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Permeability of edible coatings

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Abstract The permeabilities of water vapour, O_2 and CO_2 were determined for 18 coating formulations. Water vapour transmission rate ranged from 98.8 g/m^2.day (6% beeswax) to 758.0 g/m^2.day (1.5% carboxymethyl cellulose with glycerol). O_2 permeability at 14 ± 1°C and 55 ± 5% RH ranged from 1.50 to 7.95 cm^{3}cm cm^{-2}s^{-1}Pa^{-1}, with CO_2 permeability 2 to 6 times as high. Permeability to noncondensable gases (O_2 and CO_2) was higher for hydrophobic (peanut oil followed by beeswax) coatings as compared to hydrophilic (whey protein concentrate and carboxymethyl cellulose).

Keywords Edible coatings · Water vapour · O_2 · CO_2 · Permeability

Introduction

Edible coatings are developed for fresh commodities to control migration of moisture, exchange of gases such as O_2, CO_2 and C_2H_4, which influence the quality and shelf life of commodities. The most studied property of coated fruit is its weight loss during storage (Farooqi et al. 1988, Paull and Chen 1989, Cohen et al. 1990, Moller et al. 2004, Chauhan et al. 2005). Coatings have also been studied in relation to spoilage, especially chilling injury and browning. Prevention of spoilage was sometimes attributed to adjuncts, such as fungicides or bioregulators, but more often to the diffusion barrier formed by the coating. The barrier hinders O_2 and CO_2 diffusion, thus reducing the respiration rate (Banks 1984, 1985, Erbil and Muftugil 1986, Farooqi et al. 1988, Saftner 1999). Coatings also prevent spoilage by serving as a barrier to water vapour (Morris 1982).

A principal disadvantage of wax coatings is the development of off-flavours from their use (Tewari et al. 1980, Cuquerela et al. 1981, Krishnamurthi and Kushalappa 1985, Chen and Paull 1986, Erbil and Muftugil 1986, Dhalla and Hanson 1988, Farooqi et al. 1988, Paull and Chen 1989, Cohen et al. 1990). In general, with the exceptions of appearance and lubrication (Lidster 1981, Mellenthin et al. 1982), the literature shows that the effects of coating are directly related to gas exchange between fruit and its environment. However, the literature provides no information on the permeability properties of fruit coatings except for a few estimates made for permeance of coated vs. uncoated fruits (Banks 1984, Ben-Yehoshua et al. 1985, Paull and Chen 1989, Perez-Gago et al. 2003, Chauhan et al. 2005, Togrul and Arshan 2005) or based on relative values from storage of the same commodity with different coatings (Cuquerela et al. 1981, Rohrbach and Paull 1982). Due to lack of data on permeability in general, the considerable literature on coatings is of little value in predicting whether a coating used for one purpose is suitable for another. Therefore, determination of permeability of some coatings' components and how a coating's permeability may be used to predict its performance was carried out.

Materials and methods

Carboxymethyl cellulose (CMC), glycerol, glycerol monostearate (GMS) and Tween 20 (surfactant) were procured from Hi media, Mumbai, India and whey protein concentrate (WPC) from Mahaan Protein, Kosi Kalan, Mathura, India, peanut oil (PO) from Nature fresh, Delhi, India and beeswax (BW) from Gulzar Honey, Hisar, India. Cellulose acetate film, low density polyethylene (LDPE) bags and

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corrugated fibreboard boxes were purchased from the local market, Hisar, India.

Preparation and selection of coatings: Eighteen coating formulations were prepared: 6 were hydrophobic having 3 concentrations each of PO (5, 10 and 15%) and BW (2, 4 and 6%) and 6 concentrations each of hydrophilic ones having 3 with and 3 without glycerol of WPC (5, 10 and 15%) and CMC (0.5, 1.0 and 1.5%). GMS was used as an emulsifier for hydrophobic coatings in 1:2 (for 15% PO and 6% BW) and 1:1.5 (for 5 and 10% PO and 2 and 4% BW) ratios by weight of the base material and homogenized using hot water at 3000 rpm for 30 sec to form emulsion. Glycerol was used as a plasticizer at 15% level in hydrophilic coatings and the solution made was heated till 90°C for 30 min. Water was used as a solvent for preparation of various coatings. Before coating, all the formulations were cooled down to ambient temperature and Tween 20, as a surfactant/ wetting agent, was added at 1.0% in all the coating formulations. All the coatings were tested for water vapour transmission rate (WVTR), viscosity and O_2 and CO_2 permeability.

Uniform and healthy guava (Psidium guajava L) fruits of cv Hisar 'Safeda' were harvested at green mature stage with the help of a sharp secateure, leaving a small pedicle intact on the fruit in early December from the Horticultural Farm, CCS Haryana Agricultural University, Hisar. Four coatings viz. WPC (5%) with glycerol, CMC (1%) with glycerol, PO (5%), BW (4%) were selected based on their effect on the percent weight loss and sensory quality for guava fruits. There were 8 fruits per pack (~800 ± 60 g) and 6 replicates per treatment. Guava fruits were coated by either soft brush or dipping for 2 min depending on the viscosity of coating materials. Excess coating was allowed to drain off onto paper towels. The coated commodities were air-dried and kept in corrugated fiberboard boxes. The fruits were stored for 12 days at ambient (16 ± 1ºC, 60 ± 5% RH) and 21 days at refrigerated (10 ± 1ºC, 65 ± 5% RH) conditions. Various observations were recorded on every 4th day under ambient and on every 7th day under refrigerated storage conditions. The controls used for comparing the performance of the coated fruits were:

Control I: uncoated fruits packed in corrugated fiber boxes with newspaper lining,

Control II: uncoated fruits packed in 0.5% perforated LDPE (400 gauge) bags and,

Control III: uncoated fruits packed in unperforated LDPE (400 gauge) bags.

The WVTR was determined by a static method. Here wide-open mouth vials containing anhydrous $CaCl_2$ were closed with coated (using various edible coatings) cellulose acetate and plastic film. The thickness (measured by microguage) of the coatings was 1.4 micron. Wax was used to make it an airtight unit. Vial closed with uncoated cellulose acetate film was used as a control. All the prepared vials were then kept in desiccators containing saturated KNO_3 solution. Further, these desiccators were kept at 38ºC in the incubator for maintaining 90% RH. WVTR was expressed as $g/m^2.d$ by observing the change in weight per unit time.

Viscosity of the coatings was determined by using viscometer (Brookefield Viscometer, MA, USA) and expressed in centipoises (mPa.s). In order to avoid turbulence various spindles and spindle speeds used for different coating formulations were:

1) WPC: spindle no 02 and spindle speed 50
2) CMC: spindle no 02 and spindle speed 50 and 10 (depending on consistency)
3) PO: spindle no 07 and spindle speed 10
4) BW: spindle no 07 and spindle speed 10

Permeability was measured by coating LDPE bags of known permeability with various coating materials using a brush. The thickness of the coatings was 1.4 micron. The bags were flushed with CO_2 and sealed using a gas-flushing machine. The initial concentrations of CO_2 and O_2 were noted, thereafter, every hour for 5 h the concentrations were observed in the polythene bags using a gas analyzer (Model 1902D, Quantak, USA). The experiment was carried out at ambient conditions (14 ± 1°C, 55 ± 5% RH). The reported permeability values are mean of 3 samples. The gas (O_2 and CO_2) transmission rate was expressed as $cm^3cm\ cm^{-2}s^{-1}Pa^{-1}$.

Respiration rate of the fruits during storage were determined as per the headspace analysis procedure adopted by Banks (1984, 1985) using gas liquid chromatograph fitted with chromosorb-101 column and thermal conductivity detector. The flow rate of carrier gas (nitrogen) was 18 ml/min, oven temperature 100°C and injector and detector temperature 120°C. Respiration rate was expressed as mg CO_2/h/kg fruit. Three replicates from each group of guava fruits were tested at both the storage conditions and respiration rate was tested on 3 randomly selected fruits. Initial baseline values of respiration were established on zero day of the test period using 3 samples of guava.

A mathematical model was developed to determine the respiration rate of uncoated and coated guava fruits (coated with the selected edible coatings) in terms of ml of CO_2/h/kg as a function of the storage time at ambient and refrigerated conditions. The rate of CO_2 evolved at any time from the guava fruit was assumed to be function of surrounding atmosphere and permeability properties of the coatings. The model structure developed was

$$R\ (Cc) = \beta_o + \beta_1\ T + \beta_2\ T^2 \qquad \ldots(1)$$

where, Cc is the concentration of CO_2 (ml CO_2/h/kg fruit) in fractions and T is the storage time. The CO_2 concentrations were plotted against time and this concentration data analysis was performed to determine constant values (β) in the above model. This model is specific to selected commodity.

Results and discussion

WVTR: Under the conditions of measurement, there was quite a large variation in permeability to water vapour than

to the other gases (Table 1). The WVTR of WPC with 15% glycerol in general was lower as compared to the control. This could be attributed to the plasticizing action of glycerol (Garcia et al. 1998), which resulted in improved flexibility of coatings (Park and Chinnan 1990). However, for CMC coatings, glycerol at 15% increased the WVTR; probably at this level glycerol with CMC may have increased the permeability of coatings. However, in hydrophobic coatings there was a significant reduction in the WVTR as compared to cellophane and hydrophilic coatings. The WVTR values showed a decreasing trend with an increase in PO and BW concentrations in coatings. The minimum WVTR was observed in 6% BW (98.8 g/m^2.day). As reported previously (Hagenmaier and Shaw 1991) permeability to water vapour can be even more sensitive to RH than permeability to O_2, especially for a coating that contains polar ingredients.

Viscosity: Viscosity of the coatings increased with increase in the concentration of coating materials (Table 1). In case of CMC coatings addition of glycerol reduced the viscosity. The hydrophobic coatings, 15% PO (40 × 10^3 mPa.s) and 6% BW (52 × 10^3 mPa.s), had the maximum viscosity that may be due to higher concentration of GMS (coating: GMS::1:2) instead of 1:1.5 being used for 5 and 10% PO and 2 and 4% BW coatings. The spreadability of the coating was improved with the increase in the viscosity. Dipping, as a method of coating application, was used only for coatings having viscosity below 220 mPa.s while brushing was used for viscosities above this level.

Permeability to O_2 and CO_2: Permeability to O_2 and CO_2 was generally lower for coatings with WPC and CMC than with PO and BW (Table 1). This observation fits the findings of Ashley (1985), who noted that polymers containing hydroxyl, ester, and other polar groups tend to have a lower O_2 permeability than polymers with hydrocarbon and other non polar groups. The O_2 content was reported to be 23% for shellac (Martin 1982) and 6% for carnauba wax (Bennet 1975). Because of equipment limitations, the permeabilities were measured at 14 ± 1°C and 55 ± 5% RH rather than at temperatures used for refrigerated and high RH being maintained for fruit storage. Shellac coatings at 0°C had O_2 permeability less than at 30°C (Hagenmaier and Shaw 1991). Some of the coatings have significantly different values of O_2 permeability at 50% and 85% RH (Hagenmaier and Shaw 1992). This RH dependence is in large part due to the polar components used to solubilize the polymer. For example, shellac solubilized with NaOH is much more permeable to O_2 than that solubilized with morpholine, especially above 85% RH (Hagenmaier and Shaw 1991). Thus, at the high values of RH best suited for fruit and vegetable storage, the permeabilities can be much higher than those shown in Table 1.

Mathematical model: To predict respiration rate (as ml CO_2/h/kg fruit weight) during storage at ambient and refrig-

Table 1 Viscosity, WVTR and O_2 and CO_2 permeability of edible coatings

Coatings	Viscosity, ×10^3 mPa.s	WVTR, g/m^2.day	Permeability cm^{3}cm cm^{-2}s^{-1}Pa^{-1}	
			O_2	CO_2
Control	-	528.9 ± 1.0	3.03 ± 0.40	13.10 ± 0.70
5% WPC	0.12 ± 1.1	448.8 ± 0.8	2.90 ± 0.27	15.97 ± 0.47
5% WPC + glycerol	0.12 ± 2.1	517.0 ± 1.0	2.73 ± 0.39	24.47 ± 0.67
10% WPC	0.16 ± 1.2	464.8 ± 0.8	2.59 ± 0.38	18.15 ± 0.65
10% WPC + glycerol	0.16 ± 1.2	447.4 ± 0.5	2.17 ± 0.37	12.66 ± 0.65
15% WPC	0.20 ± 2.7	523.8 ± 0.7	2.65 ± 0.36	9.39 ± 0.62
15% WPC + glycerol	0.24 ± 2.0	462.5 ± 0.5	1.50 ± 0.37	8.15 ± 0.64
0.5% CMC	0.36 ± 2.0	657.8 ± 0.7	2.43 ± 0.36	18.70 ± 0.63
0.5% CMC + glycerol	0.40 ± 1.0	648.6 ± 0.5	3.21 ± 0.38	11.36 ± 0.65
1.0% CMC	2.60 ± 2.0	658.4 ± 0.6	2.67 ± 0.36	10.97 ± 0.63
1.0% CMC + glycerol	2.20 ± 2.0	746.7 ± 0.8	3.79 ± 0.35	9.76 ± 0.60
1.5% CMC	30.40 ± 1.0	718.4 ± 0.5	3.52 ± 0.41	12.99 ± 0.70
1.5% CMC + glycerol	22.00 ± 2.0	758.0 ± 0.7	4.06 ± 0.34	8.13 ± 0.59
5% peanut oil	12.00 ± 2.5	261.9 ± 0.8	5.62 ± 0.35	21.01 ± 0.60
10% peanut oil	20.00 ± 2.0	208.9 ± 0.7	7.77 ± 0.37	24.02 ± 0.65
15% peanut oil	40.00 ± 1.0	186.9 ± 0.6	7.95 ± 0.37	23.51 ± 0.64
2% beeswax	28.00 ± 2.0	235.2 ± 0.5	3.25 ± 0.37	14.74 ± 0.64
4% beeswax	44.00 ± 1.7	202.4 ± 0.5	4.26 ± 0.37	18.41 ± 0.65
6% beeswax	52.00 ± 1.0	98.8 ± 0.8	4.38 ± 0.38	14.73 ± 0.67

(n=3) Mean±SD WPC= Whey protein concentrate, CMC= Carboxymethyl cellulose, PO= Peanut oil, BW= Bees wax

erated temperature conditions at any time during storage of guava a model shown by equation 1 was formulated. The values of β's corresponding to various treatments have been derived by plotting the concentrations of CO_2 along Y-axis and time along x-axis. Equation 1 can be used to adequately estimate the concentration of CO_2 at any time for coated and uncoated guava fruits by substituting the value of T in particular treatment used in the study. The values of β's obtained from the model, are given in Table 2. The values of R^2 have been calculated for each model and could be seen to be near '1' verifying the goodness of fit of each model corresponding to treatments described against set of β's in Table 2. From CO_2 concentration values against storage period (days) the best treatment for guava, which delayed the respi-

Table 2 The coefficients and R^2 values (desirable '1') of model for determining the respiration rate of guava as affected by various treatments during storage at ambient (16 ± 1°C, 60 ± 5% RH) and refrigerated conditions (10 ± 1°C, 65 ± 5% RH)

	Ambient condition							Refrigerated						
	Respiration rate, ml CO_2/h/kg fruit			Coefficients				Respiration rate, ml CO_2/h/kg fruit			Coefficients			
	4d	8d	12d	β_0	β_1	β_2	R^2	7d	14d	21d	β_0	β_1	β_2	R^2
Control I	128.4	88.2	54.3	37.2595	25.6123	-2.0661	0.8992	69.5	111.6	61.2	25.2720	11.5660	-0.4586	0.9311
Control II	90.3	119.9	58.5	27.0075	25.7018	-1.9023	0.9798	71.5	99.0	60.3	27.4185	10.2726	-0.4086	0.9719
Control III	35.6	62.4	89.4	28.9595	1.10612	0.3348	0.9949	32.7	71.4	89.6	27.1910	1.43940	0.0794	0.9685
WPC (5%)	95.4	124.2	60.7	27.2240	27.1810	-2.0131	0.9844	64.6	109.2	65.7	25.1165	10.5331	-0.3985	0.9206
CMC (1%)	88.5	109.1	58.6	28.3590	23.1048	-1.7041	0.9923	58.8	100.5	81.0	26.3095	7.95420	-0.2463	0.9487
PO (5%)	91.4	111.7	49.9	27.9805	25.0951	-1.9252	0.9900	64.5	112.1	76.0	25.1765	10.2145	-0.3602	0.9292
BW (4%)	89.1	109.2	54.1	28.2005	23.7176	-1.7839	0.9911	62.7	114.3	77.9	24.6810	10.1928	-0.3527	0.9190

Initial respiration rate (0 day)= 30.0, WPC, CMC, PO, BW : As in Table 1, d: Days of storage

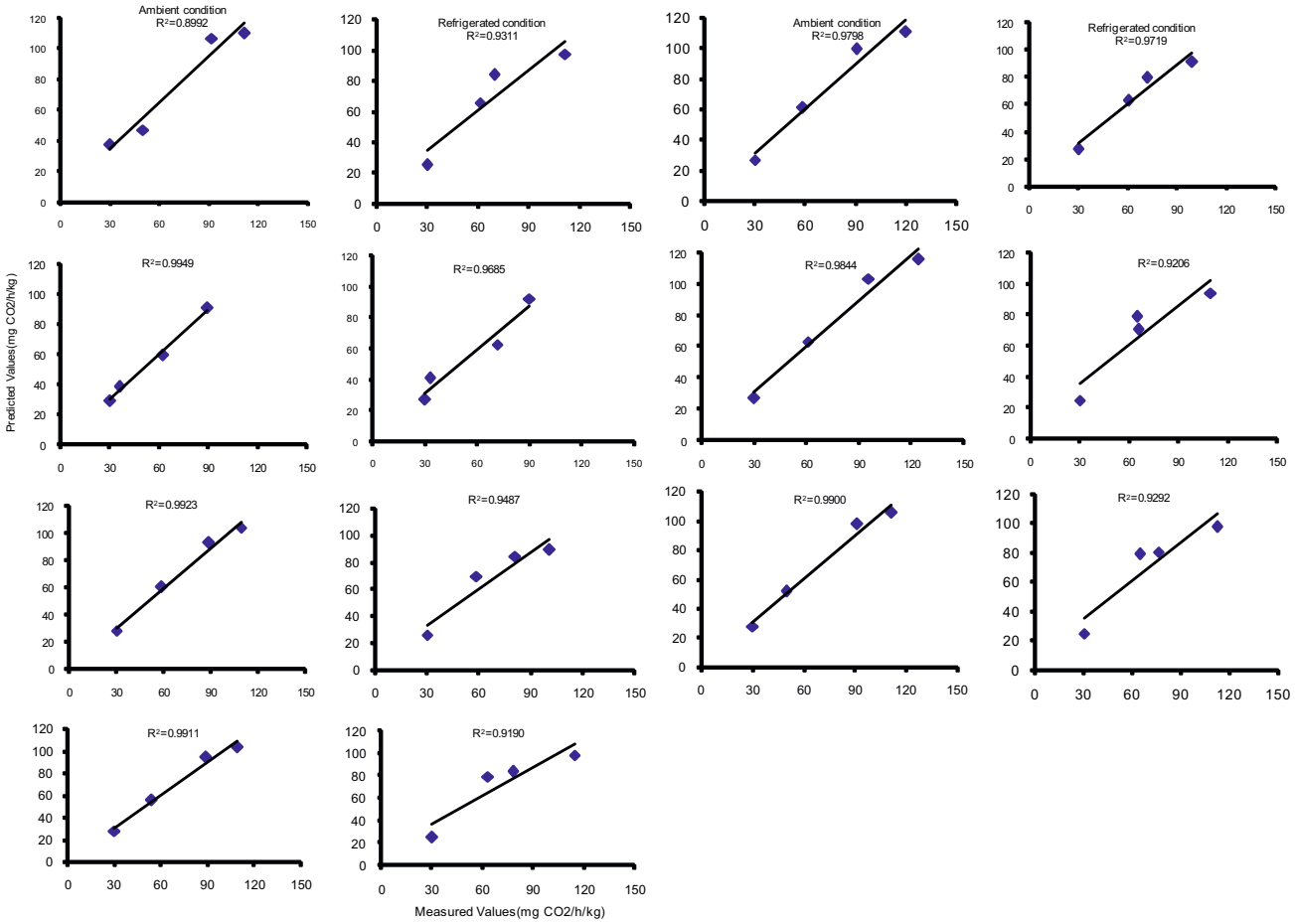

Fig. 1 Correlation graphs showing predicted vs. observed values of respiration rate under ambient and refrigerated storage conditions

ration rate, was control III under ambient conditions as well as refrigerated conditions (Table 2) while control I showed the minimum reduction in respiration rate on at both storage conditions. Amongst coatings, CMC coated guava showed maximum delay in the climacteric rise followed by BW and PO under ambient and refrigerated storage conditions. It is worth while to note that the model for each set of reading was worked out and a plot between predicted values from model and measured values was drawn, which invariably showed a straight line that could be easily fit between the points on scatter diagram (Fig. 1). This, in turn meant that a model reflected the original characteristics very well.

Conclusion

Coatings differed markedly in their permeabilities, thus some did not have the permeabilities for the purpose intended. Permeabilities for coatings should be low for O_2 and CO_2 as well as for water vapours. More data is needed on the performance of coatings of known permeability and thickness with respect to its effect on the produce physical, biochemical, physiological and microbiological parameters at higher RH values.

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