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MATHEMATICAL MODELS FOR CARBON DIOXIDE DISTRIBUTION AND LOSS IN BULKS OF GRAIN

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ABSTRACT

A mathematical model was developed for predicting the distribution of carbon dioxide (CO_2) through grain bulks. The model was solved using a finite element method and can be used for any shape or size grain bulk, provided that the bulk can be discretized into 8-node brick elements and input data can be prepared for the model. The model was developed in stages. Initially, the model was based on diffusion of CO_2 through bulk grain. In the current form, the model includes diffusion and movement of CO_2 within the grain bulk and sorption or desorption of CO_2 by the grain. The predictions of the model, in its various stages of development, were compared with experimental data on the distributions of CO_2 in three pilot-scale bins (about 1.4 m in diameter and 1.5 m high). We compared our experimental data with predictions based on two previously-published models of the loss of CO_2 from storage structures. Areas for further research in the development of mathematical models are also identified in this paper.

INTRODUCTION

A stored-grain bulk is a man-made ecological system in which deterioration of the stored-product results from interactions among physical, chemical and biological factors. Heat, moisture and carbon dioxide (CO_2) are produced during the deterioration of grain; therefore, increases in grain temperature, moisture content and CO_2 concentration in the intergranular air can all be used as indicators of incipient grain spoilage. An increase in the CO_2 concentration is a better indicator of incipient spoilage than the increase in temperature or moisture of grain because the normal concentration of CO_2 in the atmospheric air is low (0.03%) and therefore any increase can be easily detected. In addition, the movement of CO_2 through bulk grain is faster than is the movement of heat or moisture.

Modified atmospheres (MA's) provide an environment which is lethal to stored-grain pests. This is achieved by altering the proportions of CO_2, nitrogen (N_2) and oxygen (O_2) in the intergranular atmosphere. MA's containing elevated CO_2 (>60%), depleted O_2

(<10%) and low relative humidities (<50%) at high temperatures (>27°C) are, within a few days of exposure, lethal to most of the insect and mite species that are responsible for losses in stored grain (White *et al.*, 1990; Jayas *et al.*, 1991; White and Jayas, 1991; Rameshbabu *et al.*, 1991). Under a given set of environmental conditions, exposure times and gas concentrations are the critical factors, and the required minimum of these factors to cause death varies with pest species. To create MA's, CO_2 or N_2 is usually introduced into the grain mass at single or multiple points. The uniform distribution of introduced gases and the maintenance of the lethal concentration for the required duration are critical for effective control of pest populations.

The movement of CO_2 through bulk grain is dependent on the characteristics of the grain, the storage structure and the weather. Mathematical models can be developed to predict CO_2 concentrations at various locations in the stored grain when CO_2 is produced during grain spoilage or when CO_2 is introduced for disinfestation. The predictions of such models can be used either to determine the resolution required and the location of CO_2 sensors for the detection of incipient spoilage in stored grain, or to design systems that will provide a uniform distribution of CO_2 in the bulk grain. Mathematical models of the loss of CO_2 from non-airtight structures can be used to determine the amount, frequency and location of the gas injections needed to maintain lethal atmospheres.

Mathematical models, if based on the principles of physical and biological sciences and properly validated, can be used to study the effects of various parameters, such as weather, grain condition, and the size, shape and material of the storage structure, on the distribution and loss of CO_2. The major advantages of validated models are their ability to answer "what if?" questions and their transferability to different climatic regions of the world, thus making them globally applicable rather than only locally applicable (as is expected of experimental studies). Only a few studies on the movement of CO_2 within the grain bulk and the loss of CO_2 from the stored grain have been reported in the literature (Singh *et al.*, 1983; Banks and Annis, 1984; Navarro *et al.*, 1985; Jayas *et al.*, 1988; Alagusundaram *et al.*, 1991; Navarro *et al.*, 1991). Our research group has been working on the development and validation of a mathematical model of the distribution of CO_2 and on the evaluation of reported models for their ability to predict the loss of CO_2 from the storage structures. Progress on our model and the results of our evaluation of two CO_2-loss models are summarized in this paper.

MATHEMATICAL MODEL FOR CO_2 DISTRIBUTION

Earlier models

Singh *et al.* (1983) solved an axisymmetric diffusion equation using the finite element method and then predicted CO_2 concentrations in grain masses containing localized spoiling grain. The model predictions were not compared with experimental data. Based on simulations, we determined that the best location for installation of a CO_2 sensor for detection of incipient spoilage was at the centre of the grain mass. This was later confirmed experimentally by Muir *et al.* (1985). Later Jayas *et al.* (1988) used the model of

Singh *et al.* (1983) for predicting the CO_2 concentrations in a 5.8-m-diameter bin containing wheat 4.9 m deep. The predicted concentrations at various locations were compared with values measured 23 h after injecting CO_2 both at the centre and 1 m below the grain surface (all CO_2 concentrations were measured by gas chromatography). In the bottom two-thirds of the bin, the predicted CO_2 concentrations (assuming the bin wall to be impermeable to CO_2) were higher than the measured concentrations. We concluded that the model prediction could be improved by allowing some leakage through the bin wall.

In a stored-grain bulk, CO_2 may be introduced at points near the wall and then sorbed (absorbed and adsorbed) by grain, or it can be produced at various locations by pockets of spoilage containing respiring grain, microflora, mites and insects, thus making the problem of CO_2 movement three-dimensional. Alagusundaram *et al.* (1991) solved a three-dimensional diffusion equation using the finite element method for predicting CO_2 distribution in bulk-stored wheat. The predicted CO_2 concentrations were much lower than the measured concentrations in three wheat-filled, 1.42-m-diameter bins (refer to the section on experimental data for model validation). In this model, the bulk movement of CO_2 caused by the expansion of the CO_2 during sublimation was not considered. Later, Alagusundaram *et al.* (1996a) used an experimentally-determined apparent flow coefficient (D_{app}) during the application period to model the bulk movement of CO_2. Although the model predictions were improved using this approach, the errors continued to be high during the initial 3-h period after the introduction of dry ice. To use the approach of Alagusundaram *et al.* (1996a), in addition to the diffusion coefficient the values of D_{app} must be determined for every grain. Such an empirical approach is not an efficient method for use in a mathematical model.

Present model

The transient transport of miscible fluids in an anisotropic porous medium is governed by the following partial differential equation (Bachmat and Bear, 1964; Huyakorn *et al.*, 1986; Bundus *et al.*, 1996):

$$\frac{\partial C}{\partial t} = \frac{\partial}{\partial x_i}\left(D_i \frac{\partial C}{\partial x_i} - V_i C \right) + q \qquad (1)$$

subject to the boundary conditions:

$$C = C_{S1} \qquad \text{on boundary S1} \qquad (2)$$

$$D_i \frac{\partial C}{\partial x_i} l_i - V_i C l_i = Q \qquad \text{on boundary S2} \qquad (3)$$

and the initial condition:

$$C\,(i, t = 0) = C_0 \qquad \text{in the domain } \Omega \qquad (4)$$

where C = concentration of CO_2 at time $t > 0$ (kg/m^3), C_0 = initial concentration of CO_2 in the domain Ω (kg/m^3), D_i = diffusion coefficient of CO_2 through bulk grain in the ith direction (m^2/s), l_i = direction cosines of outward drawn normal to the boundary, q = amount of CO_2 sorbed or produced by the grain (kg m^{-3} s^{-1}), Q = total surface flux across the boundary S2 (kg m^{-2} s^{-1}), S1 and S2 = boundary segments (defined in the following paragraph), t = time (s), V_i = velocity of CO_2 flow in the ith direction (m/s), and Ω = domain consisting of the stored-grain bulk; subscript $i = x$, y and z in a Cartesian coordinate system.

The boundary S1 represents the segment of the boundary where concentration of CO_2 can be specified. It may consist of more than one part of the boundary. For example, part of S1 may represent the surface of the grain where concentration may be specified as constant at the atmospheric level in a ventilated head space; another part of S1 may represent the portion of a grain boundary where CO_2 is injected and thus maintained at a constant concentration. Similarly, S2 may be made up of more than one part of the boundary. For example, the bin floor may be assumed to be impermeable to the flow of CO_2, whereas bin walls may have a specified flux of CO_2 to the surroundings depending on the rate of loss through the bin wall. Segments S1 and S2 together make the total boundary of the domain Ω. The specified CO_2 concentration along boundary S1 is known as the Dirichlet boundary condition, and the specified flux across boundary S2 is known as the Neumann boundary condition. When the surface flux, Q, is zero, Eq. (3) represents a homogenous Neumann boundary condition.

To handle the period of CO_2 sublimation from dry ice, we calculated and used effective diffusivities that take into account the effects of both bulk movement and diffusion. Replacing diffusion coefficients with effective diffusivities during the CO_2-sublimation period eliminates the velocity terms in Eq. (1), thus eliminating the computational difficulties engendered by the presence of the velocity term. The effective diffusivities were calculated using the equations of Saffman (1960) and Bear (1972):

$$D_{\mathrm{L}} = \frac{1}{2} V_{\mathrm{a}} L \left(\frac{\ln 3 V_{\mathrm{a}} \tau_0}{L} - \frac{1}{12} \right) \tag{5}$$

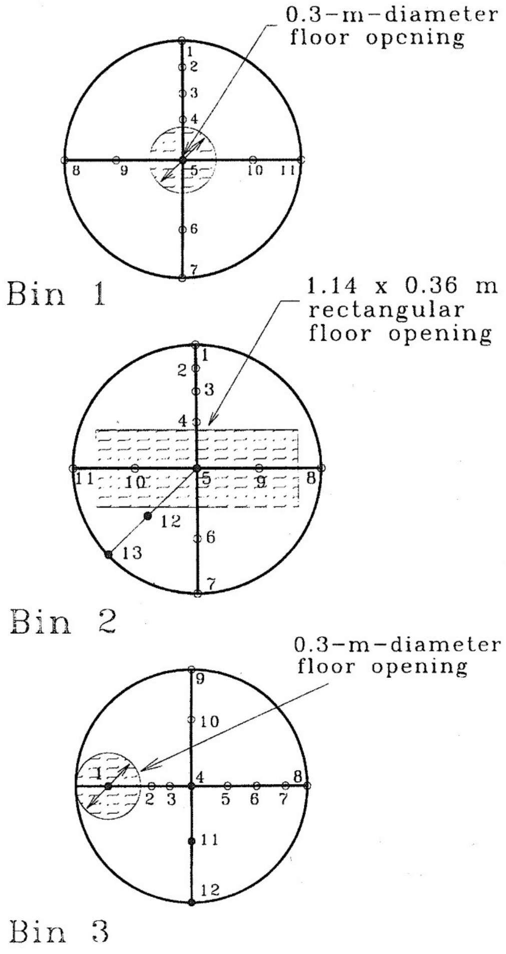
$$D_{\mathrm{T}} = \frac{3}{16} V_{\mathrm{a}} L \tag{6}$$

where D_{L} = longitudinal diffusivity (diffusivity in the major direction of flow) (m^2/s), D_{T} = lateral diffusivity (diffusivity perpendicular to the major direction of flow) (m^2/s), L = grain size (m), V_{a} = average Darcy velocity (m/s) and $\tau_0 = L^2/2D_i$ (1/s).

The diffusion coefficient of 6.46 mm^2/s (Shunmugam *et al.*, 1994) was used after the sublimation of CO_2. It was assumed that the diffusion coefficient was independent of the direction of diffusion (Singh *et al.*, 1984) and of CO_2 concentration (Cunningham and Williams, 1980).

Experimental data for model validation

The measured CO_2 concentration data of Alagusundaram *et al.* (1996b) were used to validate the model. The CO_2 concentrations were measured at various locations in wheat bulks contained in three bins 1.42-m diameter by 1.47-m height (Fig. 1). Each of the three bins was equipped with a different partially-perforated floor opening (circular at the centre, rectangular and circular near the wall). The CO_2 data were collected from five levels in each bin (near the floor and then spaced 0.33 m apart vertically). At each level there were 11, 13 and 12 sampling points for Bins 1, 2 and 3, respectively. Metal boxes (0.5 × 0.5 × 0.37 m for Bins 1 and 3 and 1.22 × 0.46 × 0.36 m for Bin 2) were mounted centrally under the floor openings. Known quantities of dry ice were introduced in these boxes to create high CO_2 concentrations in the wheat bulk. The CO_2 concentrations were recorded at 1, 3, 6, 9, 12 and 21 h after the introduction of dry ice. The tops of the grain bulks were either left open or covered with a polyvinylidene chloride (PVDC) sheet impermeable to CO_2.



Bin 1

Bin 2

Bin 3

Fig. 1 Schematic diagram of the temperature and gas sampling locations in 1.42-m-diameter by 1.47-m-tall bins. ● = temperature and gas samples; ○ = only gas samples.

Validation of the present model

The CO_2 concentrations predicted by the convective-diffusive model were compared with the measured CO_2 concentrations by calculating the mean relative-percent-error (hereafter referred to as error):

$$e = \frac{1}{N}\sum_{i=1}^{N}\frac{|M_i - P_i|}{M_i} \times 100 \tag{7}$$

where e = mean relative percent error (%), M_i = measured CO_2 concentration at sampling location i (%), N = number of data points and P_i = predicted CO_2 concentration at sampling location i (%).

The errors in all three bins (Fig. 1) at different sampling times are given in Table 1. These data can be used to judge the accuracy of the model prediction. The accuracy of model predictions improved considerably in comparison with both the predictions of a pure diffusion model (Alagusundaram *et al.*, 1991) and the predictions using experimentally determined apparent flow coefficients during the dry ice sublimation period (Alagusundaram *et al.*, 1996a). However, the errors were still high in the initial time periods. There are two probable reasons for such high errors in the initial time periods:

1. The horizontal velocity, V_x, was estimated using the airflow-pressure drop relationship of wheat. The flow characteristics of CO_2 through wheat could be different from that of the flow characteristics of air. Data on the low-velocity flow characteristics of CO_2 through agricultural grains are essential for accurately predicting the CO_2 distribution in MA storage.
2. The gravity effect was not included in the model. CO_2 is about 1.5 times heavier than air (CO_2 density at a temperature of 20°C is 1.82 kg/m^3 compared with air density of 1.19 kg/m^3 at the same temperature). The gravitational forces acting on the heavier CO_2 molecules will reduce their rate of vertical movement. This hypothesis was supported by the fact that the differences between the measured and the predicted CO_2 concentrations at 0.33 and 0.66 m above the floor were larger than the differences near the floor. For example, in Bin 1 with 180 g dry ice and a covered grain surface, the average errors at 3 h were 11.7% near the floor and 18.7 and 37.7% at 0.33 and 0.66 m, respectively, above the floor. Thus, it is essential to include the gravity effect in the model.

The predictions agreed well with the CO_2 concentrations at 12 h. In Bin 2 with 370 g dry ice and a covered grain surface, for example, the error at 12 h was only 6%. The errors, however, were higher at 21 and 24 h, when the predicted concentrations were higher than the measured concentrations. This could be due to sorption of CO_2 by the wheat. To determine the effect of sorption, we compared predicted concentrations, with and without sorption, for the 740-g dry-ice experiment in Bin 2 with a covered grain surface. The sorption of CO_2 by wheat was estimated using the data of Cofie-Agblor *et al.* (1993). The error at 21 h was 22.4% with sorption, compared with 34.9% without sorption. Therefore, including sorption appeared to improve the accuracy of the model.

We did not simulate concentrations with sorption for other experiments because

TABLE 1

Mean relative errors[1] (%) between the measured CO_2 concentrations in 1.42-m-diameter bins filled with wheat and the concentrations predicted by the convective-diffusive model. Effective diffusivities were used in the first hour and the diffusion coefficient was used thereafter

Bin	Grain surface	Mass of dry ice (g)	Replicate	Time since introduction of dry ice												
				1 h	3 h	6 h		9 h		12 h		21 h		24 h		
						5 L*	4 L**	5 L	4 L	5 L	4 L	5 L	4 L	5 L	4 L
1	Open	180	1	34.8	21.4	16.0		10.9	6.4	6.0	4.5	21.9	10.6		
			2	31.7	24.9	19.8		11.2	7.8	5.9	4.7	16.0	13.8		
			3	33.9	24.7	17.6		7.1	6.0	6.0	3.5	26.0	17.2		
			4[2]	33.7	23.6	18.0		9.5	6.4	4.5	3.5	20.0	16.3		
	Covered		1	17.9	22.2	18.1				12.1	10.3	15.6	15.9		
			2	29.6	24.0	14.9				9.5	7.4	11.1	12.4		
			3	28.7	22.1	17.8				11.5	8.5	10.5	10.5		
			4[2]	25.4	22.7	17.0				10.8	8.5	12.3	12.9		
		540	1	60.2	50.5	36.5	34.5	20.0	18.8	27.3	13.8	21.5	4.2		
			2	61.5	52.5	36.0	37.0	31.8	31.0	33.2	25.3	30.2	4.4		
			3	59.7	55.9	41.3	38.2	26.9	29.2	16.1	16.3	9.0	6.6		
			4[2]	60.5	53.0	38.4	36.7	24.4	26.8	20.6	18.6	14.3	4.5		
2	Open	370	1	47.2	32.2	20.6				11.7	7.6	21.5	20.2		
			2	46.1	28.2	17.8				10.9	10.0	26.0	21.4		
			3	42.8	31.9	22.4				9.5	4.9	13.6	15.3		
			4[2]	45.4	30.3	20.2				8.0	5.6	17.9	18.1		

TABLE 1 (continued)

Bin	Grain surface	Mass of dry ice (g)	Replicate	Time since introduction of dry ice											
				1 h	3 h	6 h		9 h		12 h		21 h		24 h	
						5 L*	4 L**	5 L	4 L	5 L	4 L	5 L	4 L	5 L	4 L
	Covered		1	47.0	31.7	22.1				5.9	5.2	15.2	13.2		
			2	45.5	31.7	21.0				8.3	7.6	15.3	13.4		
			3	46.0	31.4	19.4				7.5	7.9	18.1	15.5		
			4[2]	46.2	31.6	20.8				6.3	6.2	15.8	13.6		
		740	1	47.8	39.3	28.6				7.11	6.3	31.7	23.5	35.2	25.7
			2	46.2	36.1	22.2				8.8	7.5	37.6	28.8	43.4	31.2
			3	46.4	40.3	25.9				9.2	7.4	39.0	29.6	44.1	31.9
			4[2]	46.5	38.0	26.2				6.8	5.6	34.9	26.4	39.8	29.1
3	Open	180	1	42.0	50.7	44.5		39.1	35.5	30.8	28.9	15.9	13.3		
			2	46.0	52.5	45.6		37.8	34.3	29.7	27.7	15.3	12.3		
			3	46.5	57.1	48.5		38.8	40.1	32.1	33.9	17.6	18.7		
			4[2]	43.9	53.7	46.4		39.1	36.8	30.9	30.4	16.0	14.8		
	Covered		1	48.1	53.6	49.5		43.3	40.6	34.7	33.1	17.5	17.2		
			2	49.1	55.3	49.4		42.6	39.0	34.9	33.8	20.1	20.0		
			3	43.2	55.5	46.1		43.5	39.8	34.3	31.9	19.1	18.8		
			4[2]	43.9	54.1	48.5		42.9	39.4	35.6	32.8	18.3	18.4		

[1] Mean relative error (%) = $\frac{1}{N}\sum\frac{|\text{Measured}-\text{Predicted}|}{\text{Measured}} \times 100$. The CO_2 concentrations in the bins ranged from 5 to 60%.

[2] Measured data for the three replicates were averaged and compared with the simulation results.

N = number of data points (25 in Bin 1, 50 in Bin 2 and 45 in Bin 3).

*Mean relative percent errors were calculated for all 5 levels (5 L).

**Mean relative percent errors were calculated for the bottom 4 levels (4 L).

the available sorption data are only available for a 100% initial CO_2 concentration (Cofie-Agblor *et al.*, 1993). It is possible that at lower initial concentrations the sorption rate could be equal to the sorption rate at 100% initial concentration. However, there is no experimental evidence to prove this hypothesis, and Alagusundaram *et al.* (1996a) demonstrated, by predicting CO_2 concentrations with various rates of sorption, that the model predictions are sensitive to the sorption rate. Therefore, to obtain more accurate model predictions, we are currently determining the CO_2 sorption behaviour of wheat at various initial concentrations.

EVALUATION OF CO_2-LOSS MODELS

Mathematical models of CO_2 loss from bins (caused by wind, temperature and chimney effects) can be used to predict the amount of CO_2 needed to maintain the required concentration and thus improve the efficiency and effectiveness of MA fumigation of stored grain. Predictions of two mathematical models from the literature, those of Lawrence Berkeley Laboratory (LBL) (Sherman and Grimsrud, 1980) and of Banks and Annis (BA) (1984), were compared with experimental data on gas loss from a pilot bin (diameter 1.42 m, height 1.47 m) and a full-size bin (diameter 5.56 m, height 6.60 m), both filled with wheat (Peck *et al.*, 1994).

The effective leakage areas (ELA) of the bins were determined using fan pressurization tests. An ELA of 4.6 cm^2 was measured for the pilot bin. In the full-size bin, a CO_2 impermeable plastic sheet was attached to the inside wall at 2.5 m above the floor. This excluded the upper half of the bin, including the roof, and resulted in an ELA of approximately 7.7 cm^2.

A precise comparison of the predicted and measured rates of CO_2 loss from the two experimental bins could not be done because the effects of the three major factors (wind, temperature changes and chimney forces) could not be separated in the experimental results. In both models, however, the simulated effects of these three factors can be compared separately.

The LBL model predicted a loss due to wind that was 60 times higher for the pilot bin and 5 times higher for the full size bin than the measured total rate of CO_2 loss. The predicted CO_2 loss from the full size bin due to the temperature effect was equal to the measured total rate of loss. In the LBL model, the shielding and the terrain coefficients do not account for the direction of the prevailing wind and depend on a subjective assessment of the surroundings. In addition, an overestimation of the ELA by the pressurization test would cause significant differences between predicted and experimental data.

Predicting the rate of gas loss caused by wind with the BA model was difficult because the model was sensitive to an undefined pressure coefficient. Determination of this coefficient requires wind tunnel tests for each bin. These wind tunnel tests were not conducted and are not practical for a general model. The predicted chimney effect using the pilot bin was 75 times the average experimental gas loss rate. An accurate comparison, however, is not possible without more experimental data. For the full size bin, the predicted gas loss rate due to the chimney effect was approximately 15 times the experimental gas loss rate. Further experimental studies are being conducted in our laboratory to separate the forces of wind, temperature and chimney effects so that accurate relationships can be developed. This should provide data which could be used to validate the existing models more accurately and provide information for the development of a generalized CO_2 loss model.

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