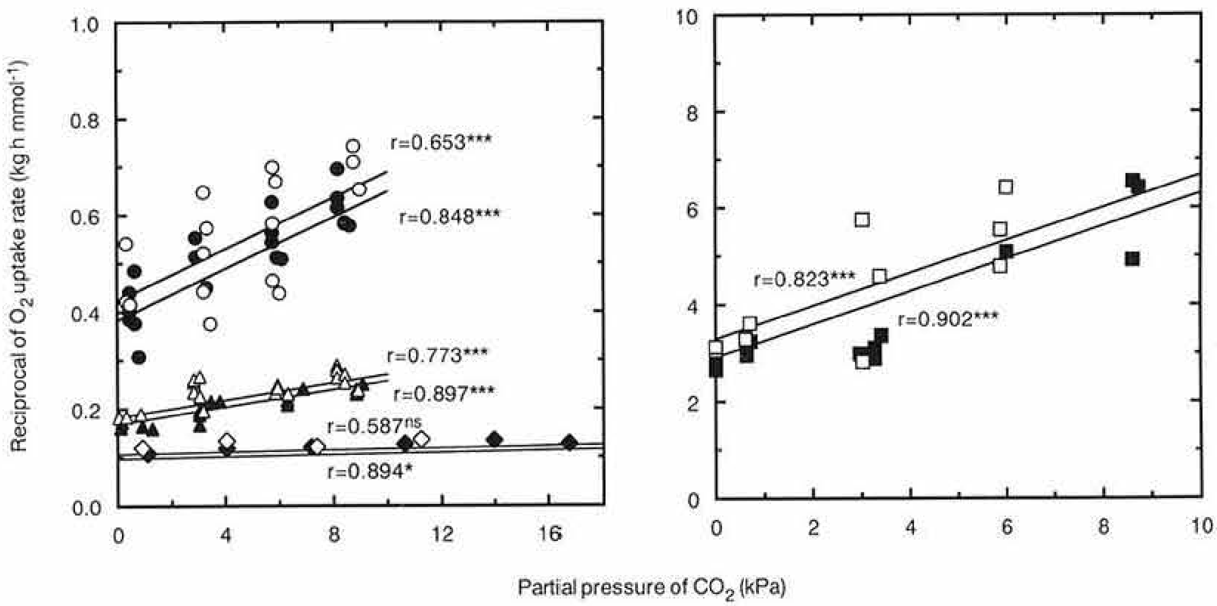

Fig. 2. Relationship between reciprocal of O_2 untake rate and partial pressure of CO_2 for shredded cabbage (○), broccoli (△) and tomatoes (□) based on the experimental data obtained in this study and for broccoli (◇) in the literature[15]

Open and closed symbols denote the data at 2.03 and 21.3 kPa O_2, respectively.

which is an indirect effect[19]. Such indirect effects are not compatible with mathematical modeling of respiration rate equations at a mechanistic level. The macroscopic approach, such as adsorption theory, which was applied to derive Eq. 2 may be more suitable for expressing complex biochemical reactions.

The O_2 uptake rate data for shredded cabbage and shredded lettuce obtained in this study and for blueberries and raspberries obtained from the literature[3,9] are shown in Figs. 3 and 4 with

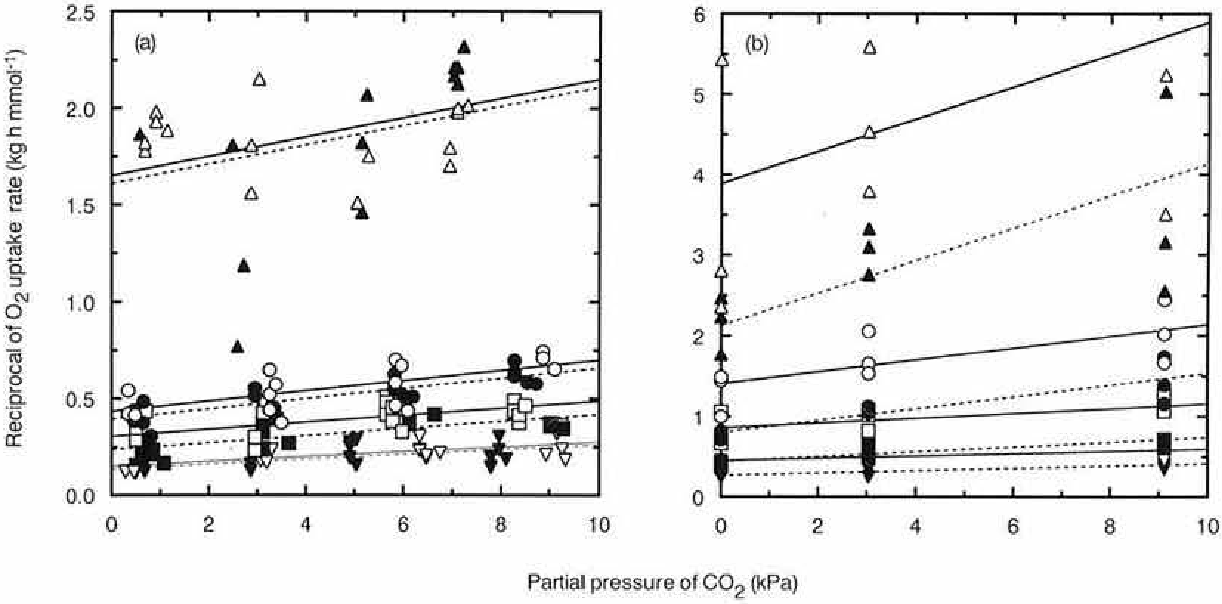

Fig. 3. Relationship between reciprocal of O_2 uptake rate and partial pressure of CO_2 for shredded cabbage (a) and shredded lettuce (b) at 5 (△), 15 (○), 20 (○) and 30°C (▽)
Open and closed symbols denote the data at 2.03 and 21.3 kPa O_2, respectively, on the basis of the experimental data obtained in this study. Solid and dashed lines denote the lines generated by the method of Makino et al.[17] in reference to the O_2 partial pressure of 2.03 and 21.3 kPa, respectively.

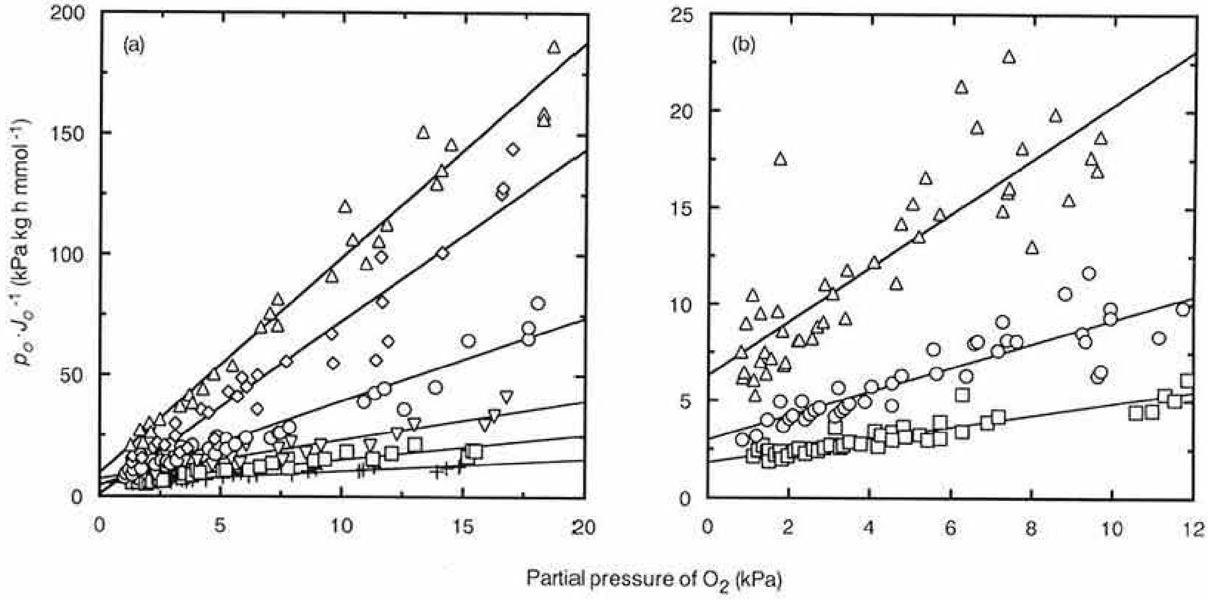

Fig. 4. Relationship between $p_o \cdot J_o^{-1}$ and partial pressure of O_2 for blueberries (a) and raspberries (b) at 0 (△), 5 (◇), 10 (○), 15 (▽), 20 (□) and 25°C (+) on the basis of the data obtained in the literature[3,9]
Solid lines denote the linear regression lines.

calculated linear regression lines. The uptake rate rose with the increase of O_2 partial pressure, decrease of CO_2 partial pressure, and rise of temperature in the environment surrounding the commodities. The trend was in agreement with that described in the previous report[12], supporting the assumption that the O_2 uptake rate data obtained in this study are suitable for the analysis of the temperature dependence of the O_2 uptake rate of the commodities.

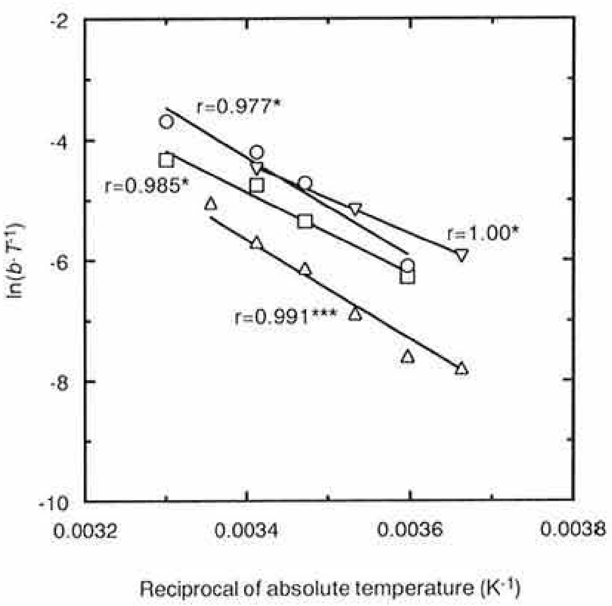

Fig. 5. Relationship between $\ln(b \cdot T^{-1})$ and reciprocal of absolute temperature for shredded cabbage (○), shredded lettuce (□), blueberries (△) and raspberries (▽)
Solid lines denote the linear regression lines.

Values of $\ln(b \cdot T^{-1})$ calculated from the values of the maximum O_2 uptake rate parameter b for various products are shown in Fig. 5. The results indicate that the values of $\ln(b \cdot T^{-1})$ were linearly correlated with the reciprocal of absolute temperature. These observations suggest that Eqs. 3 and 9 can be used for the analysis of the temperature dependence of the O_2 uptake rate of horticultural commodities. In the previous reports, Karel and Go[13] and Song et al.[23] applied the Arrhenius equation to express the respiration rates of pre-climacteric hard green bananas, cut broccoli and blueberries, respectively. I applied Eq. 3 derived from the transition state theory. The correlation coefficients in Fig. 5 indicate that the proposed Eq. 3 is suitable for the O_2 uptake rate. The proposed Eq. 3 is composed of Boltzmann's constant k, Planck's constant h, Avogadro's constant L and gas constant R which are generally adopted in the physicochemical field except for the symbol N. These findings suggest that Eq. 3 is more suitable than the Arrhenius equation for describing the temperature dependence of the O_2 uptake rate from a theoretical viewpoint.

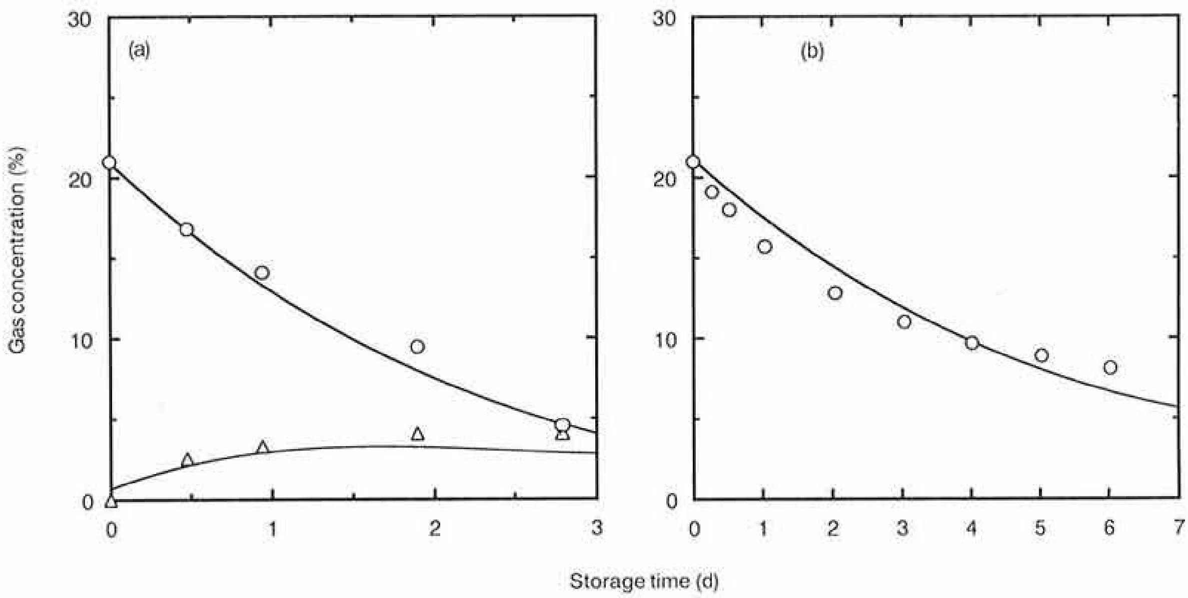

Fig. 6. Measured changes in concentrations of O_2 (○) and CO_2 (△) in modified atmosphere packing (MAP) systems for shredded cabbage (a) and shredded lettuce (b)
Simulated results are indicated by the full lines. Initial amount of O_2, CO_2 and N_2 molecules in the systems are 3.16, 0 and 11.7 mmol (a) and 7.37, 0 and 27.4 mmol (b), respectively. Partial pressures of O_2, CO_2 and N_2 outside of the systems are 21.3, 0 and 79.0 kPa, respectively.

Table 1. Oxygen uptake rate parameters for horticultural commodities

Commodity	ΔG	N	$a^{a)}$	$i^{a)}$	$Q^{a)}$
Shredded cabbage	8.81×10^8	7.74×10^{18}	3.09	0.0691	0.853
Shredded lettuce	7.28×10^8	3.50×10^{16}	0.0532	0.0838	0.962

a): Average in the range of 5 to 30°C.

Table 2. Gas permeability parameters for a low density polyethylene film

Gas	E_i	F_i
O_2	4.74×10^8	5.42
CO_2	4.81×10^8	44.8
N_2	3.74×10^8	0.126

2) Simulation of dynamic changes in atmosphere in MAP systems for horticultural commodities

Experimental data and simulation results for the changes in the O_2 and CO_2 concentrations over time in the MAP systems for shredded cabbage and shredded lettuce are shown in Fig. 6. The simulation results obtained by using the parameters presented in Tables 1 and 2, indicated by the full lines, were in good agreement with the experimental data. These facts suggest that the model derived from the Langmuir adsorption theory, mechanism of the depression of respiration caused by CO_2 and the transition state theory is suitable for expressing the O_2 uptake rate of horticultural commodities as well as for the evaluation of the parameters, gas permeability coefficients and *RQ*.

Morales-Castro et al.[20,21] calculated the changes in the O_2 and CO_2 concentrations over time in the MAP systems for sweet corn and head lettuce using respiration models suitable for changes in temperature. The respiration models were constructed by the non-linear regression analysis which is an empirical approach. Deviation of the experimental results from the simulated results was larger than that in this study. It is suggested that the simplified model proposed in this study is more effective for the design of the MAP systems than the model(s) reported in the literature.

Conclusion

An O_2 uptake model based on Langmuir adsorption theory was found to be suitable for the analysis of the actual data of O_2 uptake of shredded lettuce, tomatoes and broccoli obtained in this study and of apples, broccoli, bananas and blueberries obtained in the literature. An O_2 uptake model under an atmosphere with CO_2 based on the assumption that CO_2 gas inhibits the oxidation of the organic acids in the TCA cycle predicted well the actual rates for shredded cabbage, tomatoes and broccoli. The validity of the transition state theory for explaining the temperature dependence of the O_2 uptake rate of shredded cabbage, shredded lettuce, blueberries and raspberries was demonstrated in this study. Simulation of changes in O_2 and CO_2 concentrations over time in a MAP system for shredded cabbage or shredded lettuce was in good agreement with the experimental data. The model equations proposed in this study can thus be used for the design of MAP systems.

Notations

- a: O_2 uptake rate parameter (kPa^{-1})
- A: effective area of a packaging film (m^2)
- b: maximum O_2 uptake rate (mmol kg^{-1} h^{-1})
- B: constant (kg h mmol^{-1} kPa^{-1})
- C: constant (kg h mmol^{-1})
- E: energy of activation for gas permeation (m^2 kg h^{-2} mmol^{-1})
- F: frequency constant (mmol m^{-1} h^{-1} kPa^{-1})
- h: Planck's constant 3.976×10^{-32} m^2 kg h^{-1}
- i: O_2 uptake rate parameter (kPa^{-1})
- J: uptake or production rate of a gas in a horticultural commodity (mmol kg^{-1} h^{-1})
- k: Boltzmann's constant 4.97×10^{-20} m^2 kg h^{-2} K^{-1}
- L: Avogadro's constant 6.022×10^{20} molecules mmol^{-1}
- N: total number of active sites for O_2 uptake in a horticultural commodity (molecules kg^{-1})
- p: partial pressure of a gas in the environment surrounding a horticultural commodity (kPa)
- P: permeability coefficient of a gas through a plastic film (mmol m^{-1} h^{-1} kPa^{-1})
- q: partial pressure of a gas outside of a MAP system (kPa)
- Q: respiratory quotient (*RQ*)
- R: universal gas constant 1.08×10^5 m^2 kg h^{-2} mmol^{-1} K^{-1}
- t: storage time (h)

T: absolute temperature (K)
v: amount of a gas in a MAP system (mmol)
W: mass of fresh produce (kg)
X: thickness of a packaging film (m)
ΔG: Gibbs energy of activation for O_2 uptake (m^2 kg h^{-2} $mmol^{-1}$)

Subscripts
C, N, O: CO_2, N_2, O_2
i: symbol expressing C, N or O
1, 2: level of O_2 pressure

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