Mathematical modelling of oxygen and carbon dioxide concentration profiles in the interstitial atmosphere of silo-bags

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Summary
In this work, O_2 and CO_2 concentration in the interstitial atmosphere of silo-bags predicted by a diffusion model are compared with values obtained by use of the lumped capacitance model. The first one uses local values of grain temperature and moisture content to evaluate the grain respiration rate while the second applies average values of these magnitudes. Comparison were carried out for wheat stored at initial temperatures of 20, 25, 30 and 40C, MC in the range (12 -16 % w.b), under climatic conditions of the South East of Buenos Aires province, Argentina. For uniform initial moisture content, the difference between predicted mean concentration of CO_2 and O_2 were less than 1point %.Further, the diffusion model was applied to analyze local effects in the silo-bag: the evolution of gas concentration when there is a spot of wetter grain (16% w.b) than the rest (13% w.b) at the bottom the bag; the penetration of O_2 through a small hole located at the top surface in a silo-bag initially in anaerobiosis.

Key word: silo-bags, hermetic storage, modified atmosphere, wheat, modelling

Introduction

In 2010 more than 40 million tonnes of grains were stored in hermetic systems (silo-bags) in Argentina. 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 reduces oxygen (O_2) concentrations, promoting a suitable environment for grain conservation.
Gas concentration depends on the balance between respiration of the ecosystem, 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). Grain type and condition, MC, temperature, storage time and O_2 and CO_2 concentrations affect the grain respiration rate.
A novel technology for monitoring grain storability in silo-bags based on CO_2 detection was implemented by The National Institute of Agricultural Technologies of Argentina (INTA), Balcarce Experimental Station (EEA) (Bartosik et al., 2008; Cardoso et al., 2008). The procedure consists of comparing the measured CO_2 concentration at some locations in the silo-bag (local concentration value) with a referential value which represents adequate storage conditions. However, a better understanding of typical CO_2 concentrations profiles for a range of storage conditions in silo-bags (i.e: grain temperature, moisture content (MC), storage time, etc) is required to define sampling location and monitoring frequency of the silo-bag. This information is relevant to develop a reliable technology for controlling grain storability based on CO_2 detection.
Simulation models are very useful to rapidly analyze numerous situations and can describe the effect of different factors affecting the grain stored under hermitic conditions, especially in this case where there is a strong interaction between the external factors and the interrelated grain bulk ecosystem components in the silo-bag. Recently, the authors developed a lumped capacitance model to calculate the change in the mean O_2 and CO_2 concentrations in a wheat silo-bag, taking

into account simultaneous oxygen consumption, carbon dioxide generation, and permeability to gas transfer of the plastic bag. This model runs in combination with a 2D heat and mass transfer model which predicts grain storage temperature and moisture content (MC) as function of weather conditions. Both models were validated by comparison of predicted temperature, grain moisture content, mean O_2 and CO_2 concentrations with measured values in field test (Gastón et al.,2009; Abalone et al., 2009, 2010a, 2010b).
In present work, the diffusion of gases was incorporated into the model in order to predict gas distribution in the silo-bag and quantify the effect of averaging local temperature variations and moisture migration in the prediction of mean gas concentrations. The model was also applied to study local effects such as a non uniform initial moisture content distribution as well as the damage of the plastic layer (perforations) in the evolution of gas concentration.

Material and methods

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.

Mathematical modelling

Stating the energy and mass balances (moisture, CO_2 and O_2) for the grain and air phases in a control volume, the following coupled system is obtained:

$$c_b\rho_b\frac{\partial T}{\partial t}=\left[\frac{\partial}{\partial x}\left[k_b\frac{\partial T}{\partial x}\right]+\frac{\partial}{\partial y}\left[k_b\frac{\partial T}{\partial y}\right]\right]+\rho_{bs}L_g\frac{\partial W_g}{\partial t}+\rho_{bs}q_H Y_{CO2} \qquad in\ \Omega_1 \tag{1}$$

$$\rho_{bs}\frac{\partial W_g}{\partial t}=\frac{\partial}{\partial x}\left[D_w\left(\eta\frac{\partial W_g}{\partial x}+\omega\frac{\partial T}{\partial x}\right)\right]+\frac{\partial}{\partial y}\left[D_w\left(\eta\frac{\partial W_g}{\partial y}+\omega\frac{\partial T}{\partial y}\right)\right]+\rho_{bs}q_w Y_{CO2} \quad in\ \Omega_1 \tag{2}$$

$$\varepsilon\frac{\partial CO_2}{\partial t}=\frac{\partial}{\partial x}\left[D^*_{CO2}\left(\frac{\partial CO_2}{\partial x}\right)\right]+\frac{\partial}{\partial y}\left[D^*_{CO2}\left(\frac{\partial CO_2}{\partial y}\right)\right]+\rho_{bs}r_{CO2} \quad in\ \Omega_1 \tag{3}$$

$$\varepsilon\frac{\partial O_2}{\partial t}=\frac{\partial}{\partial x}\left[D^*_{O2}\left(\frac{\partial O_2}{\partial x}\right)\right]+\frac{\partial}{\partial y}\left[D^*_{O2}\left(\frac{\partial O_2}{\partial y}\right)\right]+\rho_{bs}r_{O2} \quad in\ \Omega_1 \tag{4}$$

where T in K is temperature, W_g in d.b is grain moisture content (*MC*), O_2 and CO_2 (% V/V) are oxygen and carbon dioxide concentrations, ε porosity, ρ_{bs} in kg m^{-3} is dry bulk density, c_b in Jkg^{-1} K^{-1} is bulk specific heat, k_b in W m^{-1} K^{-1} is bulk thermal, D_w is an effective diffusivity parameter for water vapour, L_g in J kg^{-1} is the latent heat of vaporization of moisture in the grain, η in Pa is the change in the partial pressure due to change in the *MC* at constant temperature, ω in Pa K^{-1} is the change in the partial pressure due to change in the temperature at constant *MC*, D^*_i in m^2s^{-1} (with i= CO_2 and O_2) is the effective diffusivity through the intergranular air of carbon dioxide and oxygen calculated according to Geankoplis (1998).
In Eqs. (1) to (4), last terms represents heat, water vapour and carbon dioxide released and oxygen consumed, respectively, owing to respiration of the grain ecosystem. Respiration is modeled by the complete combustion of a typical carbohydrate. Y_{CO2} is the rate of CO_2 production, in mg [CO_2] kg^{-1} [dry matter] s^{-1}, q_H is 10.738 J mg^{-1} [CO_2], q_w is 4.09 10^{-5} kg [H_2O] mg^{-1} [CO_2]. The rate of CO_2 production r_{CO2} in m^3s^{-1} kg^{-1} [dry matter] is given by:

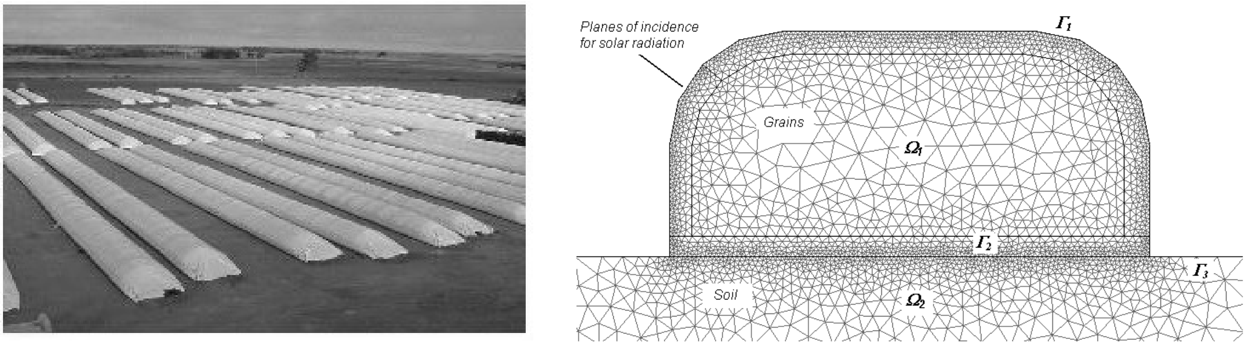
$$r_{CO2} = \frac{Y_{CO2}}{1000M_{CO2}} \frac{RT}{P_{at}} \quad ; \quad r_{O2} = r_{CO2} \tag{5}$$

Fig. 1 shows the calculation domain, which represents a cross section of the silo-bag. Boundary conditions take into account the interaction between the soil and the bottom layer of the silo-bag, solar radiation, convection and radiation. It was assumed that the silo-bag was impermeable to moisture transfer. Expressions for initial and boundary conditions associated to Eq. (1) and Eq. (2), model functions D_w, η, ω, L_g, input parameters of the thermal model and as well as bulk properties for wheat are presented in detail in Gastón et al. (2009).
Boundary conditions associated to Eq.(3) and Eq.(4) consider gas transfer through the plastic layer:

$$-D^{*}_{CO2} \frac{\partial CO_2}{\partial n} = \frac{P_{CO2} P_{atm}}{L} \left(CO_2 - CO_{2out} \right) = h_{CO2} \left(CO_2 - CO_{2out} \right) \quad on \quad \Gamma_1 \tag{6}$$

$$-D^{*}_{O2} \frac{\partial O_2}{\partial n} = \frac{P_{O2} P_{atm}}{L} \left(O_2 - O_{2out} \right) = h_{O2} \left(O_2 - O_{2out} \right) \quad on \quad \Gamma_1 \tag{7}$$

where P_{atm} in Pa, is atmospheric pressure; L in m, is the thickness of the plastic layer, P_{O2} and P_{CO2} in $m^3 ms^{-1} m^{-2} at^{-1}$, are the equivalent permeability of the plastic to O_2 and CO_2, respectively, both calculated by use of a resistance series model to take into account that the silo-bag is a mixture of high dense (HDPE) and low dense polyethylene (LDPE) (Abalone et al., 2009, 2010a).



a) b)

Figure 1: a) Picture of the silo-bag storage system; b) Schematic diagram of the calculation domain and boundaries. Cross section of the silo-bag.

Lumped model

In the lumped model, Eqs (1) and (2) are retained to obtain the evolution of the mean temperature of the silo-bag while Eqs (3) and (4) are integrated over the total volume resulting in:

$$\frac{d\overline{O_2}}{dt} = K_{O_2} \frac{\left(O_{2out} - \overline{O}_2 \right)}{\varepsilon V} - \frac{\rho_{bs}}{\varepsilon} \bar{r}_{O2} \qquad in\ \Omega_1 \tag{8}$$

$$\frac{d\overline{CO_2}}{dt} = K_{CO_2} \frac{\left(CO_{2out} - \overline{CO_2} \right)}{\varepsilon V} + \frac{\rho_{bs}}{\varepsilon} \bar{r}_{CO2} \qquad in\ \Omega_1 \tag{9}$$

The sink/source terms are evaluated at the mean temperature and moisture content of the grain. The former changes according to seasonal variation of weather conditions while the latter remains equal to the initial value as the silo-bag is considered impermeable to water vapour.

Numerical solution

The mathematical models were implemented using *COMSOL Multiphysics 3.5a* and solved numerically by the finite element method. Quadratic Lagragian elements and a fourth order numerical quadrature were applied. UMFPACK solver was selected to solve the PDE system (unsymmetrical multifrontal method and direct sparse LU factorization).

Results and Discussion

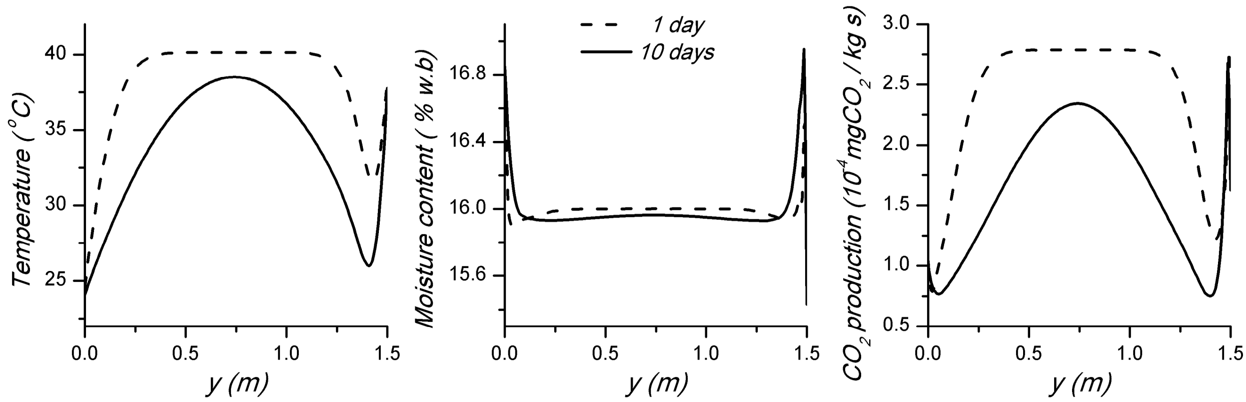
The models were applied to analyze the storage of wheat in a silo-bag from January to June (six months). Climatic conditions of the South East of Buenos Aires province, Argentina were considered. Initial grain temperatures were 20, 25, 30 and 40C. Initial grain MC ranged from 12 to 16 % w.b.
Dependence of the rate of CO_2 production Y_{CO2} was evaluated by use of the correlation developed by White et al. (1982) which takes into account grain temperature, MC and storage time:

$$log\,Y_{CO2} = -4.054 + 0.0406\,T - 0.0165\,\theta + 0.0001\,\theta^2 + 0.2389\,M \quad (10)$$

Equivalent permeability of the plastic layer to O_2 was set equal to 9.75 $10^{-8} m^3 md^{-1} m^2 at^{-1}$ and to CO_2 equal to 3.22 10^{-7} $m^3 md^{-1} m^2 at^{-1}$. Average plastic thickness L is 240 μm. For one meter long of silo bag, transfer area equals A= 5.54 $m^2$and volume to V = 4.54 m^3.
Reported values for effective diffusivity of carbon dioxide through wheat ranged from 3.7 10^{-6} m^2s^{-1} (Ileleji et al., 2006) to 7.610^{-6} m^2s^{-1} (Shunmugan et al., 2005). In this work D^*_{CO2} was set equal to 3.97 10^{-6} m^2s^{-1} and D^*_{O2} to 5.22 10^{-6} m^2s^{-1}. Wheat porosity ε is 0.38 and tortuosity τ is 1.53.

Predicted results with diffusion model

Results corresponding to the worst initial condition of 40C and 16 % w.b (hot and wet grain) were selected to illustrate model predictions. In this case, as will be shown afterwards, O_2 is consumed during the first two weeks of storage.
Fig.2 shows typical profiles of temperature (a), MC (b) and CO_2 production (c), along a vertical line located at the centre of the silo-bag cross section. The abscissa y represents the distance from the bottom of the silo-bag. The plots correspond to midday of first and tenth day of storage.
Temperature fluctuations within the first 30 cm below the surface (Fig. 2a) reflect the effect of ambient conditions as well as hourly weather data. Note that at the centre, in spite of the energy released by respiration, no temperature rise occurs because of the high ratio of energy transfer area/grain volume for the silo-bag (Gastón et al., 2009).
Moisture migration occurs in opposite direction to temperature gradients driving a redistribution of about 1 point w.b, localized within 10 cm at the top and bottom of the silo-bag (Fig. 2b).
The rate of CO_2 production shown in Fig. 2c follows the temperature trend. At the centre, the rate of production decreases, prevailing the effect of temperature decrease over the slight increase in MC (see Eq.10). In the upper layer, the behaviour is much more complex as result of the local fluctuations of temperature and moisture content.



a b c

Figure 2: Profiles of temperature (a), moisture content (b) and respiration rate (c) along the middle vertical section of the silo-bag. Initial storage conditions 40C and 16%w.b.

Fig.3 illustrates the gas concentration profiles. Although at the centre, respiration is about two to three times greater than at the bottom or surface (Fig. 2c) the concentration gradients are small because the high diffusion of gases in the silo-bag flattens the profiles of O_2 and CO_2. Differences in the order of 0.05 points% would hardly be detect in large scale field test. These numerical results are in accordance with experimental data which showed that no stratification along the vertical direction was measured during sampling gas concentration in silo-bags (Bartosik et al., 2008; Cardoso et al., 2008).

This behavior can be explained by analyzing the mass Biot Number $Bi = V h_i / A D_i^*$, the ratio between internal to external resistance to gas transfer (*i* = CO_2, O_2). Calculated values of *Bi* for CO_2 and O_2 resulted both in the order of 10^{-4}. A value smaller that 0.1 indicates that the gas concentration distribution inside the silo-bag will be almost uniform. This means that gas sampling at one or two locations would be enough to obtain reliable information of the grain storability condition, at least in the cross section of the silo-bag.

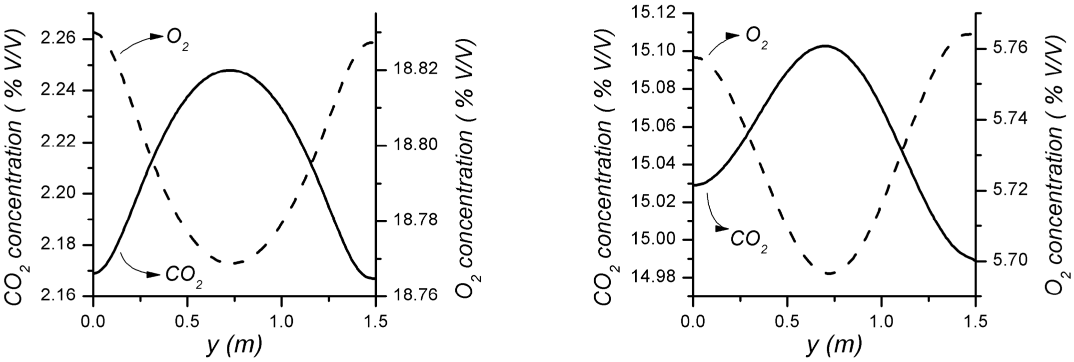


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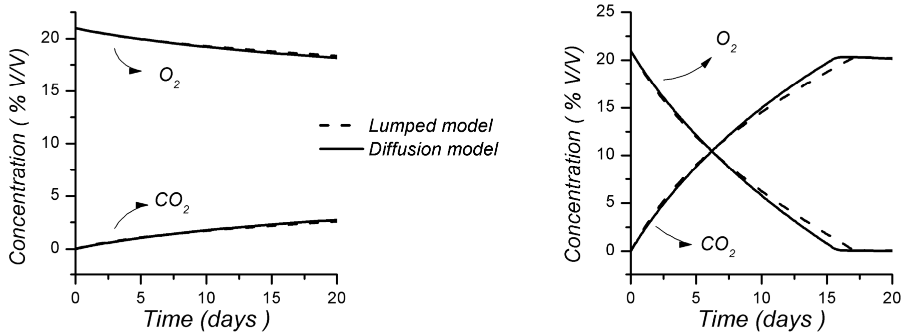
Figure 3: Profiles of O_2 and CO_2 concentration along the middle vertical section of the silo-bag for 40ºC and 16%w.b initial conditions for the first (a) and tenth day of storage (b).

Comparison of predicted mean gas concentration by use of the diffusion and the lumped model

For each time step, the local O_2 and CO_2 concentration determined by Eq. (3) and (4) were averaged over the domain to obtain mean concentration values.

For each pair of initial condition in the range (20-40C, 12 -16 % w.b), the time evolution of mean gas concentration was compared with the corresponding value calculated applying the lumped model. This comparison quantifies the effect of averaging local temperature variations and moisture migration by evaluating the CO_2 production (Eq.10) using mean values of temperature and the initial moisture content.

Fig. 4 illustrates the comparison for (40C -12%w.b) and (40C-16 %w.b), the worst range of bagging conditions. For dry and hot wheat, differences are negligible. For wet wheat, the lumped model slightly over predicts at the beginning while under predicts at the end the O_2 decrease, being the difference less than 1 point %. For CO_2 accumulation the opposite trend is obtained.



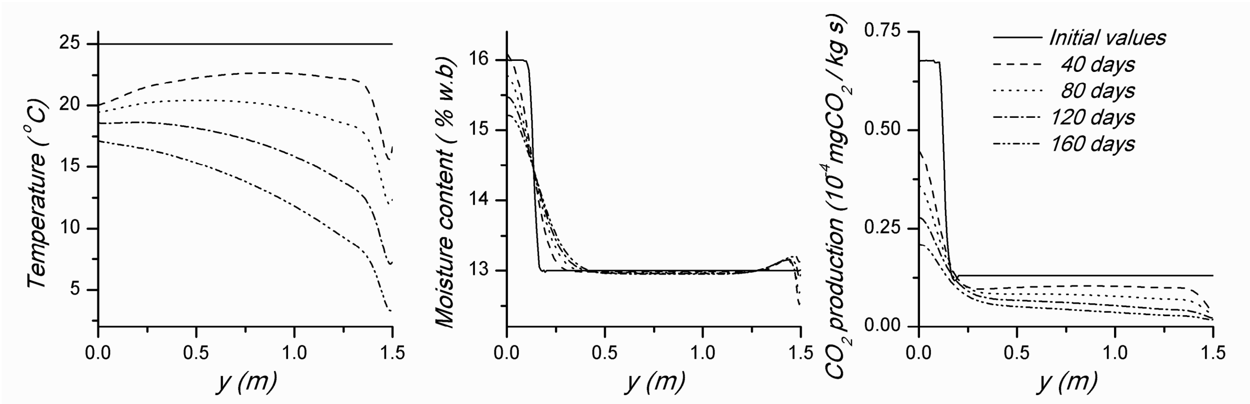
a) b)

Figure 4: Comparison between O_2 and CO_2 mean concentration evolutions predicted by use of the diffusion and lumped models. Initial grain temperature 40C. MC 12%w.b (a). MC 16%w.b (b).

The evolution of concentration values at different locations in the bag (not shown in Fig.4) is almost equal to the evolution of the mean one. It was mentioned that local information is obtained during gas sampling in the field. Therefore, it can be concluded that the lumped model is adequate for analyzing the change in gas concentration in a silo-bag when the initial moisture content distribution is assumed uniform. A difference less than 1% point such as the one obtain between both models, is in the order of magnitude of experimental errors involved in measuring gas composition in large scale field tests.

Effect of a non uniform MC distribution

A close examination of any given silo-bag in the field typically reveals a number of small perforations in the plastic cover, produced by wild animal, crop residues, etc. Water infiltration through them at the bottom of the bag can create a spot of wet grain. So, it is useful to study the evolution of gas concentration in such condition by numerical simulation.
A typical bagging condition for wheat in the South East of Buenos Aires province is 25C and 13% w.b. This condition was set as a base case. To simulate a wet spot in the bag, the diffusion model was run setting a layer 15 cm thick of grain to 16% w.b at the bottom while the rest to 13% w.b. This layer represents 10% of the grain stored per unit length of the silo-bag.
Fig. 5 shows temperature (a), moisture content (b) and CO_2 production profiles (c) every 40 days of storage. Again, although there is a spot of wet grain, temperature does not rise at the bottom of the bag. Moisture slowly migrates towards the centre. Comparison of Fig. 5c and Fig. 2c shows that the rate of CO_2 production for the base case (25C - 13% w.b MC) is significantly smaller than for (40C - 16% w.b MC). At 16% w.b MC, production rate at 25C is 5 times smaller than at 40C.
Fig.6 depicts planar profiles of gas concentration, as result of the fast diffusion through the grain mass, in spite of the localized O_2 consumption and CO_2 production.



a b c

Figure 5: Profiles of temperature (a), moisture content (b) and respiration rate (c) along the middle vertical section of the silo-bag. Initial temperature 25C. Non uniform initial MC (16-13% w.b).

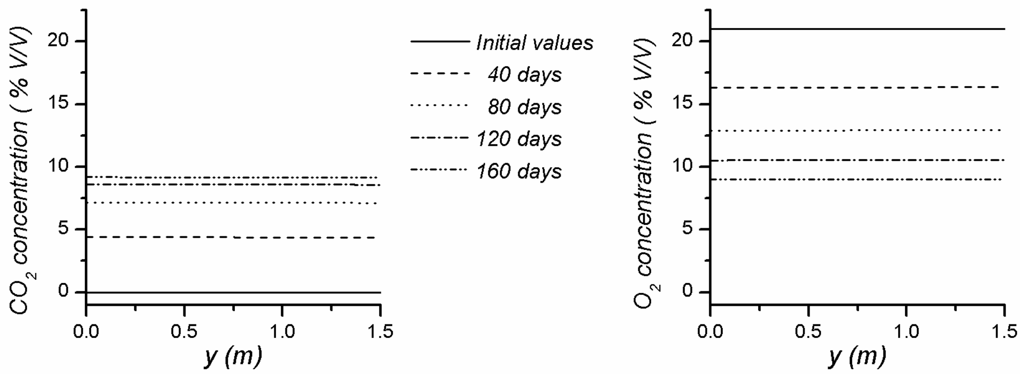


Figure 6: Profiles concentration of CO_2 and O_2 after 40, 80,120 and 160 days of storage. Initial temperature 25C. Non uniform initial MC (16-13% w.b).

Fig.7 compares the evolution of the mean gas concentrations for the base case (25C and 13%w.b MC) with the non uniform MC distribution case. The first curve is considered a referential value which represents adequate storage conditions. During the first month of storage, the difference between curves is below 1% w.b, but thereafter it increases up to 3 or 4 points. This suggests that if the silo-bag is monitored during the first month, the wet spot probably would not be detected because the measured and referential values are similar, even within experimental errors. But after two months, a CO_2 value measured 3 or 4 points above the referential value is a clear warning of possible grain spoilage in the silo-bag.

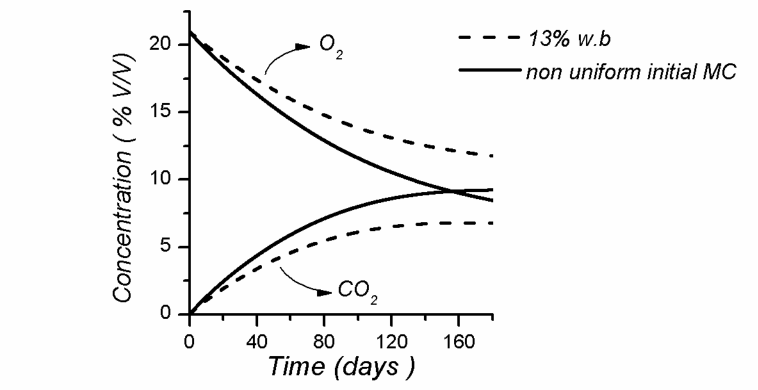
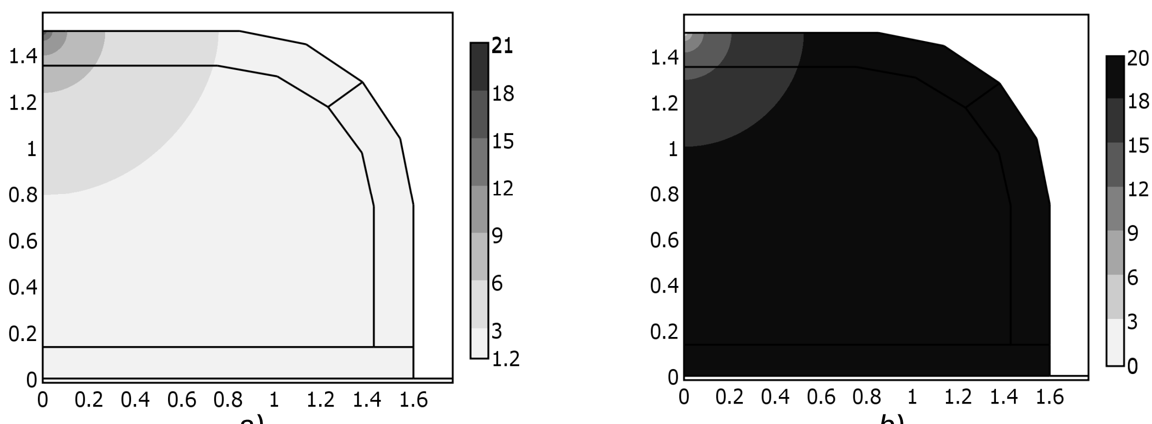


Figure 7: Profiles of O_2 and CO_2 concentration along the middle vertical section of the silo-bag

Effect of structural damage of the silobag

As mentioned previously, the silo bag may be damage in the field and a small perforation in the plastic layer modifies O_2 and CO_2 concentration significantly (Abalone et al., 2010b). The selected case for analysis is wheat bagged at 40C and 16%w.b MC, once anaerobic conditions (0 % of O_2 and 21% of CO_2) are attained (see Fig. 4b). A computational study was carried out to study the changes in gas concentration owing to the exchange of O_2 and CO_2 through a perforation of 1cm diameter located at the upper surface of the silo-bag silo.
Distributions of O_2 and CO_2 concentration after two and ten days of storage are presented in Fig. 8 and Fig. 9, respectively. Fig. 8a shows penetration radii of about 70 cm where O_2 is above 9% V/V. after two days. After 10 day, O_2 is above 9% everywhere. This condition increases the risk of grain spoilage because with such oxygen concentration biological activity may be reactivated.
Results obtained for wheat in this work are contrastingly different with those obtained for maize stored at similar conditions (35C and 16% w.b MC) by Bispo dos Santos et al. (2007), where O_2 penetration was very low. The values for maize respiration rate reported in that study were very high and are not consistent with those obtained by applying BERN et al.(2002) correlation cited by the authors.



a) b)

Figure 8: Distribution of O_2 (a) and CO_2 (b) concentration (%V/V) after two days of storage

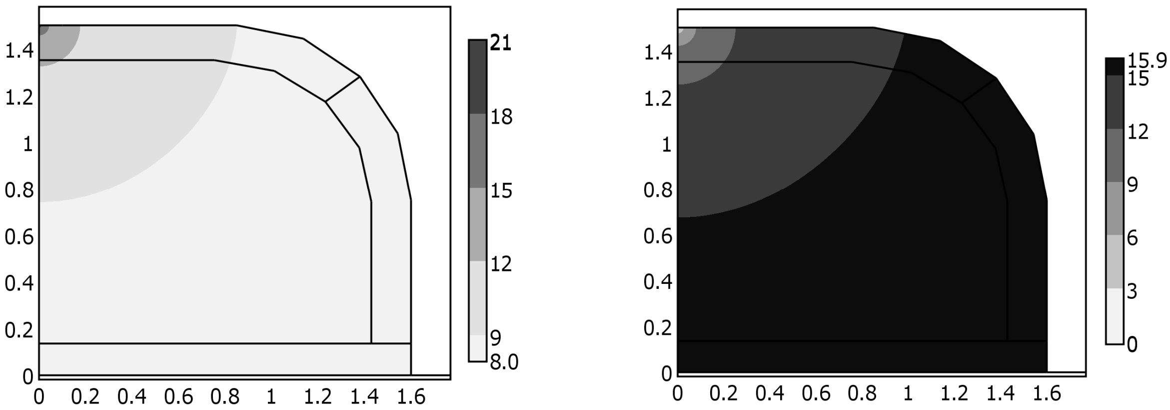


Figure 9: Distribution of O_2 (a) and CO_2 (b) concentration (%V/V) after ten days of storage

Conclusions

A diffusion model was developed to determine O_2 and CO_2 concentration in the interstitial atmosphere of silo-bags. Results were compared with those obtained by use of a lumped model to quantify the effect of averaging local variations of temperature and moisture migration when evaluating the rate of CO_2 production. The differences between predictions of mean gas concentrations were less than 1point %. It can be concluded that the lumped model is adequate for generating gas concentration curves for different grain storage conditions. This information can further be used as referential values for controlling adequate storage conditions by monitoring the CO_2 level in the silo-bags.
Local effects such as the presence of a wet stop at the bottom or a perforation at the surface of the silobag were analyzed by applying the diffusion model. A wet spot at 16% w.b affecting 10% of the grain initially loaded at 13% w.b increases 2 to 3 points CO_2 levels with respect to the referential curve. Penetration of O_2 through a 1cm diameter hole of is significant. A silobag at 40C and 16% w.b, initially in anaerobiosis, may attain O_2 levels of 10% V/V in ten days, with the potential risk of grain deterioration due to biological activity.
Further studies will be carried out, extending the diffusion model to 3D, in order to complete the analysis of local effects along the longitudinal direction on the silobag.

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