

Influence of Relative Humidity and Film Composition on Oxygen and Carbon Dioxide Permeabilities of Edible Films

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At low relative humidity (RH), an edible wheat gluten film presents very low oxygen and carbon dioxide permeabilities [1.24 and 7.4 amol/(Pa m s), respectively, at 25 °C]. For higher than 60% RH, O₂ and CO₂ permeabilities increase exponentially [to 1290 and 36 700 amol/(Pa m s), respectively, at 95% RH], presumably due to the plasticizing effect of water molecules. Pectin, chitosan, pullulan, and myofibrillar protein films are also very permeable to O₂ and CO₂ at high RH. The selectivity ratio (CO₂/O₂ permeability) of all films tested at high RH was very high (from 10 to 28) as compared to conventional synthetic films (4 to 6).

Keywords: *Edible film; gas permeability; relative humidity*

INTRODUCTION

Controlling the oxygen and carbon dioxide concentration around a food product is a well-known means to extend food shelf life. Oxygen is involved in many degradation reactions in food (fat and oil rancidity, microorganism growth, enzymatic browning, vitamin loss, etc.). Because of this, many packaging strategies seek to exclude oxygen, thereby protecting the product. For respiring commodities with living tissues, such as fresh fruits and vegetables, some permeability to oxygen and especially to carbon dioxide is essential, and moderate barrier packagings are more appropriate. The partial elimination of carbon dioxide is crucial for the maintenance of high-quality fruits and vegetables (Zagory, 1992). Some of them tolerate and even benefit from high carbon dioxide concentrations, up to 20% (e.g., strawberries, cherries, raspberries, broccoli, mushrooms). Others are injured by carbon dioxide concentrations above about 2% (i.e. pears, some apple varieties, lettuce, celery, tomatoes). If film with the appropriate permeability is chosen, a suitable equilibrium modified atmosphere will be established.

Edible films from polysaccharides, proteins, and/or lipidic compounds have already been considered for protecting various food products through gas transfer control (Guilbert, 1986; Kester and Fennema, 1986; Gontard and Guilbert, 1994) and for enhancing or replacing the natural coatings of fruits and vegetables, which could be lost during postharvest operations. Wax coating of fruits has been used in China since the 12th century to improve fruit quality and preservation. Waxing increases the carbon dioxide content and decreases the oxygen content of the internal atmosphere in oranges (Eaks and Ludi, 1960). Chitosan coatings retard ripening and prolong storage life of tomatoes, cucumber, and bell pepper fruits without affecting their ripening characteristics (El Ghaouth et al., 1990, 1991).

Lowings and Cutts (1982) reported that an edible composite coating (TAL Pro-long, which is sodium carboxymethylcellulose, sucrose esters of fatty acid, and monodiglycerides of fatty acids) produced a modified

atmosphere within fresh fruits after application. Changes in the internal concentration of oxygen, carbon dioxide, and ethylene and delayed ripening have been reported in apples, pears, limes, bananas, and mangoes (Cuq et al., 1995b). For example, this type of film (TAL Pro-long) was applied to bananas and caused a 6-fold reduction in oxygen permeability of the fruit skin, whereas carbon dioxide permeability was only reduced by about half (Banks, 1984).

The development of edible films and coatings with selective gas permeability is potentially of great interest for controlling respiratory exchange, achieving a modified atmosphere effect, and thus improving the preservation of fresh fruits and vegetables.

The main characteristics to consider in the selection of packaging materials for modified atmosphere packaging of fruits and vegetables are oxygen, carbon dioxide, and water vapor permeabilities. The gas permeability of a film depends on several factors such as the nature of the gas, the structure of the material, and the temperature. Although most plastic films are relatively unaffected by relative humidity, films made from biological materials do change their mechanical and barrier properties in high moisture conditions (Gontard et al., 1994), which is the case for fresh fruit and vegetable packagings.

Concerning edible films, numerous studies have been carried out to evaluate water vapor and oxygen permeability and the effect of the film type and structure and temperature. Although such information is quite useful for film selection, further information at more realistic high relative humidity levels on carbon dioxide and oxygen permeability is required for accurate film selection. With such information, it would be possible to determine the film required to attain a specific optimized equilibrium modified atmosphere for a given commodity. It could thus be used to match the specific properties of fruits and vegetables with tailor-made films and coatings.

The objectives of this study were to measure oxygen and carbon dioxide permeabilities of several edible films and examine the effect of relative humidity (RH) and film composition on gas permeabilities and selectivity. The effect of RH was specifically studied on a wheat gluten film developed in previous investigations (Gontard et al., 1992, 1993).

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MATERIALS AND METHODS

Preparation of Films. Wheat gluten based films were prepared according to previous investigations (Gontard et al., 1992, 1993). A film-forming solution was prepared using wheat gluten (Amylum Aquitaine, Bordeaux, France) at a concentration of 7.5 g/100 mL, ethanol (45 mL/100 mL), acetic acid (pH adjusted to 4.0), and distilled water. Glycerol was added at a concentration of 20 g/100 g of gluten as a plasticizer (Gontard et al., 1993).

All components were mixed vigorously by magnetic stirring and warmed at 45 °C. The film-forming solution was then immediately spread uniformly onto a level plexiglass surface using a thin-layer chromatography applicator and dried at 30 °C in a ventilated oven for 24 h. For each experiment, the quantity of the solution poured onto the surface was calculated so as to give a constant weight of dried matter (gluten plus glycerol plus lipid) of 11.8 mg/cm². This method allowed the formation of films with a controlled constant thickness (0.100 ± 0.005 mm), which was measured at several positions with a micrometer before the experiments.

For composite "mixed" films, diacetyl tartaric ester of monoglycerides ("Panodan" Grinsted, Marly-le-Roi, France) or white refined beeswax (La Ceresine, Marseille, France) was added at a concentration of 30 g/100 g of gluten in the film-forming solution, which was warmed to melt the lipids (Gontard et al., 1994).

Composite bilayer films were prepared by casting a thin layer of melted beeswax on a warmed base film composed of gluten and either diacetyl tartaric ester of monoglycerides or beeswax (Gontard et al., 1995). The quantity (5.3 mg/cm²) of deposited wax was finally controlled by weighing. The total film thickness was 0.140 mm.

The chitosan film preparation method was adapted from that described by Bai et al. (1988). An aqueous solution of chitosan (2 g/100 mL) and acetic acid (1.5 g/100 mL) was stirred and warmed at 70 °C and then cast onto a level plexiglass surface to obtain 3 mg/cm² dried matter. After drying at 30 °C, a 0.020 mm thick film was obtained.

For pullulan films, an aqueous solution of pullulan (Osaka Kagaku Gokin Co., Japan), at a concentration of 10 g/100 mL, was cast to obtain 3 mg/cm² dried matter and dried on a level plexiglass surface at 30 °C for 24 h (Sugimoto, 1986). The film thickness was 0.020 mm.

For pectin films, 5 g of pectin ("Unipectin", Sanofi, Paris, France) was solubilized in 100 mL of water at 60 °C. The solution was cast to obtain 5 mg/cm² dried matter, dried at 30 °C for 24 h, and then coated with an excess of 3% CaCl₂ solution. After 30 s, the excess CaCl₂ solution was eliminated and the film was allowed to dry again. The films obtained were 0.055 mm thick.

Myofibrillar protein films were prepared using the method of Cuq et al. (1995a). A film-forming solution was prepared from washed fish mince (protein concentration of 2 g/100 mL), acetic acid (pH adjusted to 3.0), glycerol (0.7 g/100 mL), and water. All components were mixed in a vacuum thermoregulated homogenizer. The solution was then stored for 6 h at 25 °C, cast on a plexiglass plate to obtain a dried matter weight of 3 mg/cm², and dried to obtain a film of 0.030 mm in thickness.

Wheat gluten, chitosan, pullulan, pectin, and myofibrillar proteins films were mechanically resistant, transparent, and easily handled when dried. Composite and bilayer films were more opaque and less resistant.

Film Permeability Measurements. Oxygen permeability (P_{O_2}) and carbon dioxide permeability (P_{CO_2}) were measured at 25 °C in a controlled-temperature room using a standard isostatic method. Samples were exposed to 100% oxygen or carbon dioxide on one surface and to a nitrogen carrier gas on the other, at atmospheric pressure and at the desired RH. The transmission rates of oxygen and carbon dioxide through the film were calculated from the steady-state partial pressure of oxygen in, and the flow rate of, the carrier gas stream.

Our specially designed permeability cell contained two circular (8 cm diameter × 0.5 cm deep) 25 mL chambers

separated by the film sample, of which 50 cm² was exposed to both chambers and sealed with an O-ring.

The RH of gases was controlled by passing each gas stream through a gas drying tower (0% relative humidity) or four successive gas washing bottles containing a saturated salt solution corresponding to the required relative humidity conditions, ranging from 6 to 100% (Spiess and Wolf, 1987; Multon, 1984; Stamp et al., 1984).

Prior to the permeability measurements, to accelerate the time to reach steady state, films were conditioned for 5 days at the desired RH in closed containers with suitable saturated salt solutions.

Wheat gluten films were tested at various relative humidities. For composite, bilayer, pectin, chitosan, pullulan, and myofibrillar protein films, the first objective was to measure permeability at close to 100% RH. This was not possible because some of them were highly sensitive to moisture and could not be measured at very high RH. Thus, in the high RH range, the RH given for each film was the highest possible.

Water activities in films were precisely determined by placing the film, immediately after measurements, in the chamber of a Novasina apparatus (ref TH-2/RTD-33/BS, Defensor France, Neuilly-sur-Marne). The results obtained correspond to the exact RH indicated for each measurement.

The partial pressure of oxygen or carbon dioxide in the carrier gas stream was determined using an oxygen (electrochemical detection) and carbon dioxide (analytical infrared gas analyzer, standardized for each RH) analyzer (PAK 12X, Abiss, France).

Measurements were obtained when the steady state was reached (constant oxygen concentration in the carrier gas stream).

The flow rate was determined using a flow meter (Chromoptic, Auxerre, France), and pressures in the chambers were equalized and maintained at atmospheric pressure.

Oxygen and carbon dioxide permeabilities were calculated as follows:

$$P = J(\Delta x)/A(\Delta p)$$

with P in amol/(m s Pa) (amol is 10⁻¹⁸ mol). P is the oxygen or carbon dioxide permeability coefficient of the film, J is the transmission rate of oxygen or carbon dioxide through the film (in amol/s), Δx is the film thickness (in m), Δp is the differential partial pressure across the film (in Pa), and A is the surface area of the film (in m²).

Three replicates of each film type were tested.

For each type of film and relative humidity, the P_{O_2} values obtained were confirmed using an Ox-Tran 2/20 ML modular system according to method D3985 of the American Society for Testing and Materials (ASTM, 1988).

RESULTS AND DISCUSSION

Effect of Relative Humidity and Film Composition on Oxygen and Carbon Dioxide Permeability of Wheat Gluten and Other Edible Films. P_{O_2} and P_{CO_2} determine the composition of the atmosphere around the fruit or vegetable; respiration of the fruit or vegetable depletes the oxygen and enriches the carbon dioxide inside the film.

The effects of RH were first studied in detail on a wheat gluten film. The P_{O_2} and P_{CO_2} of various other edible films were then considered.

At 0% RH, a wheat gluten film presented very low P_{O_2} and P_{CO_2} [1.24 and 7.4 amol/(m s Pa), respectively] as compared to other edible, biodegradable or synthetic films (Table 1). For example, P_{O_2} of wheat gluten film was 800-fold lower than that of low-density polyethylene and 10-fold lower than that of Polyamide 6, a well-known high oxygen barrier polymer at 0% RH (Billing, 1989). Wheat gluten film was 2.6- and 570-fold less permeable to carbon dioxide than Polyamide 6 and low-density polyethylene, respectively. Interesting applica-

Table 1. Oxygen and Carbon Dioxide Permeabilities of Various Films at 0% RH

film ^a	O ₂ permeability (amol/m s Pa)	CO ₂ permeability (amol/m s Pa)	temp (°C)
EVOH (1)	0.16		23
chitosan (2)	0.57	1.83	25
wheat gluten (3)	1.24	7.4	25
cellophane (1)	1.34		23
Polyamide 6 (1)	11.9	19.5	23
PET (1)	11.9	37.6	23
rigid PVC (1)	16.0		23
MC/beeswax (4)	499		25
MC/palmitic acid (5)	190		24
LDPE (1)	1003	4220	23
MC/PEG (6)		29900	21

^a According to (1) Poyet (1993), (2) Pavlath et al. (1993), (3) present study, (4) Greener and Fennema (1989), (5) Rico-Peña and Torres (1990), (6) Park and Chinnan (1990). EVOH is ethylenevinyl alcohol, PET is poly(ethylene terephthalate), PVC is poly(vinyl chloride), LDPE is low-density polyethylene, MC is methylcellulose, and PEG is poly(ethylene glycol).

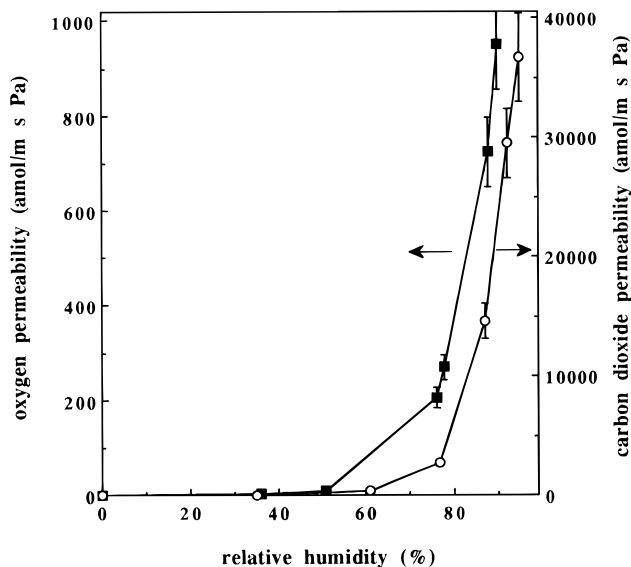


Figure 1. Effect of relative humidity on oxygen (■) and carbon dioxide (○) permeability (1 amol = 10⁻¹⁸ mol) at 25 °C of a wheat gluten film (variability between replicates is indicated only when variability exceeded size of the symbol).

tions of wheat gluten film as an oxygen and carbon dioxide barrier in dry conditions could thus be considered (e.g., for low water activity products or sandwiched between two layers of a highly water resistant material).

However, P_{O_2} and P_{CO_2} of the wheat gluten film increased substantially with increases in RH (Figure 1).

At 25 °C and RH lower than 50%, P_{O_2} remained relatively constant and lower than 8 amol/(m s Pa).

At RH higher than 50%, the curve became exponential with a sharp increase in P_{O_2} for the highest RH levels. Such behavior is typical of hydrophilic polymers including cellophane, ethylenepolyvinyl alcohol, or methylcellulose-palmitic acid films (Figure 2). This exponential effect of RH on P_{O_2} was previously observed for other edible film types (McHugh and Krochta, 1994; Rico-Peña and Torres, 1990).

When there are no pores, faults, or membrane punctures, permeability (P) is equal to the product of the diffusion coefficient (D), representing the mobility of permeant molecules in the polymer, and the solubility coefficient (S), representing the permeant concentration in the film balanced with the external pressure: $P = DS$. The increase in permeability could thus be related

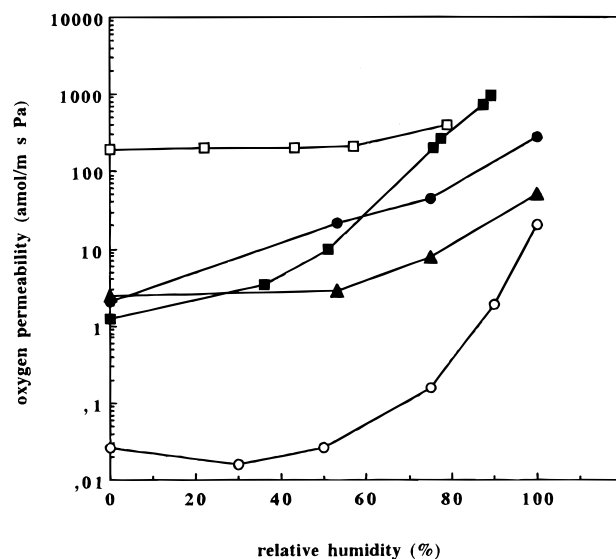


Figure 2. Effect of relative humidity on oxygen permeability of various edible and synthetic films at 25 °C according to the following studies: Poyet (1993) for EVOH (○) and Polyamide 6 films (▲); Rico-Peña and Torres (1990) for MC/palmitic acid film (□); Rigg (1979) for cellophane film (●); and the present study for wheat gluten film (■) (MC is methylcellulose, and EVOH is ethylenevinyl alcohol).

to the increase in the diffusion coefficient, due to structural modifications of protein matrix, and also to the increase in oxygen solubility in the film by an increased water content in the film (oxygen solubility in water = 1.25 mmol/L at 25 °C and 1 atm).

The carbon dioxide measurements indicated that the effect of RH on permeability was similar to that of P_{O_2} but the sharp increase in permeability was more important. Generally, carbon dioxide permeates through plastic polymer more rapidly than other atmospheric gases such as oxygen. The carbon dioxide solubility in water ($S = 34.5$ mmol/L at 25 °C and 1 atm) was higher than oxygen solubility and could explain this difference.

The sharp increase in P_{O_2} and P_{CO_2} was similar to the results obtained for water vapor permeability with the same wheat gluten film (Gontard et al., 1992) and could be correlated with a sharp modification in the protein network structure and polymeric chain mobility, corresponding to a change from a glassy to a viscoelastic state. The observed increase in the P_{O_2} , in the 40–100% RH range, could be related to the plasticization effect of the amorphous protein backbone by water molecules. Water molecules in the polymer above a water activity level of 0.4 interact with hydrophilic groups and thus may disrupt hydrogen bonding, creating additional sites for the dissolution of oxygen and increased mobility of oxygen molecules within the polymer.

Using sorption isotherms for wheat gluten films (Gontard et al., 1992), RH was converted into water content (Figure 3). The sharp increase in P_{O_2} was observed at about 10% (dry matter) water content. For higher than 20% water content, P_{O_2} increased quite linearly with the water content (5 P_{O_2} units for 1% water). This means that the effect of water on P_{O_2} could be governed by two different laws according to the water content of the film. These laws depend on the structure of the film, which could be rigid and vitreous at low water content and rubbery and viscous at high water content (Gontard, 1993). For P_{CO_2} , conversion of RH into water content led to the same conclusions as for

Table 2. Oxygen and Carbon Dioxide Permeabilities of Various Films at High RH

film ^a	O ₂ permeability [amol/(m s Pa)]	CO ₂ permeability [amol/(m s Pa)]	RH (%)	temp (°C)
pectin	297		87	25
	1340	21300	96	25
chitosan	472	8010	93	25
pullulan	17	72	30	25
wheat gluten	982	24500	91	25
wheat gluten	1290	36700	95	25
myofibrillar proteins	290		86	25
	873	11142	93	
wheat gluten/DATEM	790	8811	95	25
wheat gluten/beeswax	687	6614	91	25
wheat gluten/DATEM and beeswax (bilayer film)	11	76	56	
wheat gluten/beeswax and beeswax (bilayer film)	10	61	56	
EVOH (1)	6		95	23
Polyamide 6 (2)	10		100	23
PET (2)	12		100	23
cellophane (3)	130		95	23
starch (4)	1085	26410	100	24

^a According to (1) Salame (1986), (2) Ashley (1985), (3) Taylor (1986), and (4) Allen et al. (1963). DATEM is diacetyl tartaric ester of monoglyceride, EVOH is ethylenevinyl alcohol, and PET is poly(ethylene terephthalate).

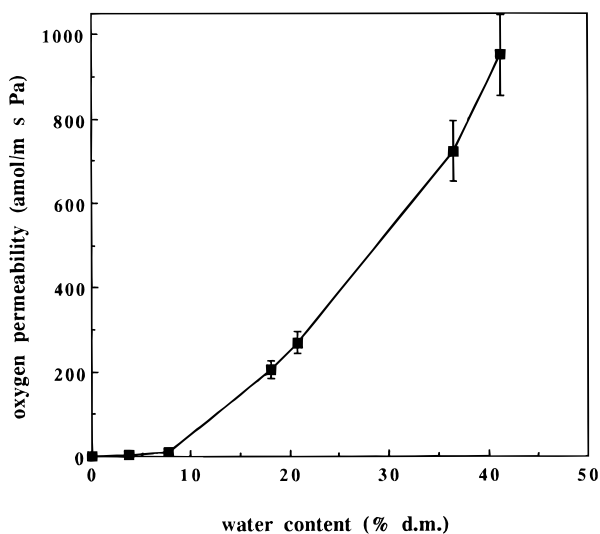


Figure 3. Effect of film water content on oxygen permeability of a wheat gluten film at 25 °C.

P_{O_2} : P_{CO_2} is linearly dependent on the water content with two distinct slopes for high and low water content ranges.

In practical terms, it was shown that P_{O_2} and P_{CO_2} of wheat gluten films varied greatly with their water content. The water activity of the food and the RH of the surrounding atmosphere may reduce or increase the concentration of oxygen and carbon dioxide available for respiration or oxidation processes when such hydrophilic compounds are used as a food packaging or coating material.

Various other films were tested for P_{O_2} and P_{CO_2} measurements in high RH conditions: pectin, chitosan, pullulan, and proteins such as caseinate, fish proteins, and wheat gluten combined or not with lipid compounds. The results are presented in Table 2.

Pullulan film was highly sensitive to moisture and became extremely viscous at high RH, thus rendering the measurements impossible in these conditions. However, this film showed good oxygen barrier properties in quite low RH conditions. These properties could explain the high effectiveness of pullulan film in preventing oxidation and rancidity and increasing the acidity in high fat and low water activity foods such as peanuts, cashew nuts, almonds, dried fish, seasonings, or dried pork (Sugimoto, 1986).

The effects of moisture on pectin, chitosan, and fish protein films were just as important as on wheat gluten film. The hydrophilicity of these macromolecules explains the high P_{O_2} and P_{CO_2} observed at high RH.

The addition of lipid components to film-forming solution resulted in a decrease in the P_{O_2} of the composite mixed film. At 91% RH, oxygen permeability was reduced by about 30% for a 20% beeswax content. At 94.5% RH, including 30% of diacetyl tartaric ester of monoglyceride in the wheat gluten formulation reduced film permeability by about 40%. Similar effects were shown on P_{CO_2} . This effect could be related to the hydrophobic characteristics of these components which reduce water content of the film for the same RH and thus oxygen or carbon dioxide solubility. It was also previously demonstrated that diacetyl tartaric ester of monoglyceride strengthens the protein structural matrix, thus improving the barrier properties of the film (Gontard et al., 1992).

At 56% RH, very low P_{O_2} and P_{CO_2} were observed for wheat gluten and beeswax bilayer films. Bilayer films were tested for P_{O_2} and P_{CO_2} at very high RH, but the wheat gluten based film swelled with water adsorption and separated from the beeswax layer.

Compared to synthetic films such as poly(ethylene terephthalate) or ethylenepolyvinyl alcohol, all edible films tested lost their oxygen barrier properties at high RH.

Effect of Relative Humidity and Film Composition on the CO₂/O₂ Permeability Ratio. The selectivity, represented by the carbon dioxide/oxygen permeability ratio, is one of the most descriptive parameters of a film (Zagory, 1992) and determines the relative proportions of carbon dioxide and oxygen in the package. Films with high ratio values will allow carbon dioxide to escape from the package relatively easily, resulting in low carbon dioxide concentration atmosphere. Films with lower ratio values will allow greater CO₂ buildup in the package. The selectivity ratio determines the possible combination of oxygen and carbon dioxide concentrations inside the film. Since fruits and vegetables vary in their tolerance to carbon dioxide and in their ability to benefit from high percentages of this gas, the selectivity ratio value of a film is very important for predicting the relative amounts of oxygen and carbon dioxide that will accumulate.

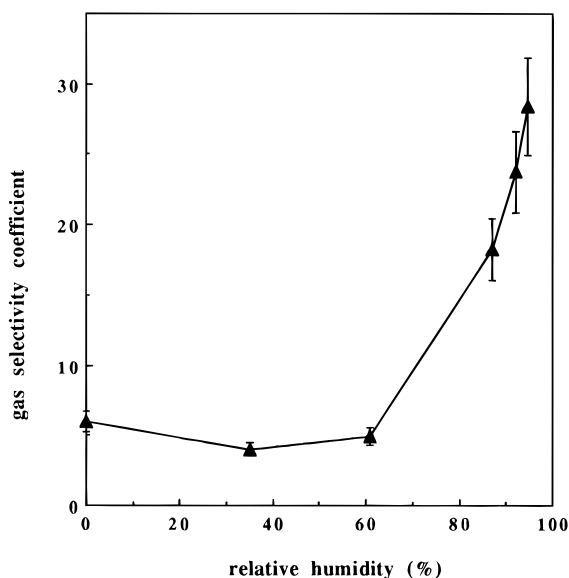


Figure 4. Effect of relative humidity on gas selectivity coefficient (carbon dioxide permeability/oxygen permeability ratio) of a wheat gluten film at 25 °C.

Table 3. Gas Selectivity Coefficient (Carbon Dioxide Permeability/Oxygen Permeability Ratio) of Various Edible Films

films	gas selectivity coeff	RH (%)
pectin	16.0	96
chitosan	17.0	93
pullulane	4.2	30
wheat gluten	25.0	91
wheat gluten	28.4	95
myofibrillar proteins	12.7	92
wheat gluten/DATEM	11.1	95
wheat gluten/beeswax	9.6	91
wheat gluten/DATEM and beeswax (bilayer film)	6.8	56
wheat gluten/beeswax and beeswax (bilayer film)	6.2	56

The P_{CO_2}/P_{O_2} ratio of a wheat gluten film (Figure 4) was dependent on the RH. Up to 60%, the selectivity ratio value was relatively low (4–6) and equivalent to the selectivity of synthetic polymers such as poly(vinylidene chloride) (5.8), polypropylene (4.0), or Polyamide 6 (4.2) (Lefaux, 1972). For higher RH, selectivity increased to 28.4 at 94.5% RH. This value corresponds to the ratio of carbon dioxide and oxygen solubility in water. At very high RH (close to 100%), solubility of oxygen or carbon dioxide molecules in the free water of the film becomes the main parameter for the transport of these molecules through the film.

At high RH, the selectivity of highly hydrophilic films, such as chitosan, pectin, or wheat gluten films, was important (Table 3). The selectivity values decreased when lipidic components were added to wheat gluten, but the ratio obtained remained more than twice as high as in common synthetic films.

Microperforated synthetic films are marketed for their ability to achieve high permeability by allowing gases to move across the film via mass flow, which is much faster than the usual permeation process. However, mass flow does not provide the differential permeabilities to oxygen and carbon dioxide that nonperforated films offer. With resulting CO_2/O_2 permeability ratio values close to 1, these films will not provide a low carbon dioxide atmosphere. The edible films tested

could thus be useful when both high permeability and high selectivity are required.

Conclusion. Fresh vegetable storage requires specific permeability properties which could be approached by microperforations in synthetic films. The high permeability and natural selectivity of the films tested in this study could be advantageously used with fresh fruits and vegetables to provide desirable storage conditions. Since both product respiration and film permeability are temperature sensitive, it is essential to determine P_{O_2} and P_{CO_2} at the low temperature at which the film will be stored. Studies are under way to study the combined action of temperature, RH, and film composition on gas permeability of edible films.

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