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## Numerical investigation on freeze-drying of aqueous material frozen with pre-built pores☆

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## ABSTRACT

Freeze-drying of the initially porous frozen material with pre-built pores from liquid material was found experimentally to save drying time by over 30% with an initial saturation being 0.28 compared with the conventional operation with the initial saturation being 1, using mannitol as the solid material. In order to understand the mass and heat transfer phenomena of this novel process, a two-dimensional mathematical model of coupled mass and heat transfer was derived with reference to the cylindrical coordinate system. Three adsorption–desorption equilibrium relationships between the vapour pressure and saturation value namely, power-law, Redhead's style and Kelvin's style equation, were tested. Kelvin's style in exponential form of adsorption equilibrium relation gave an excellent agreement between the model prediction and experimental measurement when the equation parameter,  $\gamma$ , of 5000 was applied. Analyses of temperature and ice saturation profiles show that additional heat needs to be supplied to increase the sample temperature in order to promote the desorption process. Simulation also shows that there is a threshold initial porosity after which the drying time decreased with the increase in the initial porosity. Enhanced freeze-drying is expected to be achieved by simultaneously enhancing mass and heat transfer of the process.

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## 1. Introduction

Freeze-drying or lyophilisation is a dehydration method that can significantly reduce product damage caused by thermal drying. It plays a unique role in the processing of delicate and heat-sensitive materials of high values such as food, pharmaceuticals and biological products. However, freeze-drying is the most sophisticated and expensive process of all drying techniques, both in capital investment and in operational expenses [1]. Therefore, reduction of freeze-drying time in order to lower energy consumption and raise productivity has been a worldwide challenge during the past decades [2].

There are a few ways of process enhancement that can minimize drying time on a consistent basis while maintaining acceptable quality of freeze-dried products. The obvious one is to conduct freeze-drying, from the viewpoint of energy saving, at the eutectic temperature for a liquid phase that crystallizes during freezing or the glass transition temperature for a liquid phase that remains amorphous [3]. The alternative way is to combine freeze-drying with microwave heating, which has

demonstrated its potential to enhance the internal heat transfer due to the interaction between the microwave and lossy media [4,5]. Excessive microwave energy input, however, could cause frozen materials to melt and collapse, which would ruin the overall freeze-drying process.

Although modern industry has been benefited from the existing knowledge of physical and chemical processes during freeze-drying, fundamental formulations in some areas are still incomplete. Microwave freeze-drying with the assistance of dielectric materials has successfully settled the heat transfer problem. The mass transfer problem still remains unsolved. It is necessary to re-examine the freeze-drying process. There are usually three stages in freeze-drying: freezing stage, primary drying stage, and secondary drying stage. Freezing is the first stage of a freeze-drying process, and the performance of the overall process is closely related to this stage. The shape of pores, pore size distribution and pore connectivity of porous dried layer formed by ice sublimation depend largely on the size of ice crystals during the freezing stage [3,6]. This dependence is of extreme importance because the mass transfer rate is significantly affected by the porous structure of the dried layer [7,8]. If ice crystals are small and discontinuous, the mass transfer rate of vapour in the dried layer would be limited. However, if large ice crystals are formed and homogeneous dispersion in the frozen solution can be realized, the mass transfer rate could be high and materials could be dried more quickly [9,10]. The prevailing transfer resistance of freeze-drying is from water vapour migration in the dried region,

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depending on the size of ice crystals formed during freezing [11,12]. Also, mass transfer parameters during freeze-drying are strongly dependent on the textural and morphological parameters of the ice phase [13].

Unlike fruits and vegetables with naturally formed porous structures that may be partially filled with water, liquid material to be freeze-dried is usually frozen into the solid material without initial voids. The latter freeze-drying operation is known to be lengthy and expensive. It was therefore proposed that the liquid material is first frozen into the porous material with certain porosity and then freeze-dried [14]. Generally, such a frozen material is partially filled with ice crystals like ice-cream. It would be beneficial to promoting the freeze-drying rate because of the increased area of heat and mass transfer initially. Experimental studies have showed that the freeze-drying process can be significantly enhanced using the initially porous frozen material [15,16]. The aims of the present work are to perform a numerical investigation using proper adsorption-desorption relationships to reproduce and predict the kinetics process under the experimental drying conditions, and to examine the heat and mass transfer phenomena during the freeze-drying of the initially porous material.

## 2. Mathematical Model

In the present simulation, a two-dimensional mathematical model of the cylindrical coordinate system was derived based on the understanding of heat and mass transfer mechanism developed previously [17]. The main assumptions made in the model formulation have been reported elsewhere [2]. In order to match the experimental tests, the sample geometry during simulation exactly followed the experimentally used one as shown in Fig. 1.

In the supporting pad,

$$\rho_m c_m \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \lambda_m \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left( \lambda_m \frac{\partial T}{\partial z} \right). \quad (1)$$

In the material region,

$$\varepsilon \rho_i \frac{\partial S}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r K_S \frac{\partial S}{\partial r} \right) + \frac{\partial}{\partial z} \left( K_S \frac{\partial S}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( r K_T \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left( K_T \frac{\partial T}{\partial z} \right) \quad (2)$$

$$(\rho c)_e \frac{\partial T}{\partial t} = \frac{1}{r} \frac{\partial}{\partial r} \left( r \lambda_e \frac{\partial T}{\partial r} \right) + \frac{\partial}{\partial z} \left( \lambda_e \frac{\partial T}{\partial z} \right) + \frac{1}{r} \frac{\partial}{\partial r} \left( r K_S \Delta H \frac{\partial S}{\partial r} \right) + \frac{\partial}{\partial z} \left( K_S \Delta H \frac{\partial S}{\partial z} \right) \quad (3)$$

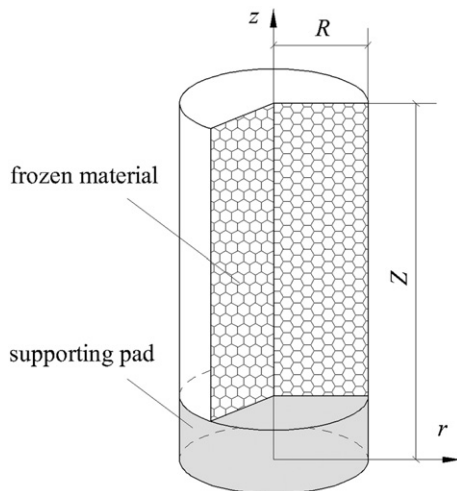


Fig. 1. Sample geometry.

where

$$K_S = \left[ \frac{K}{\mu_v} R_v \rho_v T + \frac{(1-S)\varepsilon D}{\tau} \right] \frac{\partial \rho_v}{\partial S}$$

$$K_T = \left[ \frac{K}{\mu_v} R_v \rho_v T + \frac{(1-S)\varepsilon D}{\tau} \right] \frac{\partial \rho_v}{\partial T} + \frac{K}{\mu_v} R_v \rho_v^2$$

$$\lambda_e = (1-\varepsilon)\lambda_s + \varepsilon S \lambda_i + \varepsilon(1-S)\lambda_v + K_T \Delta H$$

$$(\rho c)_e = (1-\varepsilon)\rho_s c_s + \varepsilon S \rho_i c_i + \varepsilon(1-S)\rho_v c_v + \varepsilon(1-S)\Delta H \frac{\partial \rho_v}{\partial T}.$$

Initial temperature and ice saturation are uniform.

$$T|_{t=0} = T_0 \quad (4)$$

$$S|_{t=0} = S_0 \quad (5)$$

At the central line of the sample, there are no mass and heat fluxes.

$$\nabla S|_{\text{centl}} = 0 \quad (6)$$

$$\nabla T|_{\text{centl}} = 0 \quad (7)$$

On the inner surface of the supporting pad, there is no mass flux.

$$\nabla S|_{\text{insf}} = 0 \quad (8)$$

On the sample surface, heat transfer is governed by radiation and drying chamber has a constant pressure.

$$-\lambda_e \nabla T|_{\text{surf}} = \sigma \varepsilon F (T_{\text{surf}}^4 - T_{\text{amb}}^4) \quad (9)$$

$$\rho_{v,\text{surf}} = \frac{P_{\text{amb}}}{R_v T_{\text{surf}}} \quad (10)$$

It has to be pointed out that heat and mass flux balance equations at sublimation interface were not involved in the present model as a boundary condition. A boundary is known to be a border that encloses an area or a space. The important feature of a boundary in the sense of transport phenomena is, in the authors' understanding, that there is an abrupt change in physical properties around areas adjacent to the boundary. In the modelling of freeze-drying, an artificially moving speed of sublimation interface was usually introduced as an additional boundary condition. It is reasonable for the freeze-drying of the initially solid frozen material due to the absence of initial pores. However, it is difficult to justify this introduction for the initially porous frozen material based on physical considerations. The term  $\partial R/\partial t$  is an independent term, which assumes that the moving speed of interface at any location contributes to a special heat and mass transfer that is different from other areas for the uniform, homogeneous and isotropic porous matrix. It is the additional term that results in an additional variable, *i.e.*, the interface position,  $R$ , in a mathematical model and causes additional complexity of model solving. Based on such understanding, some researchers presented their simulation results with a sharp change in ice saturation occurring at the interface although the materials to be dried were initially unsaturated [4,17,18], which seems to be physically unrealistic. More importantly, the sublimation interface should be inherently formed during drying, rather than inserted as a constraint for the initially unsaturated frozen material. Recent studies have confirmed this judgement [19]. Current simulation results further verify that the sublimation interface can be indeed formed naturally as seen subsequently.

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