

# Oxygen permeability and airtightness measuring method for breathing bags

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Vigneault, C., Orsat, V., Panneton, B. and Raghavan, G.S.V. 1992. Oxygen permeability and airtightness measuring method for breathing bags. *Can. Agric. Eng.* 34:183-187. A non destructive measuring method was developed to measure the airtightness and the O<sub>2</sub> permeability of a breathing bag system used in CA storage. The measuring method was used to test three different types of bags. The three types of bags were significantly different but the specimens of the same type of bags were not significantly different. Accurate permeability was not measurable when airtightness was not sufficient.

Une méthode non-destructive pour évaluer l'étanchéité et la perméabilité à l'oxygène de sacs tampons utilisés en entreposage AC a été développée. Cette méthode a été utilisée pour évaluer trois types différents de sac. Les résultats ont démontré des différences significatives entre les trois types de sac, mais les sacs d'un même type n'étaient pas significativement différents. La perméabilité des sacs n'a pu être mesurée lorsque leur étanchéité était mauvaise.

## INTRODUCTION

The controlled atmosphere (CA) miniature chamber developed by Vigneault et al. (1991) requires a breathing bag to compensate for any variations in total gas volume caused by fluctuations in barometric pressure or temperature (Fig. 1). Such a bag is inflated at the beginning of a storage period and is installed outside the mini-chamber to allow its free expansion when needed. Because the concentration of O<sub>2</sub> in the bags is lower than in the surrounding air, care must be taken in the choice of the bag material. The O<sub>2</sub> permeability of the bag liner must be low to avoid O<sub>2</sub> entering the mini-chamber through the bag surface. Breathing bag sealing and connections to the mini-chamber have to be airtight to avoid air infiltration and O<sub>2</sub> build up in the bag. The objectives of this work were to develop a simple method to measure the airtightness of the breathing bag system, the O<sub>2</sub> permeability of the different types of bags, and to find a better breather bag system than the double polyethylene bag used by Vigneault et al. (1991).

## MATERIALS AND METHOD

Three types of commercial bags were chosen: Type A, a double polyethylene and aluminum bag (used for wine storage); Type B, a double polyethylene bag (TwinPak BXL HY-BAR commonly used for milk storage); and Type C, a multi-layer barrier bag (Cryovac B-620). The bags called Type A and Type B were already sealed on their four edges, and were supplied with one molded outlet, 27 mm in diameter. The Type C bags were initially open at one edge. They

were sealed by folding the extremity once and snapping the bag at 10 mm from the fold with a "Zip-Lock" mechanical joint commercially sold as a winterizing window system. A commercial plastic bulkhead union (tube to tube type) was used to connect each bag to a 4.4 mm outside diameter flexible tubing. This flexible tubing was either used for connecting the bag to the mini-chamber when the bag was used as a breather bag, or for connecting the bag to the filling system, or for gas sampling through a septum connection during the test period of the three different bags.

The method used to evaluate the airtightness of the breather bag is similar to the method described by Bartsch and Blanpied (1984) for airtightness of a CA storage room. The bags were pressurized at a pressure of about 250 Pa, then the pressure differential between both sides of the surface of the bag and the barometric pressure were monitored at about 8 minute intervals. Since a 1°C temperature difference induces an average pressure change of 350 Pa, infiltration tests were conducted under constant temperature and barometric pressure. The tests were repeated when the barometric pressure was not stable, and the temperature was kept constant by putting the bag inside a closed box to prevent temperature fluctuations due to the ventilation system. Thermocouples placed inside and outside the box showed that the temperature did not fluctuate by more than 0.1°C through the testing period.

The time  $t_{1/2}$  is the time the pressure differential takes to reach half of its starting value. It is a characteristic of the bag and it is related to the airtightness of the breather bag system. The time  $t$  of the bag could be deduced from the curves of differential pressure drop as a function of time. For example, a breather bag is referred to as a "30 minute bag" when a period of time of 30 minutes is required for the differential pressure to drop to half its initial value. As a reference, a "10 minute CA storage room", when empty, roughly experiences one air change on a 30 day period (ASHRAE 1982). A "20 minute" system is considered as minimum for a CA storage system (Bartsch and Blanpied 1984).

The O<sub>2</sub> permeability of the first two types of bags was not available. A 13.1 to 20.5 mL·h<sup>-1</sup>·m<sup>-2</sup>·MPa<sup>-1</sup> membrane permeability was specified for the Type C. Since fittings were added to all bags, it was decided to measure the airtightness and the permeability of the complete system.

The O<sub>2</sub> permeability was measured as follows. Each bag was filled at atmospheric pressure with 20 L of pure N<sub>2</sub> gas with the help of a pressure vacuum system that was built to

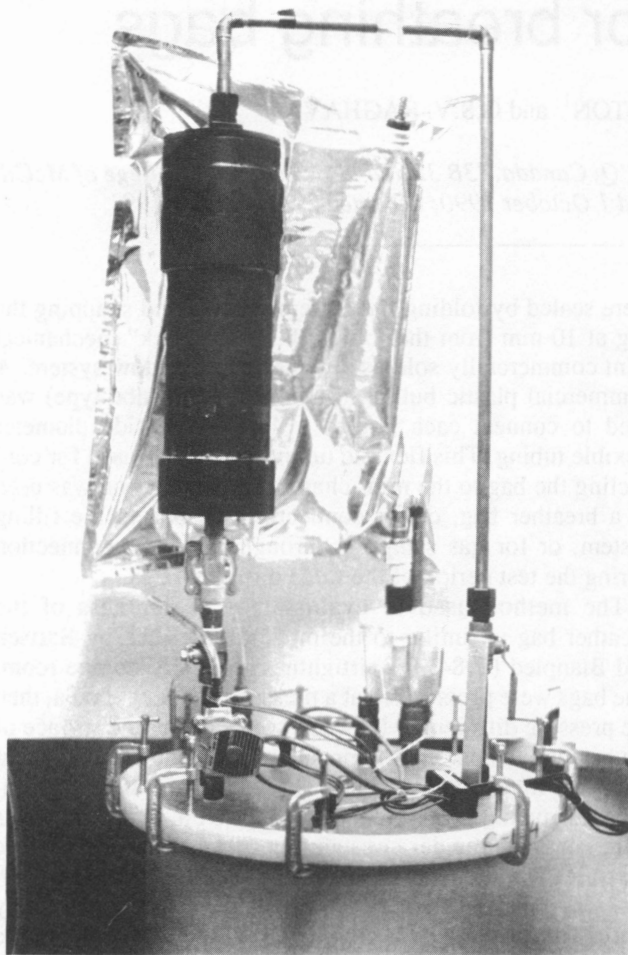


Fig. 1. Mini-Chamber CA storage system developed by Vigneault et al. (1991).

displace an exact volume of gas. Figure 2 shows a schematic of the system. A volumetric flask was connected to an airtight container with a flexible tubing. The volumetric flask was placed on a scale having a 0.1 g resolution, and the connecting tube was attached to a stand to avoid its influence on the mass of the flask given by the scale. The airtight container may be moved to two different levels. At level 1, water in the airtight container exits to the flask and creates a vacuum that sucks pure nitrogen from the gas source. The volume of gas entering the airtight container is the same as the volume of water displaced. At level 2, the water exits from the flask pushing the gas out of the airtight container into the test bag. The movements of water and gas in the system are controlled by a set of clip valves. The operation is repeated until the desired volume of gas in the bag is reached.

Two bags were emptied immediately after being filled. The difference between the volumes measured during the filling process and the emptying process did not exceed 5 mL over the 20 L volume, a difference of 0.025%.

After all the bags were completely filled, the flexible tube

used to fill the bag was replaced by a septum. The septum was squeezed by a threaded screwed cap against the fitting of the connector to obtain an airtight module. Then the bags were suspended in a well ventilated room in order to put their total surface in good contact with the atmospheric gases.

The gas concentration in each bag was measured daily over a two week period with a gas chromatograph (GC). The gas homogeneity in the bags was obtained by gently moving the sides of the bags a few times to get an air movement in the bag. Inconsistent results were obtained when the air inside the bag was not mixed before sampling. A 5 mL syringe was used for sampling the gas inside the bag through the septum. The syringe was purged a few times to eliminate any residual gases it may have contained and to mix the gas near the sampling inlet. The 5 mL samples were used to purge the 0.5 mL sampling loop of a GC and this gas was analyzed using a standard GC gas analysis method. The method used permitted the separation and the quantification of CO<sub>2</sub>, O<sub>2</sub>, and N<sub>2</sub> gases of the sample.

After two weeks of sampling, the final volumes of gas in the bags were measured. The system used to fill the bags (Fig. 2), was used to empty the bags with manipulations executed in the reverse order. The barometric pressure was noted at the filling and the emptying of the bags to determine the exact volume of gas in the bag.

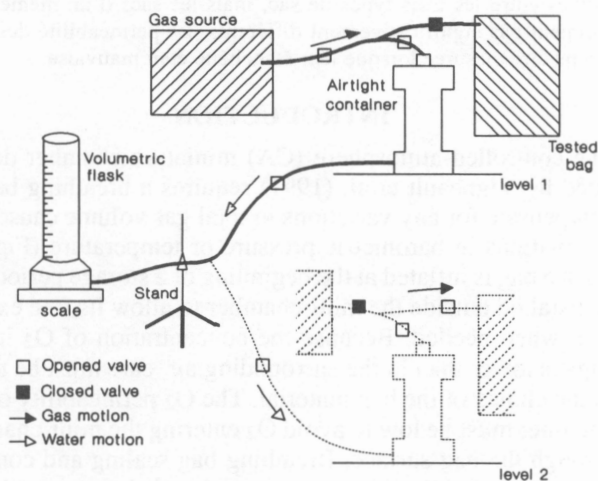


Fig. 2. Schematic view of volume measurement setup (filling).

## RESULTS AND DISCUSSION

### Airtightness testing

The analysis of the data on airtightness requires the development of a suitable model for the differential pressure drop as a function of time. This method is necessary when experimental results have to be extrapolated to obtain  $t_{1/2}$ .

The model is based on two realistic assumptions. The first one states that the differential pressure drop results from the loss of some gas exiting the bag through small orifices. The net loss resulting from the diffusion across the membrane is neglected on the basis that the time scale of this phenomenon is much larger than the time scale of the gas loss through

orifices. The second assumption is that the flow of gas through the orifices is incompressible. This is justified by the low initial pressure differential of 250 Pa. At this pressure, the incompressible approximation gives results which are within 0.09% of the one obtained for compressible flows (Van Wylen and Sonntag 1959).

The analysis starts with the incompressible Bernoulli's equation:

$$V = \sqrt{\frac{2(P_1 - P_2)}{\rho_2}} \quad (1)$$

where:

$v$  = velocity of gas exiting bag ( $\text{m}\cdot\text{s}^{-1}$ ),  
 $P_1$  = pressure inside bag (MPa),  
 $P_2$  = atmospheric pressure (MPa), and  
 $\rho_2$  = density of gas ( $\text{kg}\cdot\text{m}^{-3}$ ) (here taken as the density at atmospheric pressure by the second assumption).

From Eq. 1, the mass flow rate of gas out of the bag is given by:

$$\dot{m} = \left[ \sum_{i=1}^N C_{vi} A_i \right] \sqrt{2\rho_2(P_1 - P_2)} \quad (2)$$

where:

$\dot{m}$  = mass flow rate ( $\text{kg}\cdot\text{s}^{-1}$ ),  
 $C_{vi}$  = velocity coefficient of  $i^{\text{th}}$  orifice,  
 $A_i$  = cross sectional area of  $i^{\text{th}}$  orifice ( $\text{m}^2$ ), and  
 $N$  = number of orifices in the bag.

The summation in Eq. 2 is an unknown parameter. It includes the effect of the number of orifices and their shape. From the perfect gas law it is easy to obtain:

$$\frac{dP_1}{dt} = - \frac{\dot{m}R_aT_1}{V_1} \quad (3)$$

where:

$t$  = time (s),  
 $R_a$  = gas constant ( $\text{Pa}\cdot\text{m}^3\cdot\text{kg}^{-1}\cdot\text{C}^{-1}$ ),  
 $T_1$  = temperature inside bag ( $^{\circ}\text{C}$ ), (here  $T_1 = T_2$ ), and  
 $V_1$  = total volume of bag ( $\text{m}^3$ ).

Substitution of Eq. 2 into Eq. 3 yields:

$$\frac{dP_1}{dt} = - \alpha \sqrt{P_1 - P_2} \quad (4)$$

where:

$$\alpha = \left[ \sum_{i=1}^N C_{vi} A_i \right] \frac{R_aT_1}{V_1} \sqrt{2\rho_2} \quad (4a)$$

The parameter  $\alpha$  contains the bag characteristics  $C_{vi}$ ,  $A_i$  and  $V_1$  together with  $R_a$ ,  $T_1$  and  $\rho_2$  which depend on the test conditions. To provide a basis for comparison of bags tested under different conditions the standardized parameter  $\alpha_s$  is introduced:

$$\alpha_s = \alpha \frac{R_sT_s}{R_aT_1} + \sqrt{\frac{\rho_s}{\rho_2}} \quad (5)$$

where subscript "s" denotes standard atmospheric conditions.

Eq. 4 can be solved to obtain:

$$\sqrt{\frac{\Delta P}{\Delta P_o}} = 1 - \alpha t \quad (6)$$

where:

$\Delta P = P_1 - P_2$  (MPa), and  
 $\Delta P_o = P_1 - P_2$  at  $t = 0$  (MPa).

Equation 6 is the basic model. Solving for  $t$  at  $\Delta P/\Delta P_o = 1/2$  yields:

$$t_{1/2} = \frac{0.293}{\alpha} \quad (6a)$$

or

$$t_{1/2s} = \frac{0.293}{\alpha_s} \quad (6b)$$

where:

$t_{1/2s}$  = standardized  $t_{1/2}$ .

It is of interest to relate our results (Eqs. 5 and 6) to the method recommended by Bartsch and Blanpied (1984). The left-hand side of Eq. 6 is first expanded into a Taylor's series:

$$\sqrt{\frac{\Delta P}{\Delta P_o}} = 1 + \frac{1}{2} \ln\left(\frac{\Delta P}{\Delta P_o}\right) + \frac{1}{4 \times 2!} \ln^2\left(\frac{\Delta P}{\Delta P_o}\right) + \dots \quad (7)$$

For  $\Delta P/\Delta P_o$  close to 1, the first two terms of Eq. 7 are retained for a first order approximation. Equation 6 can then be rewritten as:

$$\ln\left(\frac{\Delta P}{\Delta P_o}\right) = -2\alpha t \quad (8)$$

Bartsch and Blanpied (1984) recommended to fit a straight line through a plot of  $\ln(\Delta P/\Delta P_o)$  versus time and to use this line to estimate  $t_{1/2}$ . Equation 8 provides a theoretical basis for this technique. When compared to Eq. 6 at  $\Delta P/\Delta P_o=0.5$ , Eq. 8 yields a  $t_{1/2}$  that is overestimated by 18%, a significant error.

The results from the first airtightness test on Type A bags are presented in Fig. 3, where the plot shows  $\sqrt{\Delta P/\Delta P_o}$  versus time. Also shown on the graph are theoretical curves for 20 and 30 minute bags. It is obvious that Type A bags leak

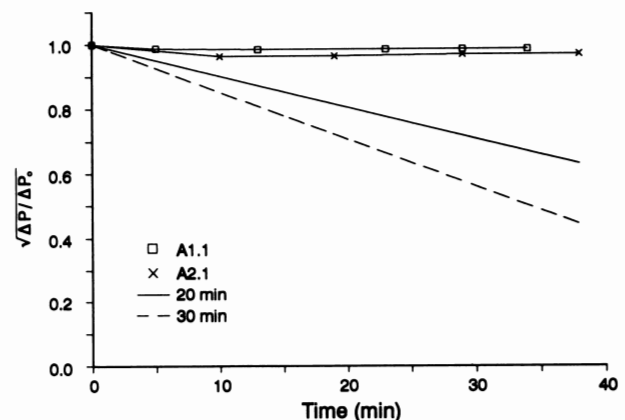


Fig. 3. Differential pressure drop as a function of time during the airtightness tests of the Type A bags, compared to "30 minute" and "20 minute" bags.

much slower than 30 minute bags. The average standardized slope  $\alpha_s$  of the plot of  $\sqrt{\Delta P/\Delta P_o}$  versus time and  $t_{1/2s}$  are presented in Table I.

Type A and Type B breathing bags were considered sufficiently airtight for our application, since each breathing bag had a  $t_{1/2s}$  superior to "30 minute", as recommended by Bartsch and Blanpied (1984) for low O<sub>2</sub> concentration storage room. The Type C bags were not considered adequate for CA storage since the  $t_{1/2s}$  of bag C1 was less than "30 minute". Further data from replicate tests revealed that the airtightness of the Type C bags decreased in time.

Some tests were also performed with Type A bags using a cheaper press-fit tubing system to connect the breathing bag to the storage room. The  $t_{1/2s}$  obtained with these bags varied between 3.4 and 16.2 min. This low value was mainly due to the poor tightness of this type of coupling. Permeability tests made with these bags were inaccurate. The bags lost respectively 48% and 15% of their initial volume during the 13 days test duration. This coupling system was considered inadequate for our application.

### Permeability tests

Permeability tests were conducted on two specimens of each type of bag and the experiment was replicated once. The measurements of the air volume contained in the bag at the end of the test period indicated no air leak for all the bags. In fact, the volumes measured were superior to 20 L. This increase in the volume corresponded to the O<sub>2</sub> entering through the bag surface due to O<sub>2</sub> permeability. Figures 4 to 6 present the results of the permeability tests. The O<sub>2</sub> concentration is

expressed as a function of time. Results for Type A and Type B bags show good repeatability. On the other hand in Fig. 6, the O<sub>2</sub> permeability of the Type C bags increases with time and/or manipulations.

The permeabilities of the bags were calculated assuming the gas concentration changed linearly with time which is a good approximation for the results presented. In this way, the volume of gas in the bag could be estimated for each sample and the O<sub>2</sub> permeabilities of the bags were calculated using Eq. 9 .

$$\mu_i = \frac{(O_{2i} - O_{2i-1}) V}{\Delta t S \Delta P_{O_2}} \quad (9)$$

where:

- $\mu_i$  = mean O<sub>2</sub> permeability of liner (mL O<sub>2</sub>·h<sup>-1</sup>·m<sup>-2</sup>·MPa<sup>-1</sup>) for i<sup>th</sup> sample,
- $O_{2i}$  = O<sub>2</sub> volume fraction for i<sup>th</sup> sample,
- $O_{2i-1}$  = O<sub>2</sub> volume fraction for i<sup>th</sup>-1 sample,
- $V$  = volume of gas contained in bag (mL),
- $\Delta t$  = time interval between i<sup>th</sup> and i<sup>th</sup>-1 samples (h),
- $S$  = surface area of membrane interface between atmosphere and stored gas (m<sup>2</sup>), and
- $\Delta P_{O_2}$  = mean difference of partial pressure of O<sub>2</sub> calculated using Eq. 10 (MPa).

$$\Delta P_{O_2} = 0.21 - \left( \frac{O_{2i} + O_{2i-1}}{2} \right) P_t \quad (10)$$

where  $P_t$  = total pressure (MPa).

**Table I: The average standardized slope  $\alpha_s$  and the time required for a half initial differential pressure drop for each bag tested**

Bag Type	Bag no	$\alpha_s$		$t_{1/2s}$ (min)	
		Rep1	Rep2	Rep1	Rep2
A	1	-0.00050	-0.00710	582.7	411.6
A	2	-0.00087	-0.00089	336.7	330.1
B	1	-0.00861	-0.00825	34.0	35.5
B	2	-0.00586	-0.00510	50.0	57.5
C	1	-0.00897	-0.03925	22.0	7.5
C	2	-0.00626	-0.00932	35.6	31.4

**Table II: O<sub>2</sub> permeability measured for each bag tested**

Bag Type	Bag no	Permeability (mL·h <sup>-1</sup> ·m <sup>-2</sup> ·MPa <sup>-1</sup> )		Mean permeability by type (mL·h <sup>-1</sup> ·m <sup>-2</sup> ·MPa <sup>-1</sup> )
		Rep1	Rep2	
A	1	14.8	25.7	
A	2	17.8	35.6	23.7
B	1	539.0	575.5	
B	2	546.9	617.0	562.7
C	1	105.6	322.8	
C	2	32.6	228.0	168.8

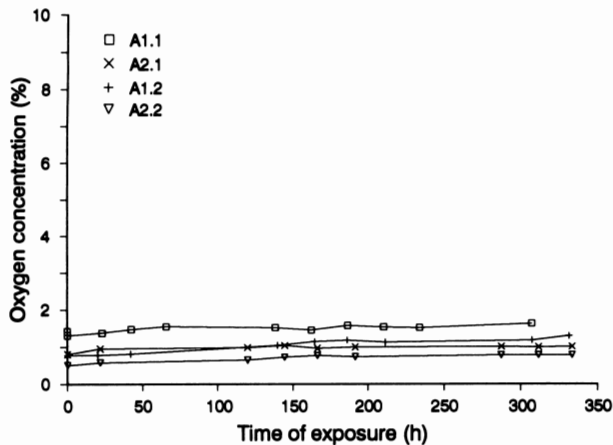


Fig. 4. Oxygen concentration as a function of time for trials of double polyethylene and aluminium bags (Type A). The legend refers to: Type, bag number - replicate.

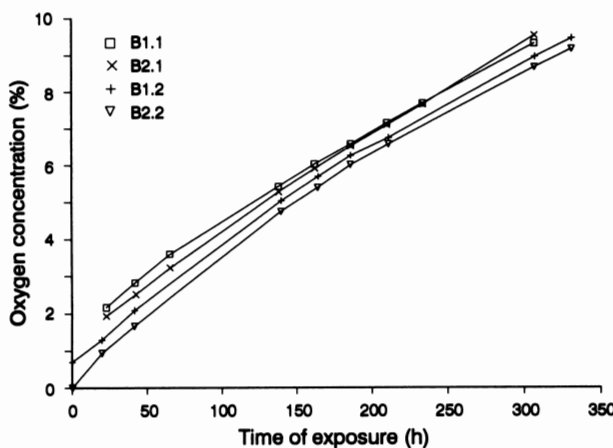


Fig. 5. Oxygen concentration as a function of time for trials of double polyethylene bags (Type B). The legend refers to: Type, bag number - replicate.

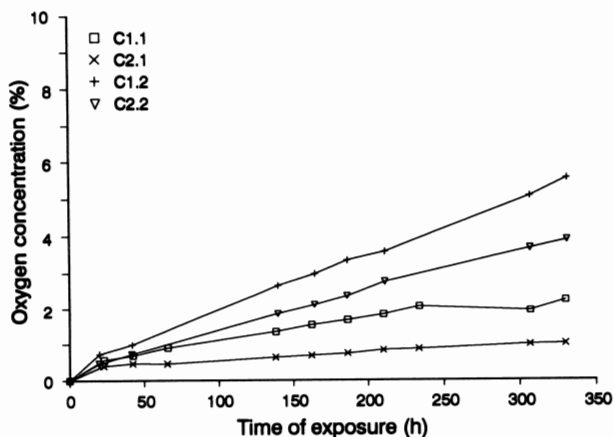


Fig. 6. Oxygen concentration as a function of time for trials of multi-layer barrier bags (Type C). The legend refers to: Type, bag number - replicate.

The  $O_2$  permeability results are presented in Table II. An analysis of variance was performed on the permeability data. At the 99.9% confidence level, the analysis revealed that the differences in permeability from type to type are significant. On average, Type B and Type C bags were respectively 24.5 times and 7.3 times more permeable to  $O_2$  than Type A bags. The differences between bags of a given type were not significant at the 95% confidence level.

The mean permeability of Type C presented here is about 10 times larger than the membrane permeability specified by the manufacturer. The observed tendency of the permeability of Type C bags to increase with time and/or manipulations can very well explain this discrepancy.

## CONCLUSIONS

Simple methods of measuring airtightness and  $O_2$  permeability of breather bags have been presented. Based on the results of this study, the following conclusions were drawn:

1. The method used to measure the airtightness of the breather bag was adequate to permit the elimination of the weak systems having a  $t_{1/2s}$  lower than "30 minutes".
2. A theoretical model was developed to analyze the airtightness data. Based on our model, it was shown that the Bartsch and Blanpied (1984) technique gives at best a rough approximation of  $t_{1/2s}$ .
3. The method used to measure the permeability of the breather bag was adequate and permitted identification of differences between the three types of bags.
4. The double polyethylene and aluminum bag used for wine storage is much more appropriate as a breather bag, since its global permeability is only 4% of that of the double polyethylene bag previously used by Vigneault et al. (1991).

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## REFERENCES

- ASHRAE. 1982. Vegetables. In *Applications Handbook*, Chapter 33. American Society of Heating, Refrigeration, and Air-conditioning Engineers, Atlanta, GA.
- Bartsch, J.A. and G.D. Blanpied. 1984. Refrigeration and controlled atmosphere storage for horticultural crops. Report NRAES-22. The Northeast Regional Agricultural Engineering Services, Cornell University, Ithaca, NY.
- Van Wylen, G.J. and R. Sonntag. 1959. *Fundamentals of classical thermodynamics*. New York, NY: John Wiley and Sons Inc.
- Vigneault, C., R. Granger and G.S.V. Raghavan. 1991. Mini-chamber for lab-scale research on controlled atmosphere storage. *Applied Engineering in Agriculture* 1(7):5-9.