Advances in Military Technology
Vol. 18, No. 2, 2023, pp. 195-206
ISSN 1802-2308, eISSN 2533-4123
DOI 10.3849/aimt.01850

Modelling of Diffusion Process in Vacuum Drying of Propellants

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The manuscript was received on 30 August 2023 and was accepted after revision for publication as research paper on 29 October 2023.

Abstract:

One crucial step in the propellant-making process that takes a lot of time and energy is drying. The study of inert propellants' vacuum drying behaviours – which mimics those of real propellants – is the main focus of this work. This study presents the development of analytical and numerical models for the vacuum drying of inert propellants, which were validated through an in-house vacuum drying experiment. The calculation of effective diffusivity, a crucial parameter characterising the rate of solvent vapour transfer during the vacuum drying of inert propellant samples, is one of the outcomes of this work. It involves removing liquid solvents from the texture of a propellant. Conclusions and suggestions for technical practice are developed based on the study's findings.

Keywords:

drying kinetics, program MATLAB, propellants, vacuum drying

1 Introduction

Vacuum drying, which works on the basis of creating a vacuum to lower the chamber pressure, is frequently used to dry hygroscopic and heat-sensitive materials. The pressure surrounding the material to be dried is reduced by using vacuum pumps. This causes a significant increase in the rate of evaporation. The result is a faster rate of drying for the product [1].

In the fields of engineering, physics, chemistry, and biology, vacuum technologies have been widely applied. Many industrial processes, including vacuum drying, liquid degassing, food processing, pharmaceutical processing, vacuum coating, and surface technology, depend on the vacuum [2].

Drying is an important stage in propellant manufacturing where liquid solvents are taken out of the propellant microstructure to give the necessary chemical and phys-

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ical properties. When compared to conventional thermal drying procedures, which necessitate extensive heating, vacuum drying stands out as an intriguing technique for drying propellants at low temperatures and pressures. Propellers should be manufactured and stored at a cold temperature in order to slow down the ageing process from a chemical perspective [3]. One possible alternative for producing wax grains for paraffin-based rocket propellants is a rotating casting technology [4].

Drying is a crucial step in the manufacturing of solid propellants that requires a lot of time and energy. As a result, predicting the effective diffusivity – a crucial property dictating the rate of solvent transfer during propellant vacuum drying – is especially helpful for figuring out the best drying conditions for drying system operation and drying process optimisation.

The paper offers two sophisticated methods for estimating the lumped value of the effective diffusivity for the mass transfer process involved in propellant drying, both analytically and numerically. The outcomes of those techniques will be examined and contrasted.

2 Method of Solution

An analytical and numerical solution to a mathematical model explaining the vacuum drying of propellants was produced. Furthermore, the proposed model was validated by conducting the experiment. Concurrently, the optimisation method was used to determine the effective diffusivity in the removal of liquid solvents from the material texture. Fig. 1 depicts the process of solving the problem.

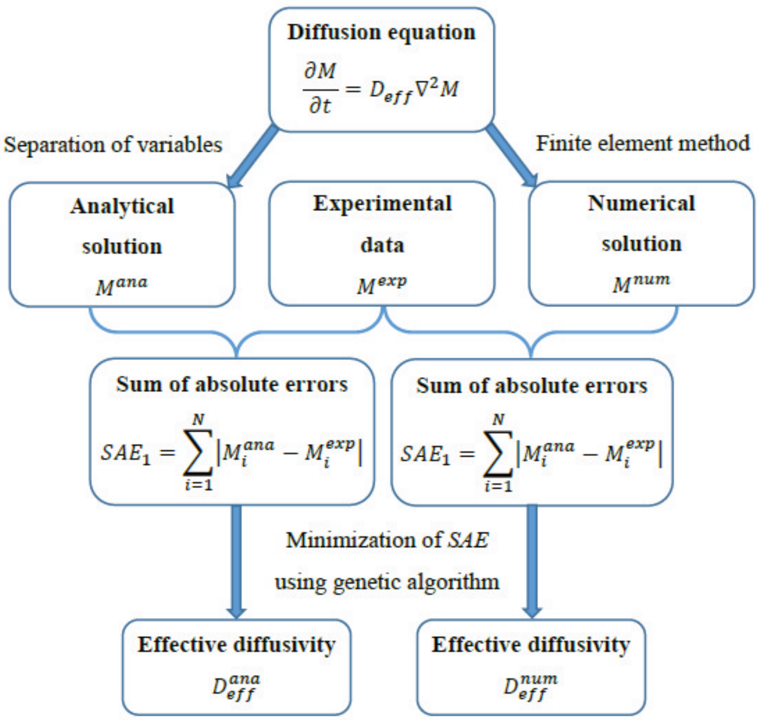

Fig. 1 The solution procedure and problem-solving method

The numerical method – finite element method and the optimization method – genetic algorithm is implemented using PDE toolbox and Global Optimization toolbox respectively provided by MATLAB software.

3 Mathematical Models of Vacuum Drying

Samples of propellants of a cylindrical shape are chosen to investigate their drying behaviours under vacuum drying. The geometry is shown in Fig. 2.

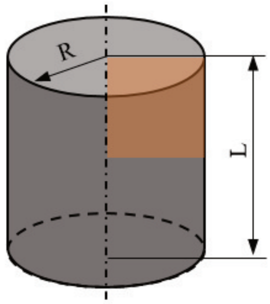

Fig. 2 Propellant sample geometry

To model the mass transfer during the drying process, the following assumptions were made:

- the materials are homogenous and isotropic,
- the liquid migration can be described by a diffusion phenomenon according to Fick's second law,
- the initial moisture content is uniform throughout the materials,
- a thermal equilibrium between the material surface and the drying air is established,
- the shrinkage effect is negligible.

Considering the assumptions, the mass transfer process of drying propellants could be modelled by the diffusion equation:

$$\frac{\partial M}{\partial t} = D_{\text{eff}} \nabla^2 M \tag{1}$$

In the diffusion equation, M is the moisture content, defined as $M = (m - m_d)/m_d$, where m denotes the material mass during the vacuum drying process and m_d denotes the absolute dry mass of the material.

The effective diffusivity D_{eff} in Eq. (1) is the key parameter, which represents the rate of moisture movement including all mechanisms including liquid diffusion, vapour diffusion, surface diffusion, capillary flow, and hydrodynamic flow [3]. Hence, it is important to develop a general procedure of recovering values of D_{eff} analytically, numerically or from experimental data.

3.1 Analytical Approach

This part is focused on the solving of the effective diffusivity D_{eff} by an analytical approach.

Considering that the samples of propellants are of a cylindrical shape (see Fig. 2) in which diffusion takes place only radially, Eq. (1) reduces to

$$\frac{\partial M}{\partial t} = D_{\text{eff}} \left(\frac{\partial^2 M}{\partial r^2} + \frac{1}{r} \frac{\partial M}{\partial r} \right), \; 0 < r < R, \; t > 0 \tag{2a}$$

with the initial and boundary conditions given by

$$M\left(r, t = 0\right) = M_0, \; M\left(r = R, t\right) = M_e, \; M\left(r = 0, t\right) - \text{finite} \tag{2b}$$

Upon introducing the so-called moisture ratio $\hat{M} = \left(M - M_e\right) / \left(M_0 - M_e\right)$, the solution of the problem given by Eq. (1) is readily obtained by separation of variables [5, 6], namely

$$\hat{M} = \sum_{n=1}^{\infty} \frac{4}{\alpha_n^2} \exp\left(-\frac{\alpha_n^2 D_{\text{eff}} \, t}{R^2} \right) \tag{3}$$

where α_n are positive roots of Bessel function of order zero ($\alpha_1 = 2.4048$, $\alpha_2 = 5.5201$, etc.). For a long drying time, the first two terms in Eq. (3) dominate, so that the general form of Eq. (3) could be simplified as

$$\hat{M} = \frac{4}{\alpha_1^2} \exp\left(-\frac{\alpha_1^2 D_{\text{eff}} \, t}{R^2} \right) + \frac{4}{\alpha_2^2} \exp\left(-\frac{\alpha_2^2 D_{\text{eff}} \, t}{R^2} \right) \tag{4}$$

As a result, moisture content could be expressed as

$$M^{\text{ana}} = \left(M_0 - M_e\right) \left[\frac{4}{\alpha_1^2} \exp\left(-\frac{\alpha_1^2 D_{\text{eff}} \, t}{R^2} \right) + \frac{4}{\alpha_2^2} \exp\left(-\frac{\alpha_2^2 D_{\text{eff}} \, t}{R^2} \right) \right] + M_e \tag{5}$$

then, the effective diffusivity is determined by fitting the analytical solution Eq. (5) to experimental data via the optimization technique.

3.2 Numerical Approach

This section is focused on the solving of the effective diffusivity D_{eff} by a numerical approach.

In this study, the finite element method (FEM), a powerful numerical method to deal with partial differential equations, is used to solve the diffusion equation. Considering that the samples of propellants are of cylindrical shape (see Fig. 2) in which diffusion takes place not only radially but also axially, Eq. (1) reduces to

$$\frac{\partial M}{\partial t} = D_{\text{eff}} \left(\frac{\partial^2 M}{\partial r^2} + \frac{1}{r} \frac{\partial M}{\partial r} + \frac{\partial^2 M}{\partial z^2} \right), \;\; 0 < r < R, \;\; 0 < z < \frac{L}{2}, \;\; t > 0 \tag{6a}$$

with the initial constant distribution $M\ (r, z, t = 0) = M_0$ and either of two most common types of boundary conditions, i.e., Dirichlet or Neumann, given by Eq. (6b) and Eq. (6c) respectively

$$\left. \begin{aligned} M\left(r = R, z, t\right) &= M_e \\ M\left(r, z = L/2, t\right) &= M_e \end{aligned} \right\} \tag{6b}$$

$$\left.\begin{aligned} D_{\text{eff}} \left.\frac{\partial M}{\partial r}\right|_{r=R} &= h_{\text{m}}\left[M_e - M\left(r=R,z,t\right)\right], \quad \left.\frac{\partial M}{\partial r}\right|_{r=0} = 0 \\ D_{\text{eff}} \left.\frac{\partial M}{\partial z}\right|_{z=L/2} &= h_{\text{m}}\left[M_e - M\left(r,z=L/2,t\right)\right], \quad \left.\frac{\partial M}{\partial z}\right|_{z=0} = 0 \end{aligned}\right\} \tag{6c}$$

where h_{m} is the convective coefficient.

Due to the symmetry, 3D cylindrical domain is simplified to 2D rectangular domain (orange area) to reduce the computation as shown in Fig. 2.

When Eq. (6) is solved using FEM, the average moisture content over the entire orange domain could be expressed by

$$M^{\text{num}} = \frac{1}{n_{\text{element}}} \sum_{i=1}^{n_{\text{element}}} M_{\text{element}i} \tag{7}$$

where n_{element} is the number of elements over the domain.

Then, the effective diffusivity as well as the convective coefficient are determined by fitting the numerical solution to experimental data via the optimization technique.

3.3 Optimization – Genetic Algorithm

The so-called genetic algorithm was used as an optimisation tool to fit the drying data to analytical and numerical models.

The genetic algorithm is a technique that relies on natural selection, the mechanism underlying biological evolution, to solve optimisation problems that are both confined and unconstrained. An individual solution population is modified repeatedly by the genetic algorithm. The genetic algorithm chooses individuals from the present population to be parents at each phase, using them to create the offspring for the following generation. Over time, the population "evolves" in the direction of the best solution [7]. The primary algorithmic steps are shown in Tab. 1.

Prior to initiating the genetic algorithm optimisation process, the fitness function, ideal variables, and restrictions on these variables must be established as follows:

Tab. 1 Optimization problem description

Description	Analytical model	Numerical model	
		with Dirichlet boundary condition	with Neumann boundary condition
Objective function	$\sum_{j=1}^{N}\left\lvert M_j^{\text{ana}} - M_j^{\text{exp}}\right\rvert$ M_j^{ana} is the analytical solution given by Eq. (5) at a given time	$\sum_{j=1}^{N}\left\lvert M_j^{\text{num}} - M_j^{\text{exp}}\right\rvert$ M_j^{num} is the numerical solution given by Eq. (7) at a given time	
Optimal variable	D_{eff}	D_{eff}	D_{eff}, h_m
Constraint	$D_{\text{eff}} > 0$	$D_{\text{eff}} > 0$	$D_{\text{eff}} > 0$, $h_{\text{m}} > 0$

4 Vacuum Drying Experiment

In experiments, inert samples of propellant, provided by the Explosia company, Pardubice, Czech Republic, were used. The samples were of a cylindrical shape and their dimensions are shown in Fig. 3.

Fig. 3 Inert samples of propellant (left) and their geometry (right), dimensions in [mm]

The inert samples of propellant were dried in a vacuum drying chamber (VACUCELL 55, Laboratory of Department of Mechanical Engineering, University of Defence, Brno), as shown in Fig. 4, with various drying conditions – different pressures and temperatures, respectively. During experiments, the mass of inert samples of propellant was recorded every hour in first 4 hours and then every 12 hours using an electronic scale with a sensitivity of 1 mg. The vacuum drying experiments lasted until the mass change was less than or equalled to 1 mg.

Fig. 4 Vacuum drying chamber (left) and electronic scale (right)

Experimental moisture content M and moisture ratio $\widehat{M}$ were calculated based on the collected weight data as follows:

$$\left.\begin{aligned} M &= \frac{m - m_{\mathrm{d}}}{m_{\mathrm{d}}} \\ \widehat{M} &= \frac{M - M_e}{M_0 - M_e} \end{aligned}\right\} \qquad (8)$$

5 Results and Discussion

Results of the solution provide us with the time variations of the moisture ratio for the above described approaches to modelling of diffusion process in vacuum drying of the given propellant.

5.1 Drying Kinetics

The moisture ratio time variations of inert samples of propellant resulted from the analytical and numerical models are shown in Figs 5-7 for two scales of time.

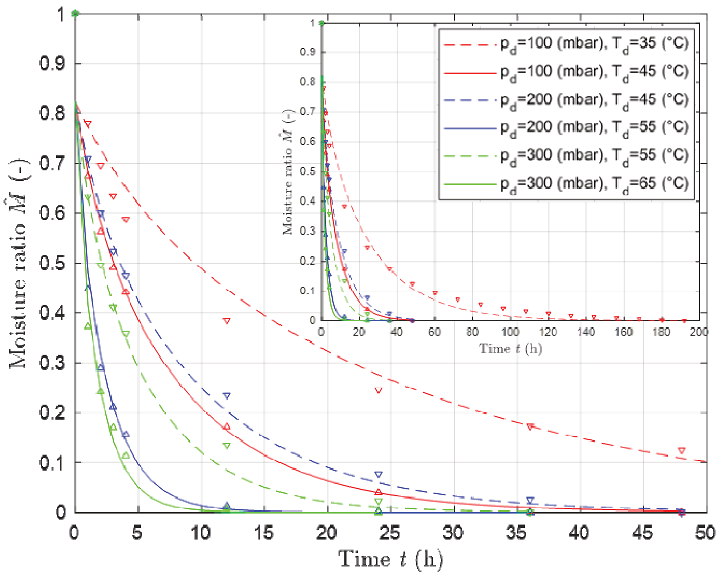

Fig. 5 Results of analytical model for moisture ratio of 5 mm *long and* 3.5 mm *in diameter inert samples of propellant*

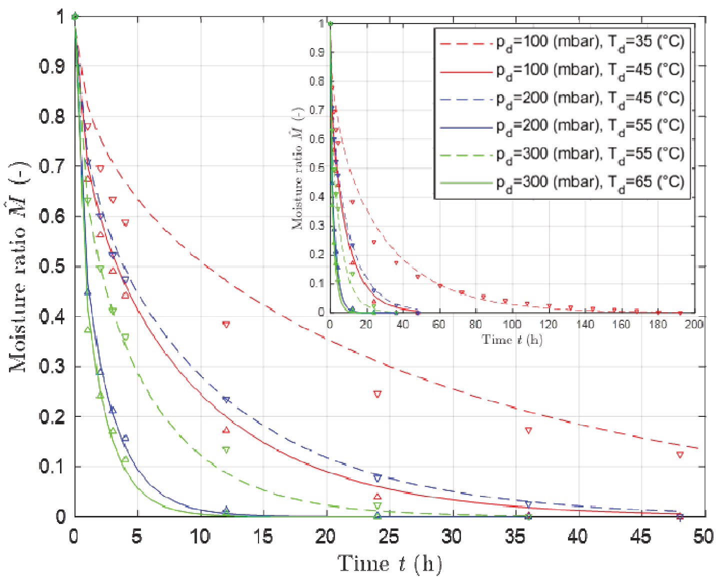

Fig. 6 Results of numerical model with Dirichlet boundary condition for moisture ratio of 5 mm *long and* 3.5 mm *in diameter inert samples of propellant*

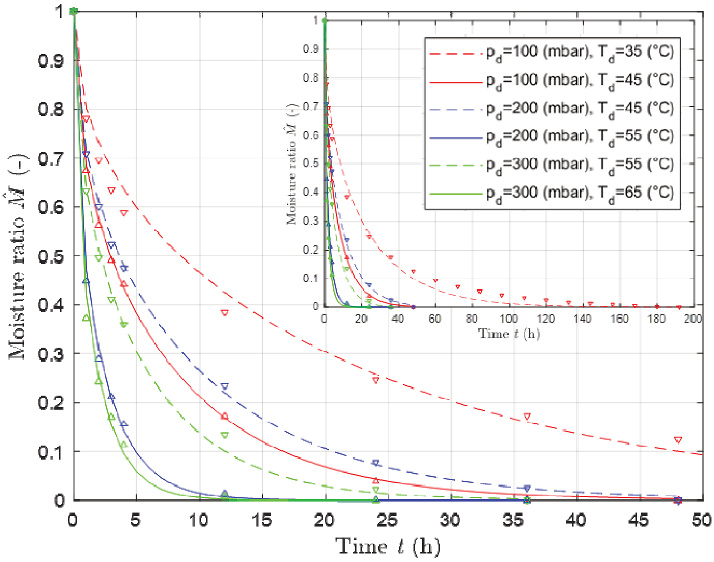

Fig. 7 Results of numerical model with Neumann boundary condition for moisture ratio of 5 mm *long and* 3.5 mm *in diameter inert samples of propellant*

It is evident that, in general, the moisture ratio determined by the analytical and numerical models fits quite well with the experimental data, which is shown as points in diagrams. The best agreement, though, seems to be with the numerical model that makes use of the Neumann boundary condition. Tab. 2 provides a qualitative assessment of model accuracy in order to corroborate these intuitive remarks.

5.2 Effective Diffusivity

The optimal values of effective diffusivity are detailed in Tab. 2. Besides, the coefficient of determination R_d^2 and sum of absolute errors (SAE) are calculated and serve as the criteria for evaluating the models. The higher the value of R_d^2 or the lower the value of *SAE* is, the better is the goodness of fit.

In this study, the effective diffusivity values resulted from the analytical model range from 5.6629×10^{-12} to 77.4075×10^{-12} m^2/s, while the ones resulted from the numerical models range from 7.3329×10^{-12} to 113.5898×10^{-12} m^2/s and from 9.0465×10^{-12} to 111.3805×10^{-12} m^2/s.

It is obvious that the drying temperature and drying vacuum pressure affect the effective diffusivity. Specifically, the drying temperature has a considerable influence on the effective diffusivity. At a given pressure, its values roughly triple with every 10 °C increase in temperature, with the exception of vacuum pressure 20 kPa, when they roughly quadruple.

5.3 Simulation of Vacuum Drying of Propellants

MATLAB can be used to simulate the moisture content field of the propellant inert samples during vacuum drying once the effective diffusivity has been established. Figs 8-13 provide an example of the numerical model's simulation results using the Neumann boundary condition. The moisture content field of inert propellant samples 5 mm long and 3.5 mm in diameter is displayed with different drying conditions.

Regarding the impact of the drying conditions, which include the drying temperature and drying pressure, it is seen that the drying conditions have a major influence on how long the vacuum drying process takes for the propellant samples that are inert. The drying process proceeds more quickly with the higher drying temperature and the lower drying pressure. As a result, drying temperature has a much bigger influence on accelerating the drying process than drying pressure.

Tab. 2 Effective diffusivity of 5 mm *long and* 3.5 mm *in diameter inert samples*

Model	**Drying conditions**		**Optimal variables**		R_d^2	**SAE**
	Vacuum pressure p_d [kPa]	**Temperature** T_d [°C]	**Effective diffusivity** $D_{eff} \cdot -10^{12}$ [m²/s]	**Convective coefficient** h_m [m/s]		
Analytical	10	35	5.6629	—	0.9750	0.0329
		45	17.6151	—	0.9668	0.0123
	20	45	15.0341	—	0.9652	0.0139
		55	58.1495	—	0.9577	0.0137
	30	55	25.7673	—	0.9568	0.0143
		65	77.4075	—	0.9515	0.0152
Numerical (with Dirichlet boundary condition)	10	35	7.3329	—	0.9854	0.0282
		45	22.9617	—	0.9942	0.0107
	20	45	19.8268	—	0.9970	0.0063
		55	90.2182	—	0.9967	0.0056
	30	55	38.8591	—	0.9940	0.0075
		65	113.5898	—	0.9941	0.0072
Numerical (with Neumann boundary condition)	10	35	9.0465	0.0065	0.9940	0.0217
		45	26.1291	0.0081	0.9997	0.0023
	20	45	21.1031	0.0056	0.9995	0.0027
		55	88.2280	0.0046	0.9987	0.0034
	30	55	36.1022	0.0031	0.9980	0.0039
		65	111.3805	0.0048	0.9962	0.0050

DOI 10.3849/aimt.01850

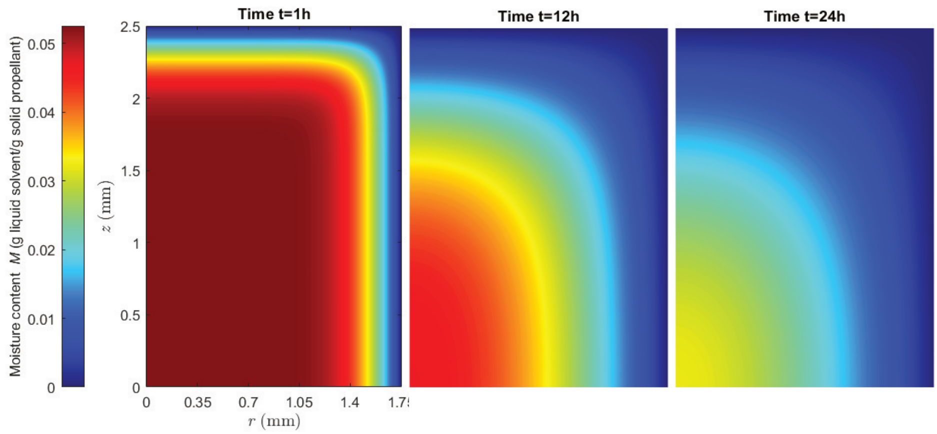

Fig. 8 Simulation for drying conditions: p_d = 10 kPa *and* T_d = 35 °C

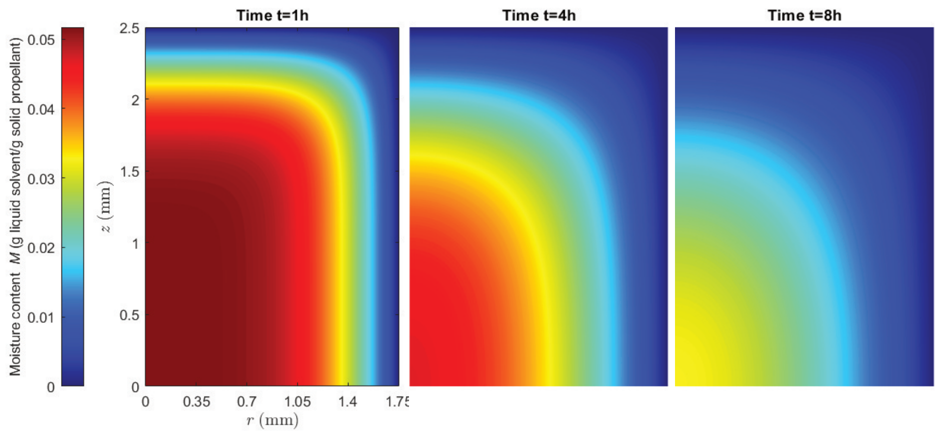

Fig. 9 Simulation for drying conditions: p_d = 10 kPa *and* T_d = 45 °C

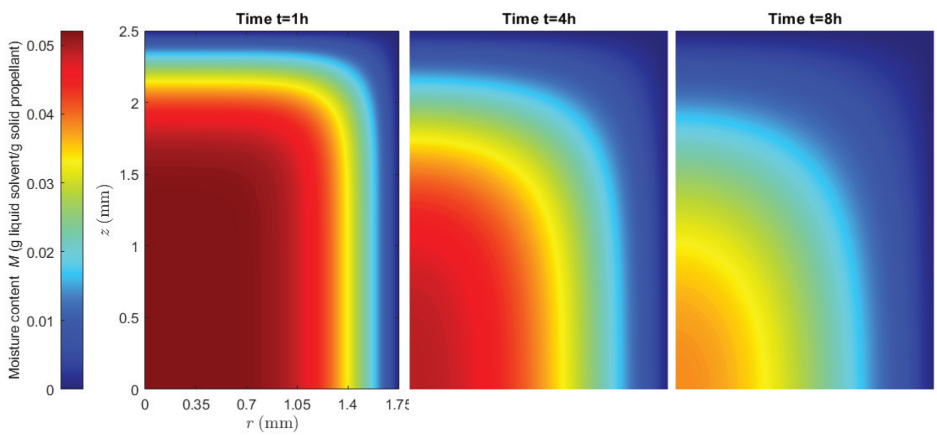

Fig. 10 Simulation for drying conditions: p_d = 20 kPa *and* T_d = 45 °C

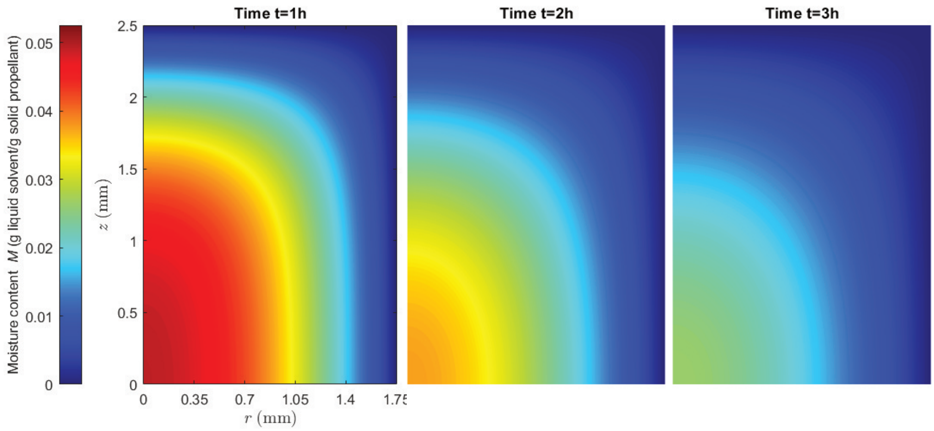

Fig. 11 Simulation for drying conditions: p_d = 20 kPa *and* T_d = 55 °C

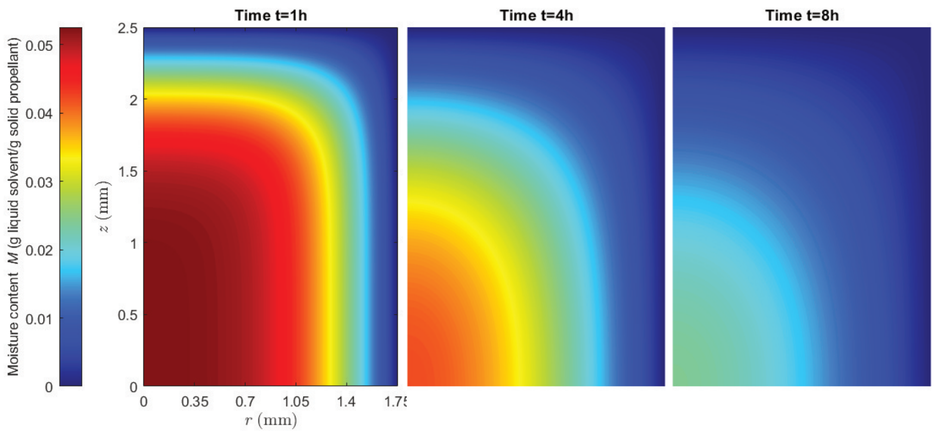

Fig. 12 Simulation for drying conditions: p_d = 30 kPa *and* T_d = 55 °C

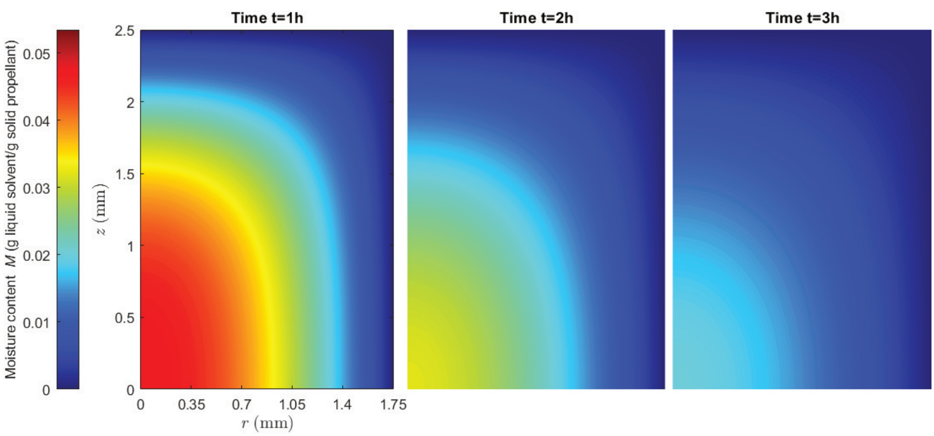

Fig. 13 Simulation for drying conditions: p_d = 30 kPa *and* T_d = 65 °C

6 Conclusion

As part of the study, several mathematical models based on Fick's second law of diffusion were created to forecast propellant drying behaviours under various vacuum drying settings. Furthermore, an estimation was made of the effective diffusivity, a crucial characteristic parameter that defines the path of the vacuum drying process as a whole. The MATLAB software's PDE toolbox and Global Optimisation toolbox were utilised to solve the issue. When liquid solvents are removed from propellants, the solution's outcome shows the time variations of the propellants' drying kinetics as well as the effective diffusivity values.

The experiment validated the models that were provided. Results from the experimental drying data are in good agreement with the analytical and numerical models. It was evident how temperature and pressure affected the propellant samples' inert samples' drying behaviours. Propeller inert samples dry more quickly at higher temperatures and lower vacuum pressures.

Maintaining low temperatures inside the vacuum chamber during the drying of hazardous materials like propellants reduces the probability of unplanned explosions and maintains the stability of the material, which is crucial to the propellants' long-term viability. This is one of the main benefits of vacuum drying.

Determining the propellants' effective diffusivity and drying kinetics is a highly helpful application. This is the way how to guarantee efficient drying, which is a crucial requirement for low-vacuum drying system design and operation.

Acknowledgement

The authors would like to thank the Faculty of Military Technology for the support by the Institutional Funding DZRO VAROPS and the specific research project SV23-216.

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