OZONE: SCIENCE & ENGINEERING
https://doi.org/10.1080/01919512.2017.1404902

Numerical Simulation and Validation of Mass Transfer Process of Ozone Gas in Rice Grain Bulks

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ABSTRACT

Ozone is a potential alternative to commercial fumigants. The development of ozone fumigation technology for insects control requires a precise prediction of the distribution of introduced ozone gas in grain bulks. Hence, this study was conducted to investigate the mass transfer process of ozone gas in rice grain bulks. Experiments were conducted in a 0.6-m diameter flat-bottom bin filled with rice grains and ozonated using 5 g h^{-1} capacity ozone generator. Simulation of transfer process of ozone through the rice grain bulks was conducted using the principle of the law of conservation employing the continuity equation. The experimental data for ozone transfer through connected the column of rice was used to validate the model. The relative error between the actual and predicted ozone concentration for the entire bin geometry was less than 33%. The predictions from the projected model were in accordance with the ozone concentrations obtained from the experimental results.

ARTICLE HISTORY
Received 12 July 2017
Accepted 10 November 2017

KEYWORDS
Ozone; Continuity Equation; Fumigations; Half-Life; Mass Transfer; Rice; Saturation Time

Introduction

Ozone is a viable alternative fumigant to manage phosphine-resistant insect pests and fungi in the stored grains. It is an environmentally safe alternative in the food industry because ozone decomposes into oxygen within hours under the regular conditions (Kells et al. 2001; McClurkin, Maier, and Ileleji 2013); hence it is a residue-free technology (Pandiselvam et al. 2017a; Zotti et al. 2008). Due to its rapid decomposition, ozone could be generated onsite to ensure its continuous supply (Pandiselvam and Thirupathi 2015a) and eliminates the handling and storage of chemicals (Pandiselvam et al. 2017c). Moreover, ozone is Generally Recognized as Safe (GRAS) by the U.S. FDA, and it is approved for use as an antimicrobial agent on processed food (FDA 2001).

Ozone is a highly reactive gas and the decay rate of ozone gas is highly affected by ambient temperature and relative humidity (McClurkin, Maier, and Ileleji 2013). In addition to temperature, the bed thickness and moisture content of grain (Pandiselvam, Thirupathi, and Anandakumar 2015b; Ravi, Venkatachalam, and Rajamani 2015), selection of ozone concentration and exposure time (McDonough, Mason, and Woloshuk 2011) and grain surface characteristic (Steponaviciene et al. 2012) have an impact on the efficacy of ozone against stored product insect pests.

Understanding the mass transfer behavior of ozone is a fundamental requsite for its effective utilization as a fumigant at the industrial level (Pandiselvam et al. 2017b; Pandiselvam et al., 2017c). Assessing the concentration profile and flow characteristic of ozone gas through grain bulks is important to determine the required concentration and time of exposure to achieve efficient and effective ozone fumigation process (Pandiselvam et al. 2017c). Based on the knowledge of the ozone gas flow characteristic in a grain bulks, it is plausible to evaluate the technical feasibility of the fumigation system and to calculate the dimensions of storage bins for efficient fumigation process (Paes et al. 2017). Hence, to assess the adaptability of ozone fumigation system in the storage bins, a study on the concentration profile and flow characteristics of ozone is imperative. Therefore, this research aims to investigate the numerical simulation of ozone concentration and flow characteristics in rice bulks as a function of time.

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Materials and methods

Materials

Rice (ADT 43) grains were procured from the local mill in Thanjavur of Tamil Nadu, India. The moisture content of the rice was determined by an oven method (ASAE 1998).

Ozone generation

The ozone was obtained through an ISM 5 ozone generator developed by Creative Oz-Air Pvt. Ltd. Noida, India (http://www.oz-air.com). The ozone gas generation process used oxygen at a purity level of greater than 95%, and was obtained by using an OZ-AIR HG5 Oxygen Concentrator (Creative Oz-Air Pvt. Ltd, Noida, India).

Storage bin

The concentration profile and flow characteristic of ozone gas was observed in a cylindrical storage bin (1.5 m height and 0.6 m diameter), which is made up of food grade stainless steel (SS 304). The storage bin was designed in such a way that the rice grains can be loaded at the top and unloading can be done at the bottom. The bin consists of the perforated sheet at the bottom, which helps to distribute the ozone gas uniformly throughout the bin. Ozone gas was injected at the bottom of the storage bin. The inlet ozone concentration and flow rate was fixed at 500 ± 25 ppm and 2 LPM, respectively. Experiments were conducted at a temperature of 30 ± 4 ° C and 60% RH. To measure the concentration of ozone in the storage bin, five concentration ports were placed. The ozone concentrations present in rice column were measured at every 5-min interval using a continuous ozone monitor (BMT MESSTECHNIK GMBH-964, Gueterfelder Damm, Stahnsdorf, Germany). The leftover ozone gasses passes through the thermal ozone destructor developed by Creative Oz-Air Pvt. Ltd (Material: stainless steel, Size: 150D×500L (mm), Power consumption: 800 Watts, Power supply: 230V/AC, Insulating material: Glass wool, Temperature: 200 °C) to convert the ozone into oxygen.

Numerical modeling

A one-dimensional numerical model by applying the principle of the law of conservation along with continuity equation used to assess the concentration profile of ozone in the stored rice bulks. Cylindrical coordinates were selected for the equation development. The first law of thermodynamics is used for developing the conservation equation for the fluid flow in the packed element at any time (Geankoplis 2003). Assumptions made for the numerical simulation of an ozone concentration profile are same as Pandiselvam et al. (2017a) and Pandiselvam et al. (2017c) for paddy and green gram, respectively.

Assumptions made for the numerical simulation are a) stored rice grains are considered as uniform in size, shape, and continuous mass at same moisture content and variety; b) porosity of the grains are uniform throughout the bed; c) frictional forces of grains are negligible during fumigation; d) grains have the same physical, mechanical and aero dynamical properties in any direction of bed; e) grains are linearly distributed in ozone entry zone; and, f) mass transfer of ozone takes place in the vertical (Z-axis) axis of the storage bin in unsteady steady state condition.

The mass transfer of ozone gas into intergranular air in the rice grain column describing by partial differential equation is given as

$$\frac{\partial c}{\partial t} = D_e\left(\frac{\partial^2 c}{\partial z^2}\right) - v_z\left(\frac{\partial c}{\partial z}\right) \tag{1}$$

The algebraic form of the equation is

$$\begin{aligned} c_{i,j+1} - c_{i,j} = A\big(&c_{i-1,j+1} - 2c_{i,j+1} + c_{i+1,j} \\ &+1 + c_{i-1,j} - 2c_{i,j} + c_{i+1,j}\big) \\ &-B\big(c_{i,j+1} - c_{i,j}\big) \end{aligned} \tag{2}$$

in which,

$$A = \frac{\partial\ tD_e}{2\ \Delta\ \partial z^2}$$

and

$$B = \frac{\partial t}{\partial z}$$

where, c is Ozone concentration, ppm; t is time, s; D_e is diffusivity, $m^2\ s^{-1}$; and V_z is the velocity of ozone in 'Z' direction.

Solution for numerical model

Experiments were conducted to measure the ozone concentration profile and flow characteristics of ozone in the stored rice bulks at 34 ± 2 °C and 56% relative humidity. A MATLAB (MATLAB 7.5; Math-Works, Inc., Natick, MA, USA) code was written to solve the algebraic equation [2]. The initial ozone concentrations in the bottom of the chamber were used as inputs for predictions. The time interval for prediction of ozone concentration fixed at 5 min for each time step in the simulation study. The governing partial differential equation and the associated boundary conditions, on which the model was based, assume that the mechanism of convective mass transfer. Predicted values from the simulation and the experimental results were compared.

Results and discussion

Flat bottom bin leveled with rice grain surfaces fumigated with ozone using fully perforated floor provide the nonlinear ozone distribution through the grain bulks shown in Figure 1. Although ozone application in rice grain bulks is performed through continuous flow, it is difficult to maintain stable concentrations and distribution in it. The diffusion process describing the distribution of ozone particles and self-dispersion in the grain layer depends on the air filtration velocity (Mendez et al. 2003; Raila et al. 2006), grain bed thickness (Pandiselvam and Thirupathi 2015a; Pandiselvam, Thirupathi, and Anandakumar 2015b), presence of impurities in grains (Hardin et al. 2010) and temperature (Allen, Wu, and Doan 2003; McClurkin, Maier, and Ileleji 2013) through the grain column. However, air velocities higher than 0.03 m/s cannot be recommended in the commercial storage structures with grain depths of 35–40 m due to excessive energy requirements needed to overcome the static counterpressure (Mendez et al. 2003), thus increasing the cost of ozone fumigation (Mohapatra et al. 2017).

This study has shown that ozone gas highly diffuses in the lateral direction as compared to a vertical direction (Figure 1). The diffusivity of ozone gas in rice grains bulks 0.0133 $m^2\ s^{-1}$. When the bin is filled with rice grains the positioning of slender shaped rice kernels that lie with the major axes horizontal, could provide more resistance to ozone flow in the vertical direction (Jayas, Sokhansanj, and Moysey 1987). Also, the density of ozone (2.14 kg m^{-3}) is greater than the density of air (1.43 kg m^{-3}) provides more resistance to vertical flow. In the systems, where ozone movement is not complicated by grain surface adsorption or temperature, the rate of diffusion is inversely proportional to the square root of the density of the ozone. The density difference may result in mass flow in addition to molecular diffusion; hence the rate of movement of ozone in the direction of the force of gravity (downward) would be greater than upward movement of ozone, against the force of gravity.

Grain bed thickness is one of the key parameters to consider in characterizing the mass transfer of gaseous ozone in an unsaturated porous media (Pandiselvam et al. 2017c). The predicted concentrations of ozone at different bed thickness are shown in Figure 2. A significant decrease in the ozone concentration from the plenum to the surface of the rice grains was observed. Ozone concentration value shows a significant decrease from 504 ppm (plenum) to around 135 ppm at 0.5 m distance from the perforated floor. This is due to concentration gradient in the rice grain column, which is nonlinear. These differences in ozone concentration across the grain bed are narrowed down with a decrease in grain bed thickness.

The moisture content of the rice grain was 11.55 ± 0.14 (%) (d.b.). Two important factors affect the ozone gas flow rate through the rice grains were bulk density and porosity of the sample; known to be 712 kg.m^{-3} and 27%, respectively, at 11.55% (d.b.) moisture content (Pandiselvam, Thirupathi, and Mohan 2015c; Ravi and Venkatachalam 2014). Mendez et al. (2003) observed that ozone penetrates to a greater depth in more porous grains like in maize in less time as compared with less porous grains like wheat. The development of the non-parallel ozone flow streamlines around the ozone injection point could be attributed to the effects of porosity and density. The

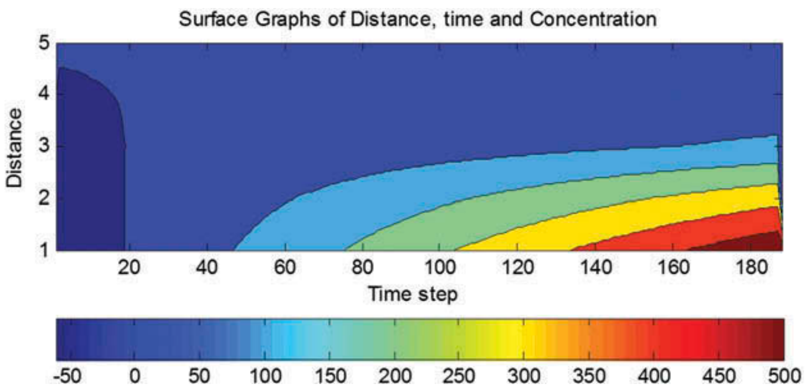

Figure 1. Simulated ozone concentration as a function of time in rice bulks.

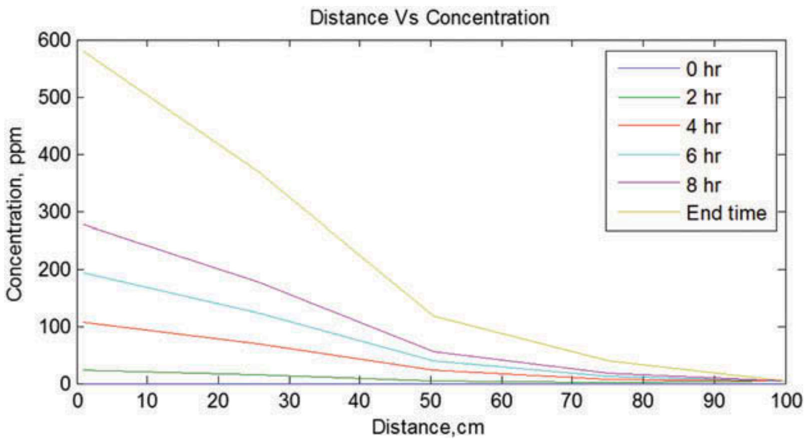

Figure 2. Residual concentration of ozone as a function of bed distance at different fumigation time.

effective flow of ozone comes to an end at an approximate height of 0.5 m from the ozone injection point. This is characterized by the formation of longer ozone flow paths towards the lateral direction and shorter ones at the upper most region of the bin.

The ozone concentration along the grain bed thickness is mostly affected by fumigation time. Figure 3 shows that ozone concentration increases with increase in fumigation time. The time required to reach desired concentration is based on flow rate, initial concentration, and ozone delivery pattern. The lines of variable traverse time give a quick method of identifying regions where the ozone concentration will be low. Fully perforated storage bin floors would create parallel ozone flow streamline and lower the risk of nonozonation zones in the grain at the bottom of the bin. However, ozone is a surface active material, fumigation of grains using ozone gas, initially reacts with grain surface because of the organic nature (active tissue) of the stored material, the high surface contact area of the grains (Hansen, Hansen, and Jensen 2013) and diffuse the internal layer (Churmasov, Rezchikov, and Gavrilova 2002). Therefore, it is difficult to maintain steady state ozone concentrations in the interstices of the material to ensure uniform treatment to all the material in a storage container.

The grains considered to be in phase one state at the start of the fumigation progress, ozone gas consumed soon after it entered the stainless steel bin, similar to wheat (Hardin et al. 2010), green gram (Pandiselvam and Thirupathi 2015a), paddy (Pandiselvam, Thirupathi, and Anandakumar 2015b) and peanut (Roberto et al. 2016). As the ozone fumigation progresses, the rate of consumption gradually decreases resulting in ozone diffusion into the grain mass (Hardin et al. 2010).

It indicates the free flow of ozone gas through the grain column with little degradation once the active tissues responsible for ozone decomposition are saturated (Mendez et al. 2003). However, the higher rice grain bed thickness configuration in the present case has resulted in the nonflow paths at grain surface. Such

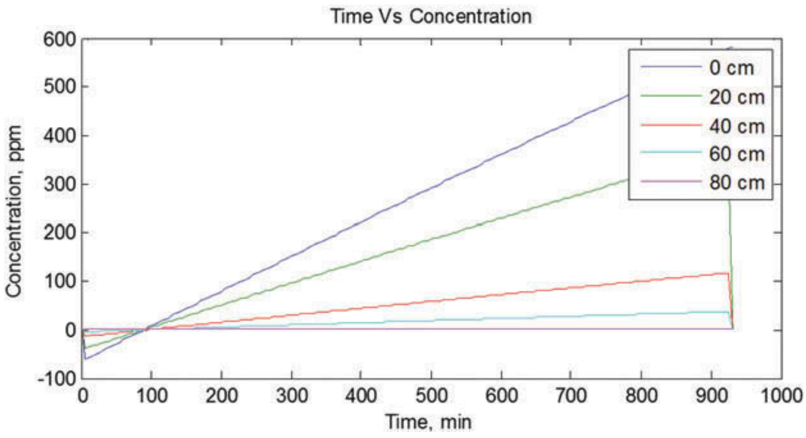

Figure 3. Residual concentration of ozone as a function of time.

nonuniformity in ozone flow pattern within the grain bed can lower the efficiency of the fumigation process causing discoloration in the food materials as it tends to oxidize the surface color (Qi et al. 2016; Tiwari et al. 2010). Multiduct designs may facilitate in providing uniform ozone flow distribution across the storage bin (Pandiselvam et al. 2017c).

Validation against experimental data is necessary for developing accurate computer simulations. The simulations run with the assigned boundary conditions and the initial concentration at every node in the grid set zero concentration. From the Figure 4, it was observed that the regression line follows the diagonal pattern. It shows that the predicted concentration has higher accuracy. The relative error of prediction and chi-square was calculated as

$$\text{Relative error}(\%) = \frac{(O_{\text{experimental}} - O_{\text{predicted}})}{O_{\text{predicted}}} \times 100 \quad (3)$$

$$\text{Chi-square}(\chi^2) = \frac{(O_{\text{experimental}} - O_{\text{predicted}})^2}{O_{\text{predicted}}} \times 100 \quad (4)$$

Experimental and predicted ozone concentration values were compared and presented in Table 1.

Due to the potentially large errors in the measurement of little amounts of ozone during the early part of the experiment, relative error values were high for the second, third and fourth hour of fumigation. For instance, at the ozone concentrations of around 16 ppm measured using ozone gas analyzer, error of ±8 ppm could have occurred even after careful calibration, resulting in the relative error of 33%. Across the entire grain bed, the mean relative error value between the experimental and predicted ozone concentration values was 12.33%. Alagusundaram et al. (1996) reported that the mean relative error of prediction was 55% for the diffusion model. Ozone concentration values were comparatively over predicted an up to 6 h and under predicted from 6 to 13 h. But the predicted ozone concentration values are closer to experimental values from 6 to 13 h fumigation. The accuracy of the simulation model is higher at 7 h prediction. However, at all points, the relative error values were within the acceptable range. The chi-square value (*df* = 12) varied from 0 (1 h) to 10.90 (4 h). The problem of ozone leakage during the ozone fumigation process is meager when compared with the phosphine fumigation (Pimentel et al. 2012) because of the low mobility of insect pests observed in the presence of ozone (Sousa, Faroni, and Guedes 2017), which reinforces its potential use in stored rice grains.

Table 1. Relative error and chi square between experimental and predicted values of ozone concentration.

	Ozone concentration (ppm)			
Time (h)	Experimental	Predicted	Relative error (%)	Chi-square
1	0	0	0	0.00
2	16	24	33	2.67
3	44	65	32	6.78
4	72	106	32	10.91
5	118	147	19	5.72
6	174	187	7.34	0.90
7	230	228	0.59	0.02
8	284	269	5.38	0.84
9	328	310	5.68	1.05
10	368	351	4.78	0.82
11	418	392	6.62	1.72
12	464	432	7.18	2.37
13	504	474	6.38	1.90

Conclusions

The success of ozone fumigation depends on maintaining the required concentration of ozone gas to control the insect pests throughout the grain bed. This article is helpful in understanding the parameters required for governing

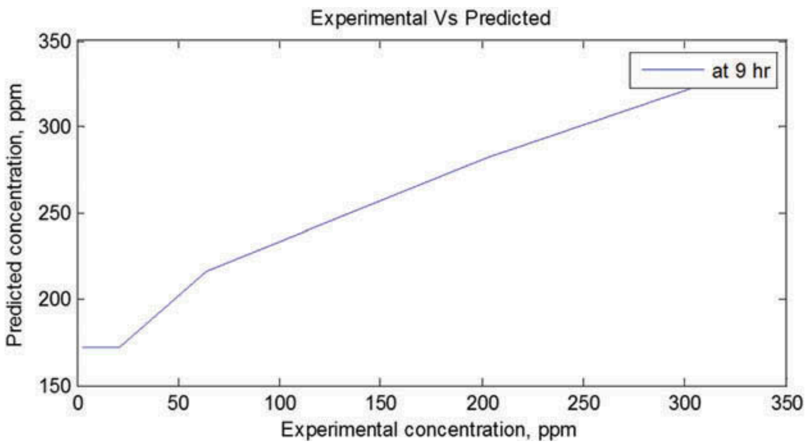

Figure 4. Comparison between experimental and predicted ozone concentrations.

mass transfer between the gaseous ozone and rice grains column. The results suggest that ozone gas fumigation in rice grains will be more effective in still air at minimum bed thickness. The present numerical simulation model is simple, provides acceptable accuracy in prediction, and requires less computational time. This study can offer solutions in predicting the total time required for ozone gas fumigation, designing the bin geometry and management of cost-effective ozone fumigation system.

Acknowledgment

The first author would like to thank Dr. K. Alagusundaram, Former Director, Indian Institute of Crop Processing Technology, Thanjavur, India for providing the ozonation facilities.

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