Mathematical Modelling of Oxygen and Carbon Dioxide Composition in the Interstitial Atmosphere of Silo-Bags

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ABSTRACT

The CO_2 and O_2 concentration in hermetic storage systems depends on the balance between respiration of the ecosystem (consumption of O_2 and generation of CO_2), the entrance of external O_2 to the system, and the loss of CO_2 to the ambient air. A preliminary model was developed to simulate the gas exchange in silo-bags, considering the aerobic grains and microflora CO_2 production and neglecting the contribution by insect respiration. Dependence of the rate of CO_2 production on grain temperature, moisture content and storage time was taken into account and evaluated by use of the correlation developed by White et al. (1982) for wheat. The influence of initial grain temperature (25, 30 and 43ºC) and moisture content (12 -16 % w.b.) as well as degree of gas-tightness of the silo-bag was investigated. The model was run for the climatic conditions of the south east of Buenos Aires province, Argentina, for a storage period of 6 months, from January to July. The gas concentration predicted by the model under different storage conditions was compared with data available from a series of field experiments developed by INTA-EEA Balcarce. Globally, predicted values were in good accordance with the measured levels of gas concentrations. For all temperatures, at 12% w.b, the CO_2 concentration remained below 4%. At 30ºC and 14% w.b., CO_2 level increased to 12%. At 15% and 16% w.b, O_2 was consumed after 80 and 40 days, respectively. CO_2 reached 21% and then, because of the loss to ambient air (differential permeability for O_2 and CO_2), it fell below 16%. At 43ºC, O_2 was consumed after 80 and 25 and 10 days, for 14, 15 and 16% w.b. respectively. Under these conditions, CO_2 reached 21% and then fell below 15%.

Keywords: Hermetic storage, modified atmosphere, wheat, Argentina.

1. INTRODUCTION

In 2007 about 35 million tonnes of grains were stored in hermetic systems (silo-bags) in Argentina, and about 5 million tonnes corresponded to wheat. The wheat stored in these silo-bags is mostly used for milling (internal and external market), but also for seeds for the next planting season. This technique; originally used for grain silage, consists in storing dry grain in hermetically sealed plastic bags. The respiration process of the biological agents in the grain ecosystem (grain, insects, mites and microorganisms) increases carbon dioxide (CO_2) and

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reduces oxygen (O_2) concentrations. This modified atmosphere inhibits the biotic activity, promoting a suitable environment for grain conservation.

In a conventional bin (round bin made of concrete or corrugated metal sheets), any temperature increase detected at the core by thermocouples is associated to local heating due to respiration of the ecosystem and spoilage. The silo-bag has a higher ratio of transfer area/grain volume compared to that of conventional bins (1.42 m^2/m^3 and 0.79 m^2/m^3, respectively) (Bartosik et al., 2008). Thus, the temperature change at the core resulting from the balance between the heat released by respiration and heat transferred to the environment is highly influenced by the external temperature (Rodríguez et al., 2001, 2002), and therefore, temperature monitoring is not reliable to detect biological activity in silo-bags. Recently, a procedure for monitoring biological activity and storability based on carbon dioxide measurement in the interstitial air of grains stored in silo-bags was implemented (Bartosik et al., 2008; Cardoso et al., 2008, Rodríguez et al. 2008). However, a better understanding of typical CO_2 concentrations in silo-bags is required to develop a reliable technology for monitoring grain storability.

Simulation models are very usefull to rapidly analyze numerous situations and describe the critical limits of the different factors in view of the complexity of the grain bulk ecosystem prevailing under hermitic conditions, as it was mentionned by Navarro et al. (1994).

A comprehensive model of heat and moisture transfer for silo-bags deals with the respiration of the biological agents that modifies the atmospheric gases composition in the interstitial air of silo-bags. All the chemical components involved in the respiration process must be accounted for: water vapor, carbon dioxide, oxygen and non-reacting elements in the interstitial space. The problem statement is described by momentum, mass, and energy component that must balance out with those of the grain substrate. Inclusion of moisture and temperature dependent properties as well as realistic boundary conditions in the model definition makes the partial differential equations nonlinear and solution must be performed by numerical methods.

Unlike what was found for fruits and vegetables (Fonseca et al., 2001; Song et al., 2002), correlations to model the rate of CO_2 production depending on O_2 and CO_2 concentrations of the atmosphere yet have to be developed to properly account for grain respiration under the anaerobic conditions of silo-bags. The correlations available in literature for wheat (White et al., 1982), corn (Bern et al., 2002) and soybean (Rukudunin et al., 2004) were developed under aerobic conditions. They depend on temperature, moisture content and storage time, but not on gas concentrations.

This work is part of a general study that aims to develop a comprehensive model for the silo-bag storage system, taking progressively into account all the aspects mentioned above. In the first stage of the study a heat and mass transfer model for non-aeration periods was adapted to the silo-bag (Gastón et al., 2007, 2009). The model was used to analyze the storage of wheat and validated by comparing predicted and measured temperatures data (provided by The National Institute of Agricultural Technologies (INTA) of Argentina, Balcarce Experimental Station (Rodriguez et al., 2001)). This is the second, and more comprehensive, stage of the study that aimed:

1. to develop a mathematical model to predict the change of O_2 and CO_2 concentrations taking into account grain respiration and permeability of the plastic bag. The correlation developed by White et al. (1982) for wheat was applied to model the CO_2 production rate due to grain and microflora respiration.

2. to carry out a preliminary validation of the simulation model with experimentally available measured CO_2 and O_2 concentrations in wheat silo-bags.

2. MATERIALS AND METHOD

2.1 Silo-bags

Silo-bags are 60 m long, 2.70 m diameter and 230 - 250 microns thick. The bags are made of a three-layer plastic, black in the inner side and white in the outer side with UV stabilizers. The plastic layers are a mixture of high dense (HDPE) and low dense polyethylene (LDPE). Approximately 200 tonnes of grains (wheat, corn and soybean) can be held in the bag and usually farmers store their production during six to eight months.

2.2 Mathematical modelling

The CO_2 and O_2 concentration in the silo-bag depends on the balance between respiration (consumption of O_2 and generation of CO_2), the entrance of external O_2 to the system, and the loss of CO_2 to the ambient air.

The transfer of gases depends on the gas partial pressure differential and the effective permeability of the plastic cover (openings and natural permeability of the plastic layer to gases). There is a strong interaction between the external factors and the interrelated grain bulk ecosystem components in the silo-bag. Grain type and condition, MC, temperature, storage time and O_2 and CO_2 concentrations affect the respiration rate.

In a first approach, a lumped model was adopted to calculate the change of the mean concentration of CO_2 and O_2 in the silo-bag (Navarro et al., 1994). The following assuptions were considered to simplify the mathematical model:

(1) CO_2 and O_2 are distributed uniformly throughout the grain mass and no stratification occurs.
(2) the contribution to CO_2 production by insects respiration and CO_2 sorption by grain are not included in the present study.
(3) the temperature is uniform throughout the grain mass and its evolution during storage according to seasonal variation of weather conditions is known.
(4) moisture content remains equal to the initial value.
(5) a resistence series model was applied to derive an effective permeability of the plastic layer.
(6) no head-space volume exists, the volume of the silo-bag is occupied by the grain mass.

2.2.1 O_2 and CO_2 mass balance in the silo-bag

Stating the mass balances of the species involved in the respiration of the grain in a volume of one meter long of the silo-bag shown in Figure 1, the following coupled system is obtained (Song et al., 2002):

$$\frac{dO_{2i}}{dt}=\frac{A_S\,P_{O_2}\,P_{atm}}{\varepsilon V\,L}\left\{O_{2out}-O_{2i}\right\}-\frac{m_{grain}}{\varepsilon V}r_{O2} \qquad in\ \Omega_1 \tag{1}$$

$$\frac{dCO_{2i}}{dt}=\frac{A_S\,P_{CO_2}\,P_{atm}}{\varepsilon V\,L}\left\{CO_{2out}-CO_{2i}\right\}+\frac{m_{grain}}{\varepsilon V}r_{CO2} \qquad in\ \Omega_1 \tag{2}$$

where O_2 and CO_2 (V/V m^3/m^3) are oxygen and carbon dioxide concentrations; P_{O2} and P_{CO2} in $m^3 md^{-1} m^{-2} at^{-1}$, are the permeability of the plastic to O_2 and CO_2, respectively; P_{atm} in Pa, is atmospheric pressure; L in m, is the thickness of the plastic layer, A_s in m^2, is the transfer area of the silo-bag, m_{grain} in kg, is the dry mass of grain, εV in m^3, is the free volume; ε is the grain bed porosity; V in m^3, is the total volume of the silo-bag; r_{O2} and r_{CO2} in m^3/s kg dry matter, are the rate of consumption of O_2 and production of CO_2, respectively. The first term on the right side of Eq. (1) and Eq. (2) accounts for the gas exchange through the plastic bag. The second one, the O_2 sink and CO_2 source as result of the respiration process. For short, the following constants were defined, where ρ_s in kg/m^3, is the grain bulk density:

$$c_1=\frac{A_S\,\overline{P}_{O_2}\,P_{atm}}{\varepsilon V\,L} \quad ; \quad c_3=\frac{A_S\,\overline{P}_{CO_2}\,P_{atm}}{\varepsilon V\,L} \quad ; \quad c_2=\frac{m_{grain}}{\varepsilon V}=\frac{\rho_s V}{\varepsilon V}=\frac{\rho_s}{\varepsilon} \tag{3}$$

The sink and source terms can be expressed using the following respiration equation, based on the oxidation of hexose:

$$\begin{aligned}&C_6H_{12}O_6+6O_2\rightarrow 6CO_2+6H_2O+2835\,kJ/mol\\&180\,gr\ +\ 192\,gr\rightarrow 264\,gr+108\,gr+2835\,kJ\end{aligned} \tag{4}$$

This expression shows that the rate of oxygen consumed equals that of carbon dioxide produced during aerobic respiration.

If the evolution of the average temperature of the silo-bag during the storage period is known, under the hypothesis that the average moisture content remains constant, the rate of O_2 and CO_2 consumption/production can be modeled by an explicit function of time:

$$\frac{dO_{2i}}{dt}=c_1\left\{O_{2out}-O_{2i}\right\}-c_2 r_{O2}(t) \qquad in\ \Omega_1 \tag{5}$$

$$\frac{dCO_{2i}}{dt}=c_3\left\{CO_{2out}-CO_{2i}\right\}+c_2 r_{CO2}(t) \qquad in\ \Omega_1 \tag{6}$$

Since the silo-bags contains air, the initial conditions (t = 0) become:

$$O_{2i}^{0}=0.21 \quad ; \quad CO_{2i}^{0}=0.0003 \tag{7}$$

Similarly, as the bag is placed in the field (air):

$$O_{2out} = 0.21 \quad ; \quad CO_{2out} = 0.0003 \tag{8}$$

Equations (5) and (6) form an ordinary differential equation system that was solved numerically.

2.2.2 Model respiration for wheat

White et al. (1982) carried out numerous experiments on the carbon dioxide released rates of cereal grains and have established robust models, expressed by the following generic equation, where Y_{CO2} is in mg/kg of dry matter in 24 h, θ is the storage time in days, T_c is grain temperature in ºC and M moisture content in % w.b.:

$$\log Y_{CO2} = -4.054 + 0.0406\, T_c - 0.0165\, \theta + 0.0001\, \theta^2 + 0.2389\, M \tag{9}$$

Equation (9) accounts for grain and microflora CO_2 production. In the present study, the contribution to CO_2 production by insect respiration is not included.

Owing to seasonal variation of weather conditions, the mean temperature of the silo-bag varies during storage. Thus, providing an explicit relationship for T_c as function of storage time, Eq. (9) becomes an explicit function of time since, M was assumed to remain at the initial moisture content. The rate of consumption of O_2 and production of CO_2 are expressed by:

$$r_{CO2} = \frac{Y_{CO2}}{1000 M_{CO2}} \frac{R T_k}{P_{at}} \quad ; \quad r_{O2} = r_{CO2} \tag{10}$$

2.2.3 Time evolution of the mean temperature of the silo-bag during storage

The time evolution of the temperature distribution of the silo-bag was calculated applying the heat and mass transfer model previously developed (Gastón et al., 2007), which is solved applying by the Finite Element Method. The mean temperature of the silo-bag was calculated by integration of the temperature distribution over the silo-bag domain, as follows:

$$\overline{T}_c(t) = \frac{1}{\Omega} \int_{\Omega} T(x,y,t)\, d\Omega \tag{11}$$

In this model, the heat released by respiration is modeled with White et al. (1982) relation, but as this equation is independent of CO_2 and O_2, the heat and mass transfer problem are decoupled form the gas balance analysis.

2.2.3 Effective permeability of silo-bag

Permeability at 25ºC of HDPE to O_2 is 6.5 $10^{-8} m^3 md^{-1} m^{-2} at^{-1}$ and of LDPE is 1.95 $10^{-7} m^3 m\, d^{-1} m^{-2} at^{-1}$. Permeability at 25ºC of HDPE to CO_2 is 1.9 $10^{-7} m^3 md^{-1} m^{-2} at^{-1}$ and of LDPE is 1.05 $10^{-6} m^3 md^{-1} m^{-2} at^{-1}$ (Osborn et al., 1992). A resistance series model was applied to derive an effective permeability to O_2 and CO_2. It was assumed that half of the plastic layer thickness was HDPE and the other LDPE polyethylene:

$$\overline{P}_i = \frac{2P_{iHD}\ P_{iLD}}{P_{iHD} + P_{iLD}} \quad ; \quad i = O_2, CO_2 \tag{12}$$

Effective permeability of the plastic layer to O_2 was 9.75 $10^{-8}m^3md^{-1}m^2at^{-1}$ and to CO_2 was 3.22 $10^{-7}m^3md^{-1}m^2at^{-1}$. For an average thickness of 240 μm, estimated permeance to O_2 was 4.06 $10^{-4}m^3d^{-1}m^2at^{-1}$ and to CO_2 was 1.34 $10^{-3}m^3d^{-1}m^2\ at^{-1}$, resulting 3 times that of O_2.

2.3 Experimental field tests

Recently, INTA-EEA Balcarce conducted a series of field experiments in order to identify the main factors affecting carbon dioxide (CO_2) and oxygen (O_2) concentrations as indicators of biological activity and appropriated wheat storability. The experiments consisted of monitoring the gas composition of the interstitial air, grain commercial quality, grain moisture content (MC), and grain temperature of several silo-bags The tests were carried out at grain elevators and farms in the south east of Buenos Aires province, Argentina, through three storage seasons (from January 2006 to May 2008). Most of the wheat silo-bags were filled in December-January and stored until June or July. A detailed discussion of the results of these tests are presented in Bartosik et al. (2008), where the authors derived a correlation between the measured CO_2 concentration in the silo-bag and the moisture content of the stored wheat.

3. RESULTS AND DISCUSSION

To validate the model, the CO_2 and O_2 concentration change inside the silo-bag was simulated and the numerical results were compared with the experimental data. Simulations were carried out for 12-16% w.b., typical moisture content range for wheat harvest. The initial temperature was assumed to be 25, 30 and 43ºC, which represent the minimum, average and maximum grain temperature of the grain stored in silo-bags during the harvest time. Wheat porosity was set to 0.38. For one meter long of the silo-bag, A_s= 5.54 m^2 and V = 4.54 m^3 (Figure 1). Figure 2 shows the dependency of the rate of CO_2 production with moisture content and temperatures. Rate of production doubles every 5ºC or every 1% w.b. increment.

First, the heat and mass transfer model was run for the climatic conditions of the south east of Buenos Aires province to determine the mean temperature change of the silo-bag from January to June. Figure 3 shows the mean temperature change of the silo-bag from summer to winter. The effect of the heat released by respiration did not have a strong effect (Gastón et al., 2009) being the difference in the mean temperature change for dry (12%) and wet (16%) at most 2ºC. This can be explained by the high energy exchange with the surroundings as a consequence of the high ratio of transfer area/grain volume of a silo-bag (~1.43 m^2m^{-3} for a 200 tonnes silo-bag).

The evolution of O_2 and CO_2 concentrations for 43ºC initial temperature are shown in Figure 4a. For wheat at 12 and 13% w.b. initial moisture content, CO_2 increased to 6 and 11% while O_2 decreased to 13 and 6%, respectively. For wheat at 14, 15 and 16% w.b. initial moisture content, 0% O_2 level is achieved after 80, 25 and 10 days, respectively. Thereafter, the O_2 concentration remained constant at 0% because the O_2 that ingresses through the plastic bag was consumed by respiration. CO_2 increased up to 18, 20 and 21%, and then started to decline as results of the losses through the cover as permeance to CO_2 was three times greater than to O_2. This figure also illustrates the effect of initial moisture content on the rate of respiration, reinforced by a high initial storage temperature. Results for 30ºC (Figure 4b) showed that 0% O_2 level was achieved only for 15 and 16% w.b., after 85 and 40 days while for 25ºC, after 100 and 70 days (Figure 4c).

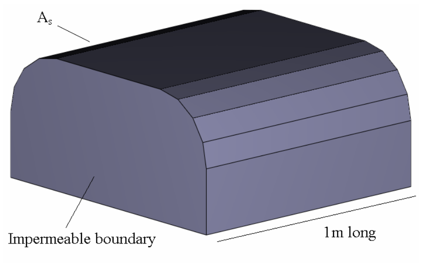


Figure 1. Schematic diagram of the silo-bag domain

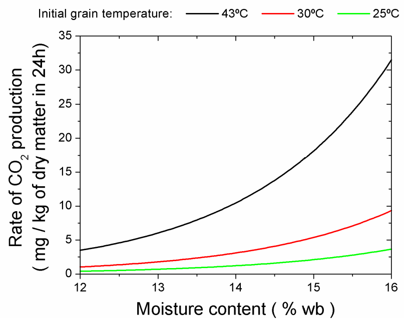


Figure 2. CO_2 production rate vs moisture content

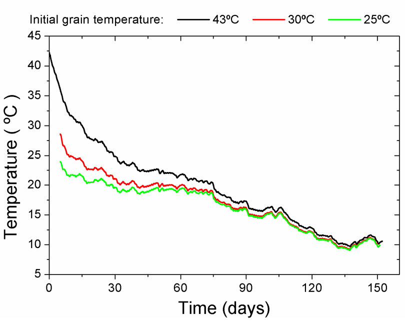
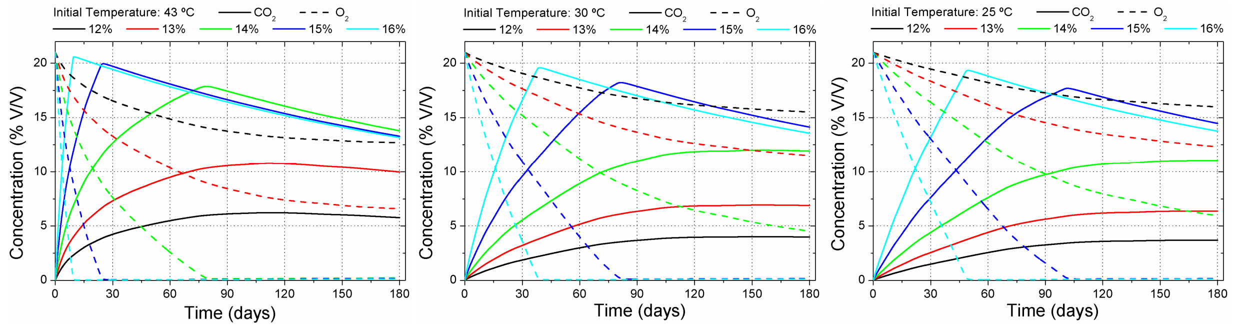


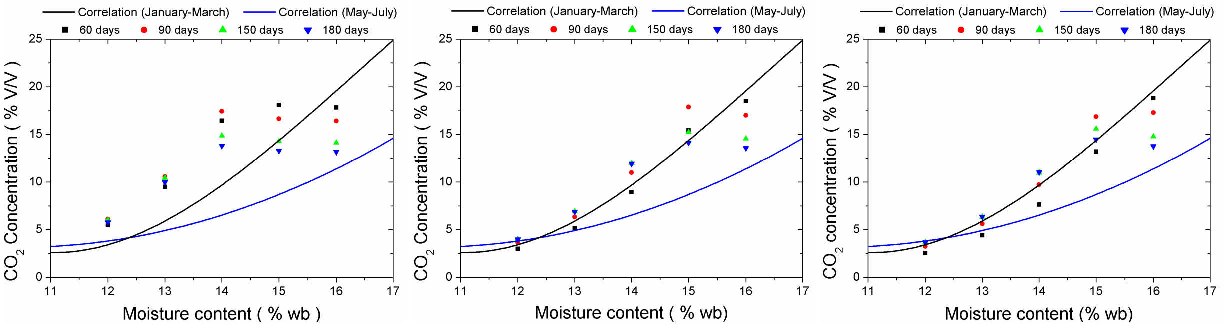
Figure 3. Mean temperature change of the silo-bag



(a) Initial Temperature 43ºC (b) Initial Temperature 30ºC (c) Initial Temperature 25ºC

Figure 4. Predicted O_2 and CO_2 concentrations in silo-bags storing wheat from January to July

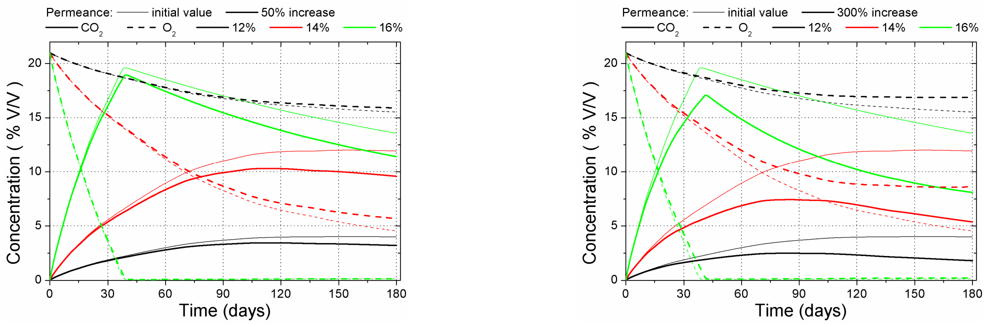
Figure 5 shows the correlations developed by Bartosik et al. (2008) between grain MC and CO_2 concentration, which collected data sampled during the warm and cold storage season from silo-bags with a wide range of initial storage conditions. The CO_2 concentration increased with the increase in grain MC, as a consequence of the biological activity. When the wheat MC was lower than 13% the average CO_2 concentration was below 5%. When the wheat MC increased to the point at which molds became active (between 13.5 and 14.5%) the CO_2 concentration increased to 15% with wheat at 16% MC, and to 30% with wheat MC above 19%. These data corresponds to silo-bags without visible structural problems. Since information about the initial storage grain temperature and moisture content was lacking, simulation were carried out for the range of storage conditions mention above to cover the most representative ones. The predicted evolution of CO_2 concentration was in accordance with experimental results. For dry wheat (12-13% w.b.) and 30ºC (average condition) CO_2 remained below 6%. Predicted values after 60, 90, 150 and 180 days are plotted in Figure 5. Predicted concentrations for the exceptionally high initial temperature of 43º present the greater deviation with both correlations for all the range of moisture contents. For 25ºC and 30ºC good agreement was obtained with the summer correlation. Concentrations were over predicted during the cold season for wet grain (14-16% w.b.).



(a) Initial Temperature 43ºC (b) Initial Temperature 30ºC (c) Initial Temperature 25ºC

Figure 5. Comparison between correlations and predicted CO_2 concentrations after 60, 90, 150 and 180 days

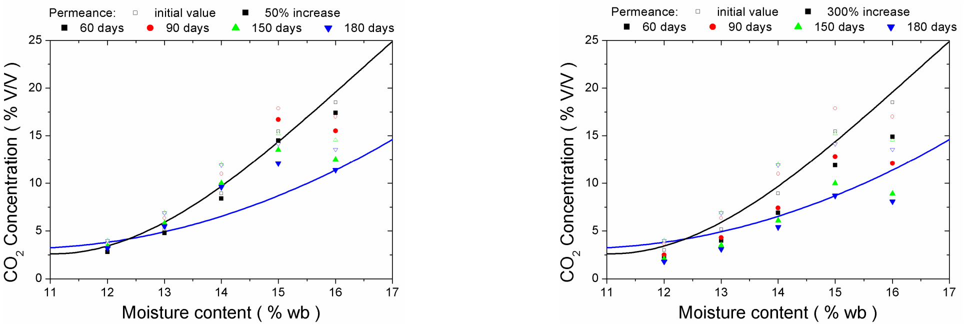
The effect of the plastic permeance to O_2 and CO_2 on model predictions was analyzed. In the first case, the initial estimated values were increased 50% and in the second one were set equal to that of LDPE. In the last case, permeances are three times greater (300%) that the initial values and may be representative of the worst permeability condition for a plastic cover without structural damage. For the initial permeance estimation and a maximum partial pressure difference of 21%, the O_2 inflow was about 0.027%/day and the CO_2 outflow of 0.09%/day. Results for the initial storage temperature of 30ºC are presented in Figure 6a and 6b, for 50% and 300% increase, respectively. Comparison of the predicted concentrations after 60, 90, 150 and 180 days with the summer and winter correlations are plotted in Figure 7a and 7b. Both figures include the prediction of O_2 and CO_2 concentration with the initial permeance values.



(a) 50% Permeance increase (b) 300% Permeance increase

Figure 6. Effect of permeance increase in the predicted O_2 and CO_2 concentrations

As expected, a 50% increase in CO_2 permeance produced a decrease in the concentrations levels of 2.5% points for 14-16% w.b. range, while for 12% w.b. at most 1% point. For 300%, concentration drop was 5% point for wet grain and of 2.5% points for dry grain. These results show that an increase of 50% improves the overall model fitness with the summer and winter correlations. When permeance of LDPE is used, results for winter correlation are improved but those for summer are worsen.



(a) 50% Permeance increase (b) 300% Permeance increase

Figure 7. Effect of permeance increase in the comparison between correlations and predicted CO_2 concentrations after 60, 90, 150 and 180 days

4. CONCLUSIONS

A preliminary model was developed to simulate the gas exchange in silo-bags, considering the aerobic grains and microflora CO_2 production and neglecting the contribution by insect respiration. The rate of wheat CO_2 production was calculated with the correlation developed by White et al. (1982), which depends on grain temperature, moisture content and storage time.

The model was implemented for the climatic conditions of the south east of Buenos Aires province, Argentina, from January to July. The gas concentration predicted by the model under different storage conditions were compared with experimental correlations developed by INTA-EEA Balcarce. Globally, predicted values were in good accordance with the measured levels of gas concentrations.

The simulations showed that for wet grain and high initial temperature O_2 is consumed in a few days. As initial temperature decreases it may take between one to three months to achieve anaerobic conditions. When dry grain is stored O_2 level remained above 10% and CO_2 level below 4%. For wet grain CO_2 level were in the range 15-16% after six month storage.

The measurement of the plastic permeance is a key parameter to improve the model performance to assist in the design of a monitoring protocol of O_2 and CO_2 concentrations as a tool for predicting grain storability for the silo-bag system.

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