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Aggregation and caking processes of granular materials: continuum model and numerical simulation with application to sugar

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Abstract—Aggregation and caking of particles are common severe problems in many operations and processing of granular materials, where granulated sugar is an important example. Prevention of aggregation and caking of granular materials requires a good understanding of moisture migration and caking mechanisms. In this paper, the modeling of solid bridge formation between particles is introduced, based on moisture migration of atmospheric moisture into containers packed with granular materials through vapor evaporation and condensation. A model for the caking process is then developed, based on the growth of liquid bridges (during condensation), and their hardening and subsequent creation of solid bridges (during evaporation). The predicted caking strengths agree well with some available experimental data on granulated sugar under storage conditions.

Keywords: Caking; aggregation; heat transfer; moisture migration; granular materials; simulation.

NOMENCLATURE

A	surface area of a single particle (m^2)
b	radius of narrowest part of the solid bridge (m)
c_{ps}	specific heat of granulated sugar (J/°C kg)
C_1, C_2	constants in (10)

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D_a	diffusion coefficient of water vapor in air (m^2/s)
D_e	effective diffusion coefficient of water vapor in air around a solid (m^2/s)
ERH	equilibrium relative humidity of air
F	sum of areas of all particles in unit volume (m^2)
k	mass transfer coefficient for sugar dehydration (l/s)
K	total solids moisture and air vapor content in an element (kg)
L_h	latent heat of water vaporization (J/kg)
m_a	mass of air in an element (kg)
m_s	mass of sugar in an element (kg)
m_{syrup}	mass of syrupy layer around a single sugar grain (kg)
m_{water}	mass of moisture around a single sugar grain (kg)
N	number of particles in unit volume
P	ambient pressure (Pa)
R	average radius of sugar particles (m)
RH	relative humidity of air
R_v	individual gas constant for water vapor (J/kg/K)
T	temperature of air or granular sugar (°C)
t	time (s)
V	volume of a single particle (m^3)
v	total volume around a single sugar grain available for solid bridge formation (m^3)
v_b	volume of solid bridge for a single sugar grain around the contact area between two grains (m^3)
v_{syrup}	total volume around a single sugar grain available for liquid bridge formation (m^3)
v_{tot}	total volume around a single sugar grain available for both solid and liquid bridge formation (m^3)
W	moisture content of granulated sugar (% of kg water/kg sugar)
W_0	initial moisture content of granulated sugar (% of kg water/kg sugar)
W_{eq}	equilibrium moisture content of granulated sugar (% of kg water/kg sugar)
x	Cartesian coordinate
Y	absolute humidity of air (kg water/kg dry air)
Y_{eq}	equilibrium absolute humidity of air at the grain surface (kg water/kg dry air)
Y_m	oscillating absolute humidity average of air at exposed boundary (kg water/kg dry air)
Y_s	saturated absolute humidity (kg water/kg dry air)

Greek

α	constant in (14)
$\gamma_1, \gamma_2, \gamma_3$	constants in (14)
ΔV	elemental volume in the domain containing solid and gas (m^3)
ΔY	amplitude of oscillation for the square-wave absolute humidity pulse (kg water/kg dry air)
ε	porosity of granulated sugar
ξ	dimensionless geometry-dependent coefficient
λ_s	thermal conductivity of granulated sugar (J/s m °C)
ρ_a	density of air (kg/m^3)
ρ_s	solids density of granulated sugar (kg/m^3)
ρ_{syrup}	density of syrupy layer (kg/m^3)
σ_{cake}	tensile strength of caked solid (Pa)
$\sigma_{crystal}$	crystalline strength of material making up the solid bridge (Pa)

1. INTRODUCTION

Caking is an undesirable phenomenon in which free-flowing powder agglomerates, compromising quality and handling. The problems occur in a wide range of powder industries and natural environments, such as foods, fertilizers, pharmaceuticals and soils. Caking is a major issue in the storage and transportation of granulated sugar, illustrating the relevance of the present work.

Despite a number of mechanisms being responsible for caking, the two dominant causes are compressive caking and moisture migration caking. As sugar tends to be transported under the absence of high loading, this paper is primarily concerned with moisture migration caking. Moisture migration caking occurs mainly through a cycle of adsorption/desorption of moisture between air and solid particles, largely because of the particles being crystallized from a mother liquor. Some moisture is retained in the crystal structure, whilst a thin layer of mobile moisture within an amorphous syrup forms outside [1–4].

Generally, the processes of powder caking due to moisture migration can be summarised in the following steps [5–7]: (i) wetting and moisture absorption, (ii) liquid bridging, (iii) drying and moisture desorption, (iv) hardening and solid bridging, and (v) compacting and caking. When particles take up moisture their surfaces are hydrated or wetted by moisture. Inter-particle liquid bridges are then formed by cohesion and adhesion. Recrystallization follows and solid bridges are formed when excess water evaporates from the particles' surface during a drying cycle. The recrystallized solid bridges formed exhibit the required strength to maintain the two particles in contact. This irreversible consolidation of bridges results in particle agglomeration. If the wetting and drying processes are repeated, this will result in a moisture migration cycle. Interparticle space is reduced and

interparticle bridges are thickened. Thus, caking strengths would depend on the rate of moisture migration and the solubility of the hygroscopic granular materials.

Although considerable work has been carried out, both theoretical and experimental, on the subject of caking [2, 4, 5], numerical simulation of caking processes is relatively recent [8, 9]. The present work is believed to be unique in its coupling between heat/mass transfer and predictions of caking strength due to the migration of moisture in a computational continuum mechanics framework. The motivation was essentially a better understanding of the behavior of caked solids, and a quantitative measure of caking which would help explain changes in the condition of a powder over time. In practice, it is particularly important to predict the ultimate caking strength of a specific material, which varies with ambient environmental conditions such as temperature, pressure and humidity. In order to achieve this, it is necessary to develop a predictive model based on the caking mechanism, which accounts for the dependence of the caking strength on the environmental conditions, as well as the characteristics and properties of the solid.

The aim of this paper is to introduce a mathematical model of moisture migration and solid bridge growth, which can then be employed to predict the increase in caking strength under wetting/drying cycles. In Section 2, moisture transfer in porous media is discussed. In Section 3, the equations for the solid bridge and caking strength are derived. Section 4 details the solution procedure. In Section 5, predictions of caking strength during cyclic processes and comparisons against experimental data are presented, and the dependence of caking strength on environmental conditions and material properties is discussed.

2. MOISTURE TRANSPORT MODEL

2.1. Energy and mass balance equations, and initial and boundary conditions

Previous experimental studies into sugar caking by Leaper *et al.* [10] used a cylindrical tensile test cell, which was used in a carefully controlled experiment to determine tensile strength in a sample of granulated sugar. The diameter of the test cell was 95 mm and its height was 12.7 mm. The cell was placed in a humidity cabinet, where the only interaction allowed with the environment was through the exposed top surface. The side walls and bottom of the cell were non-permeable to moisture. This experimental setup was used as framework for the simulations carried out in this study, and allowed the use of a simplified arrangement of a one-dimensional slab through the central axis of the test cell as the computational domain for the modeling of moisture transfer and caking (Fig. 1). Hence, the energy and mass balance equations need only be solved in one dimension. For the construction of the mathematical model, the following simplifications have been made:
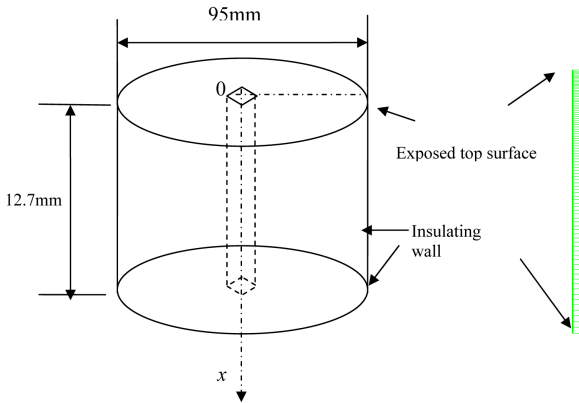
Figure 1. Experimental tensile test cell and computational domain. This figure is published in colour on http://www.ingenta.com

- The material is considered to be homogenous and isotropic in the macroscopic sense.
- Hysteresis in the relation between the moisture potential and the moisture content is not taken into account.
- Local thermal and moisture equilibrium conditions exist among all phases.
- Shrinkage and swelling effects in sugar are neglected as solid absorbs and desorbs moisture, so that the porosity remains constant.
- Convection and radiation transport phenomena are negligible compared to diffusion.
- Ambient pressure is constant.
- The ambient temperature was kept constant and isothermal conditions are assumed for moisture migration, since the temperature gradient inside the moisture layer is negligible, as predicted by the equations.

Based on the above assumptions, the one-dimensional governing equations for heat and mass transfer are previously given as follows:

2.1.1. Heat transfer.

$$(1 - \varepsilon)\rho_s c_{ps} \frac{\partial T}{\partial t} = \frac{\partial}{\partial x}\left(\lambda_s \frac{\partial T}{\partial x}\right) + L_h(1 - \varepsilon)\rho_s \frac{\partial W}{\partial t}, \tag{1}$$

where c_{ps} and λ_s are the specific heat capacity and thermal conductivity for granulated sugar, respectively, L_h is the latent heat of water vaporization, ε is the porosity (fraction of the total volume of the package occupied by air), and W is the moisture content of the solid (in %, representing the mass of liquid water in the solid per solid mass).
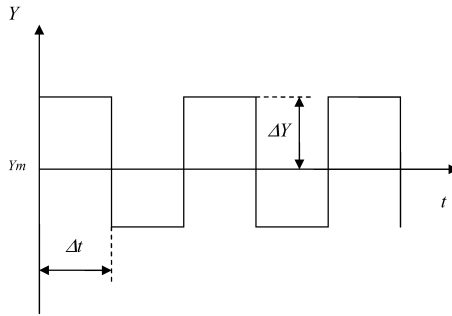
Figure 2. Square wave profile of boundary absolute humidity.

2.1.2. Mass transfer.

$$\frac{\partial Y}{\partial t} = \frac{\partial}{\partial x}\left(D_e \frac{\partial Y}{\partial x}\right) - \frac{(1-\varepsilon)\rho_s}{\varepsilon\rho_a}\frac{\partial W}{\partial t},\tag{2}$$

where D_e is the effective diffusion coefficient for water vapor in air around the solid, and Y is dimensionless and represents the mass of water vapor in the air around the solid particles.

2.1.3. Initial and boundary conditions.

The initial conditions are $T = T_0$, $W = W_0$ and $Y = Y_{eq}$ (as dictated by the initial values of T and W, assuming that the system was initially at equilibrium) at $t = 0$. At the sides of the computational domain, free-slip conditions have been assumed, hence the only interactions occurred along the x-direction. There was also no interaction between material and environment through the bottom of the cell. In the humidity cabinet the ambient temperature was kept constant and only the relative humidity was changed in a square-wave pattern as shown in Fig. 2, in order to impose the wetting/drying cycles, hence the boundary value of the air absolute humidity at the exposed surface was determined through the temperature and the ambient air relative humidity.

2.2. The effective diffusion coefficient

The effective diffusion coefficient of vapor in the air around the sugar granules is the product of the porosity and the diffusion coefficient of vapor in dry air, D_a. By employing the formula for D_a, which is a function of temperature and total pressure [11], the effective diffusion coefficient can be expressed as follows:

$$D_e = \varepsilon \cdot 2.11 \times 10^{-5}\left(\frac{273.15 + T}{273.15}\right)^{1.94}\left(\frac{1013.25}{P \times 10^{-2}}\right)^2,\tag{3}$$

where T is in °C and P is in Pa.

For the present case, the ambient pressure may be considered as constant and equal to atmospheric pressure. Therefore, the above equation may be simplified as:

$$D_e \approx \varepsilon \cdot 2.11 \times 10^{-5} \left(\frac{273.15 + T}{273.15} \right)^{1.94}. \tag{4}$$

2.3. Source terms

The source term on the right-hand side of (1) and (2) represents the increase or decrease in temperature due to the release or absorption of heat during condensation or evaporation and the subsequent change in the mass of vapor in the air around the solid particles. Effectively, this term describes moisture production or destruction in a unit volume of material at the micro-scale, and accounts for the solid and gas (air + vapor) phases, the solid bound moisture, and the thin layer of liquid syrup around the grains, which results from the mixing on the interface between the condensed vapor and the sugar grain. The transfer rate of moisture from/onto the solid surface is dependent on concentration gradients, a rate of mass transfer and the area of solid–air interface. In this way, the net mass transfer flux of moisture may be obtained on the basis of micro-mass balance:

$$(1 - \varepsilon)\rho_s \frac{\partial W}{\partial t} = -\frac{\varepsilon \rho_a \xi D_a}{F}(Y_{eq} - Y), \tag{5}$$

where F is the area of solid–air interface in an elemental volume ΔV, Y_{eq} is the equilibrium absolute humidity at the grain surface, ξ is a dimensionless geometry-dependent coefficient and D_a is the diffusion coefficient of vapor in dry air. As an approximation, F is equal to the sum of all particle surfaces in ΔV:

$$F = NA,$$

where A is the surface area of a single particle and N is the number of particles in the elemental volume, which is equal to the total solids volume in an element divided by a single particle volume:

$$N = \frac{(1 - \varepsilon)\Delta V}{V}. \tag{6}$$

Assuming spherical particles, A and V are given by: $A = 4\pi R^2$ and $V = (4/3)\pi R^3$.

Substituting N, A and V into (5), the following expression for the source term was derived:

$$(1 - \varepsilon)\rho_s \frac{\partial W}{\partial t} = -\frac{\varepsilon \rho_a \xi R D_a}{3(1 - \varepsilon)\Delta V}(Y_{eq} - Y), \tag{7}$$

where R is the average particle radius.

Based on (7), we can define a mass transfer coefficient, k, as follows:

$$k = \frac{\xi R D_a}{3(1 - \varepsilon)\Delta V}. \tag{8}$$

Thus, (7) may be written as:

$$(1 - \varepsilon)\rho_s \frac{\partial W}{\partial t} = -\varepsilon \rho_a k (Y_{eq} - Y),\tag{9}$$

where, following Christakis [11]:

$$Y = RH \cdot Y_s = RH \cdot \frac{C_1 e^{-C_2/(273.15+T)}}{\varepsilon \rho_a R_v (273.15 + T)},\tag{10}$$

hence:

$$Y_{eq} = ERH \cdot Y_s = ERH \cdot \frac{C_1 e^{-C_2/(273.15+T)}}{\varepsilon \rho_a R_v (273.15 + T)}.\tag{10a}$$

In practice, the particles are not regular spheres of equal radii; however, this approach will greatly simplify the solution and will still indicate trends. Presently, all undetermined factors have been included into a dimensionless geometry-dependent coefficient, ξ. In this study, ξ was taken to be unity — an approximation which will be demonstrated to account satisfactorily for the behavior of granulated sugar. Since there can be no direct measurements of the mass transfer coefficient [3], the parameter ξ must be calibrated to experimental data for a particular material in order to fit the sorption/desorption behavior of that material. It is interesting to note that (7) is very similar to the expressions employed by Rastikian and Capart [3], Warren and Root [12], and Zimmerman *et al.* [13] to describe the rate of change of the solids moisture content.

2.4. Sorption isotherms

The total amount of moisture within a control volume must be balanced. As the relative humidity of air changes, this results in disturbing the equilibrium of the system. Consequently, either vapor will be absorbed by the grains or evaporation from the grain surface will occur, so that the system again reaches equilibrium. There are numerous empirical correlations which link the equilibrium relative humidity to the moisture content of hygroscopic materials. Toledo [14], for example, quoted 12 different expressions often applied to food substances. In this paper, an alternative analysis is offered for the derivation of the sorption isotherms for granulated sugar, based on a semi-empirical relation, by Leaper *et al.* [8] which fits experimental data. The moisture balance in a control volume can be obtained from the equation:

$$K = m_a \cdot Y + \frac{m_s W}{100},\tag{11}$$

where, following (10), $Y = RH \cdot Y_s$. Hence, at equilibrium, (11) may be re-written as:

$$K = m_a \cdot ERH \cdot Y_s + \frac{m_s W_{eq}}{100}.\tag{12}$$

The semi-empirical relationship between W_{eq} and *ERH* for granulated sugar can be obtained from experimental data [3]:

$$W_{eq} = \left(\gamma_1 + \gamma_2 ERH^{\gamma_3} \right) \left(\frac{T}{20} \right)^{\alpha}. \tag{13}$$

Substituting from (13) for W_{eq} into (12), the equilibrium relative humidity can be obtained through the solution of the quadratic:

$$\frac{m_s \gamma_2}{100} \left(\frac{T}{20} \right)^{\alpha} ERH^2 + m_a Y_s ERH + \frac{m_s \gamma_1}{100} \left(\frac{T}{20} \right)^{\alpha} - K = 0, \tag{14}$$

where γ_1, γ_2, γ_3 and α are dimensionless constants, specific to granulated sugar, as given in Leaper *et al.* [8], K may be calculated via (11) (where the current values of Y and W may be used), and Y_s can be obtained following Christakis [11]:

$$Y_s = \frac{C_1 e^{-C_2/(273.15+T)}}{\varepsilon \rho_a R_v (273.15 + T)}.$$

3. SOLID BRIDGE FORMATION AND CAKING MODEL

The process of moisture migration, which causes caking is continuous: an increase in local relative humidity and moisture uptake by the particles, liquid bridge formation, a decrease in local relative humidity, drying and moisture evaporation, formation and hardening of solid bridges. When relative humidity increases beyond its equilibrium value, either due to a decrease in temperature or an increase in the vapor content of the ambient air, hygroscopic particles will take up moisture in their attempt to drive the system into equilibrium, with particles combining to form liquid pendular bridges at contact points. Drying occurs as relative humidity reduces below its equilibrium value, so that moisture will evaporate from the particles surface. Christakis [11] gives a comprehensive discussion on the equilibrium behavior of hygroscopic materials. An expression for the radius of the solid bridge can be found in Tanaka [1]:

$$b = 0.82R \left(\frac{v_b}{R^3} \right)^{\frac{1}{4}}, \tag{15}$$

where v_b is the volume of the bridge for a single sugar grain around the contact area between two grains, R is the radius of the grain and b is the radius at the narrowest part of the bridge. The volume v_b is given by the equation:

$$v_b = 2v\varepsilon/\pi, \tag{16}$$

hence, (15) becomes:

$$b = 0.82R \left(\frac{2\varepsilon}{\pi R^3} \right)^{\frac{1}{4}} v^{\frac{1}{4}}, \tag{17}$$

where v is the total volume available around the surface of a single sugar grain for solid bridge formation.

Volume v can be calculated by tracking changes in the total volume available around the surface of a grain for the formation of both liquid and solid bridges, v_{tot}, during a wetting/drying cycle. This volume v_{tot} is the sum of two volumes: the volume v available for solid bridge formation and the volume v_{syrup} available for liquid bridge formation. Volume v_{syrup} is the volume of the syrupy layer formed around a single grain during the wetting process. The creation of the syrupy layer occurs when the topmost layers of a sugar grain get saturated by the condensing water. It has been found that this layer is saturated with water by 40% [10]; hence, the mass of the syrupy layer for a single sugar grain can be obtained through the equation:

$$m_{syrup} = \frac{m_{water}}{0.4} = \frac{W(1 - \varepsilon)\rho_s \Delta V/N}{100 \cdot 0.4}, \qquad (18)$$

where m_{water} is the mass of moisture around the surface of a single grain and N is the total number of grains in an elemental volume ΔV, as given by (6). By knowing that the density of the syrupy layer ρ_{syrup} is approximately equal to the solids density of sugar ρ_s, v_{syrup} can be calculated as:

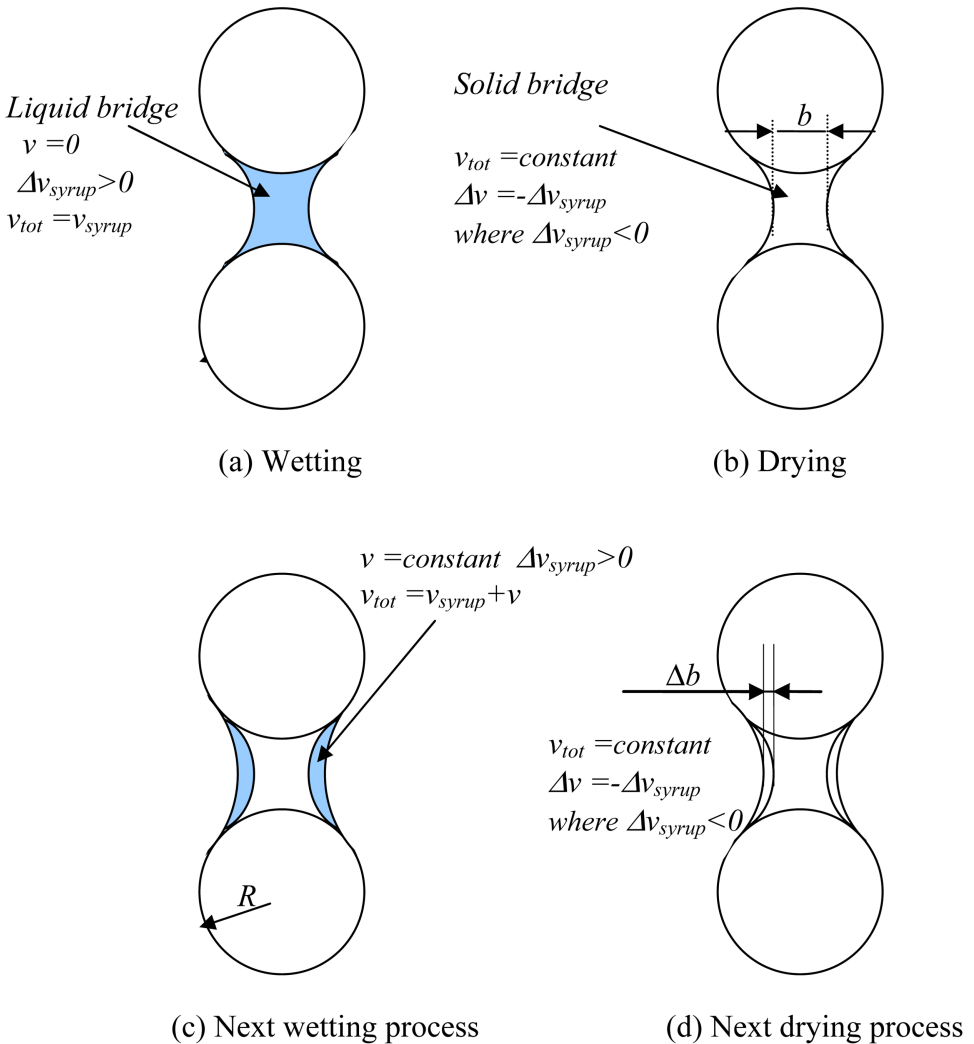$$v_{syrup} = \frac{m_{syrup}}{\rho_{syrup}} \approx \frac{W(1 - \varepsilon)\Delta V}{100 \cdot 0.4N}. \qquad (19)$$

Then, by tracking changes in the moisture content of the solids in an elemental volume, changes in v_{syrup} may be obtained. Initially, v is equal to zero, with no solid bridges formed; hence, $v_{tot} = v_{syrup}$. A schematic representation of the formation and growth of a solid bridge between two grains is given in Fig. 3. In this way, the radius of the solid bridge b may be calculated (through changes in v) and the tensile strength σ_{cake} of the caked material may be determined using Rumpf's equation [1, 10]:

$$\sigma_{cake} = \sigma_{crystal}\pi b^2 \frac{9}{8}\frac{(1 - \varepsilon)}{\varepsilon}\left(\frac{1}{2R}\right)^2, \qquad (20)$$

where $\sigma_{crystal}$ is the crystalline strength of the material making up the bridge. A value for $\sigma_{crystal}$ for granulated sugar can be found in Leaper *et al.* [10].

For the above analysis, it was assumed that every sugar grain in an elemental volume will have a neighboring grain with which they will combine to form a liquid–solid bridge. All solid bridges in the elemental volume are considered to be identical, much smaller in volume than the sugar particles and have equal strength, and the formed cake will only fail at the narrowest part of the bridges and nowhere else. The sugar grains were approximated as spherical with an average diameter of $2R$. The porosity of granulated sugar is known not to vary

Liquid bridge
$v = 0$
$\Delta v_{syrup} > 0$
$v_{tot} = v_{syrup}$

Solid bridge
$v_{tot} = constant$
$\Delta v = -\Delta v_{syrup}$
where $\Delta v_{syrup} < 0$

b

(a) Wetting

(b) Drying

$v = constant$ $\Delta v_{syrup} > 0$
$v_{tot} = v_{syrup} + v$

Δb

$v_{tot} = constant$
$\Delta v = -\Delta v_{syrup}$
where $\Delta v_{syrup} < 0$

R

(c) Next wetting process

(d) Next drying process

Figure 3. Schematic diagram of solid bridge formation and caking during a wetting/drying cyclic process. This figure is published in colour on http://www.ingenta.com

significantly with ambient conditions [10] and was kept constant in (20). Hence, the only parameter varying with time in (20) was the solid bridge radius b and by tracking changes in b, the increase in the tensile strangth of the formed cake can be determined. As will be demonstrated during the presentation of the results, despite the simplifying approximations employed, the numerical model managed to predict accurately both qualitatively and quantitatively the effects of moisture migration on a sample of granulated sugar which was subjected to a cycling wetting/drying process.

4. NUMERICAL CONSIDERATIONS AND SOLUTION PROCEDURE

The governing equations are discretized by using the finite volume method [15]. Non-uniform hexahedral cells were employed in this paper. The mesh used is shown in Fig. 1. After discretization, complete elimination of non-linearities in the set of equations for temperature and absolute humidity is not possible, since the source term in these equations is a non-linear function of temperature and absolute humidity. Hence, an iterative procedure is employed with the following steps: (i) start with initial temperature and absolute humidity, (ii) using these starting values an equilibrium absolute humidity can be calculated, which is then used to update temperature and moisture content, (iii) update absolute humidity using the updated values of equilibrium absolute humidity and moisture content, (iv) return to Step 2 and repeat the calculation procedure using the updated values of temperature and absolute humidity until convergence is achieved for all solved parameters, and (v) use the converged solution for solids moisture content in order to calculate the increase in solid bridge radii (during drying) and update the tensile strength of the cake. Thermodynamic and transport properties can be updated simultaneously, as the solution is iterative. This solution procedure has been implemented within PHYSICA, a three-dimensional, fully unstructured, Fluid Dynamics/Solid Mechanics Finite Volume code, developed at the University of Greenwich [9].

In the experiment which was simulated in the present paper, the ambient temperature was kept constant and only the relative humidity was varied. The heat transfer equation (1) was solved in the computational domain and the predicted temperature gradients were very small; hence, maintaining the temperature constant in the domain throughout the simulation. In reality, it is possible for both environmental temperature and relative humidity to vary over time, and the numerical model has been shown to be capable of simulating moisture migration under varying environmental temperature conditions [16]. Predictions of tensile strength during cycling environmental conditions, with both external temperature and relative humidity varying, will be discussed in the following sections.

5. RESULTS AND DISCUSSIONS

5.1. Simulation conditions and numerical errors

A description of the simulated tensile test cell, the mesh employed and the application of boundary conditions is given in Section 2.1. Initially, the granulated sugar in the test cell was assumed at equilibrium at 20°C and its moisture content was 0.033%. This value did not include the inherent bound moisture of granulated sugar (approximately 0.035% [10]), which did not migrate and, hence, did not influence the caking process. Initial absolute humidity (assuming equilibrium) was taken as 0.00835 kg/kg. The relative humidity at the exposed boundary was set to 70% for 4 h, followed by a 4-h period of 20% relative humidity, with the
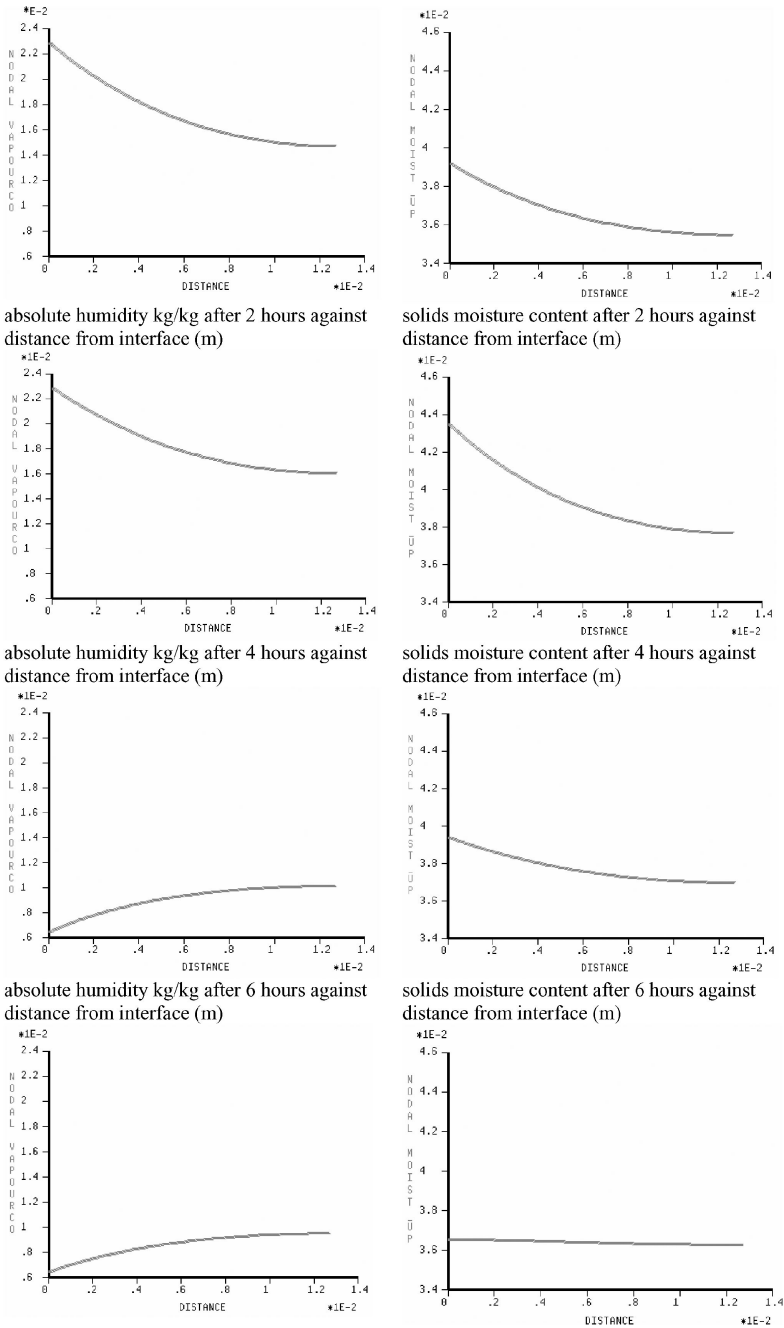
Table 1.
Thermodynamics constants and material properties

Material properties and coefficients	Value	Unit
C_1	2.53×10^{11}	Pa
C_2	5.42×10^3	K
c_{ps}	2087.3	J/kg/°C
L_h	2272000	J/kg
R_v	461.5	J/kg/K
α	0.6	
ε	0.365	
λ_s	0.208	J/m s °C
γ_1	0.03	
γ_2	0.075	
γ_3	2	
ξ	1.0	
$\sigma_{crystal}$	1.5×10^3	Pa
ρ_a	1.275	kg/m³
ρ_s	1660	kg/m³

temperature in the humidity cabinet being kept constant at 20°C. A total of 32 such wetting/drying cycles were performed. Equation (10) was employed for the calculation of the oscillating absolute humidity values at the material–environment interface (0.0228 kg/kg for 70% relative humidity and 0.0065 kg/kg for 20% relative humidity, both at 20°C). Material properties and constants used in the simulations are given in Table 1. After some preliminary optimization, a 90-element mesh and a 10-s time step were chosen, in order to guarantee stability in the solution.

5.2. Dynamic processes of moisture migration and caking

Profiles of absolute humidity in kg of water per kg of air and percentage solids moisture content of the caked material along the direction of the axis of the tensile test cell are shown for the first two cycles at 2-h intervals in Fig. 4a with the corresponding tensile strength in Pascals in Fig. 4b. In the first 4 h, the boundary absolute humidity was at its peak and the absolute humidity in the domain increased due to diffusion. Hence, the relative humidity in the simulated strip would increase (since temperature did not significantly deviate from 20°C and could be considered as constant) and the granulated sugar would take up some of the excess moisture in its attempt to bring the system back into equilibrium [11]. The formation of liquid bridges between particles has occurred; however, as these have negligible tensile strength compared to solidified bridges, Fig. 4b shows the bulk tensile strength at this stage as zero. When the exposed boundary relative humidity dropped, as shown at $t = 6$ and $t = 8$ h, the boundary absolute humidity also dropped to a minimum and because of diffusion, the absolute humidity in the domain started
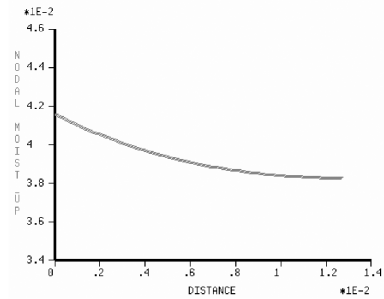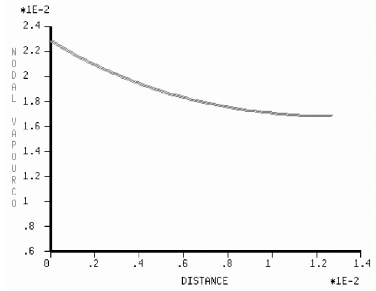
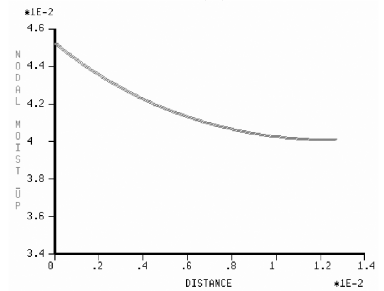absolute humidity kg/kg after 2 hours against distance from interface (m)

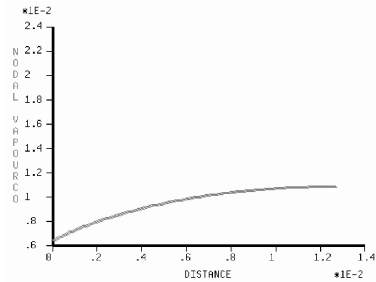solids moisture content after 2 hours against distance from interface (m)

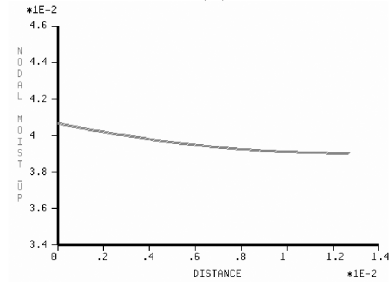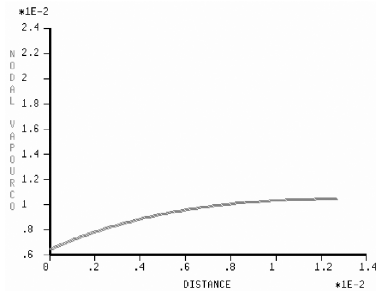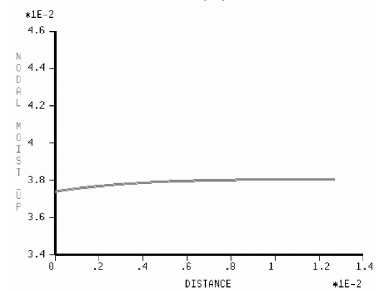absolute humidity kg/kg after 4 hours against distance from interface (m)

solids moisture content after 4 hours against distance from interface (m)

absolute humidity kg/kg after 6 hours against distance from interface (m)

solids moisture content after 6 hours against distance from interface (m)

(a, left)

Figure 4. (a) Profiles along the central axis of the test cell of humidity and solids moisture content for two of the 8-h wetting/drying cycles. (b) Profiles along the central axis of the test cell of bulk tensile strength (Pa) for two of the 8-h wetting/drying cycles.

absolute humidity kg/kg after 8 hours against distance from interface (m)
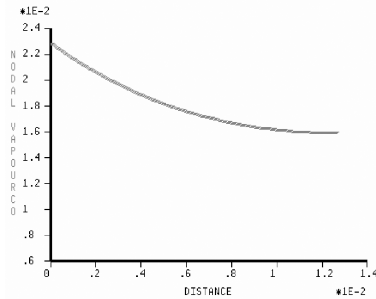
solids moisture content after 8 hours against distance from interface (m)

absolute humidity kg/kg after 10 hours against distance from interface

solids moisture content after 10 hours against distance from interface (m)

absolute humidity kg/kg after 12 hours against distance from interface

solids moisture content after 12 hours against distance from interface (m)

absolute humidity kg/kg after 14 hours against distance from interface

solids moisture content after 14 hours against distance from interface (m)

absolute humidity kg/kg after 16 hours against distance from interface

solids moisture content after 16 hours against distance from interface (m)
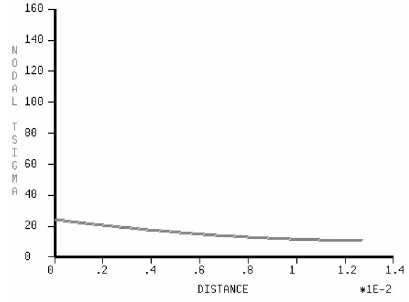
(a, right)

Figure 4. (Continued).
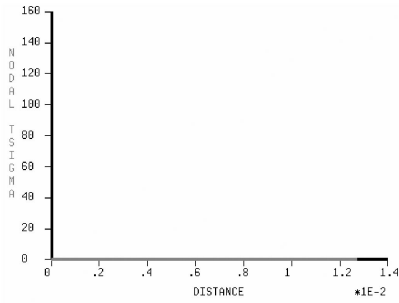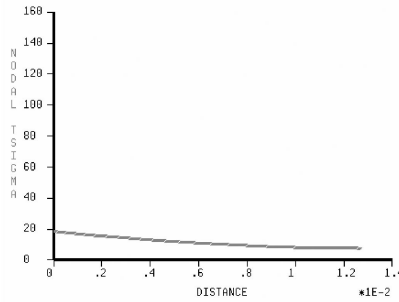
bulk tensile strength after 2 hours (Pa)
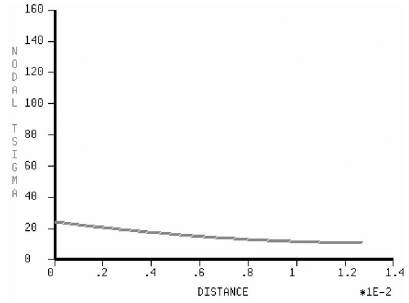
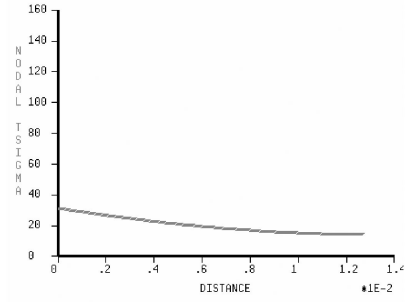bulk tensile strength after 10 hours (Pa)
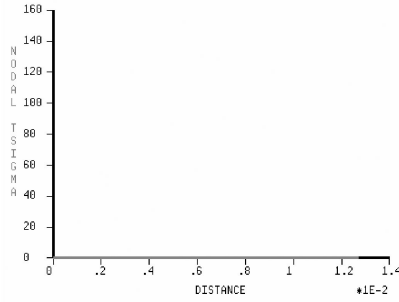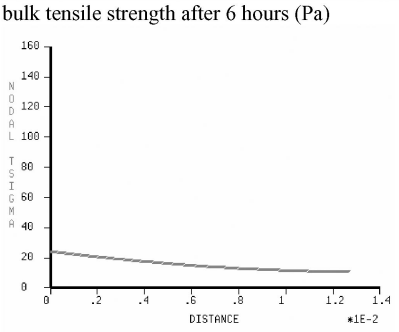
bulk tensile strength after 4 hours (Pa)

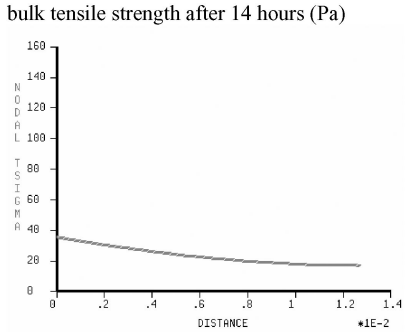bulk tensile strength after 12 hours (Pa)

bulk tensile strength after 6 hours (Pa)

bulk tensile strength after 14 hours (Pa)

bulk tensile strength after 8 hours (Pa)

bulk tensile strength after 16 hours (Pa)

(b)

Figure 4. (Continued).

decreasing. Hence, the relative humidity in the simulated strip would decrease (at constant temperature) and moisture would evaporate from the surface of the granulated sugar in order to bring the system back to equilibrium [11]. During this drying process, as has already been discussed, liquid bridges hardened and led to the formation of solid bridges; hence, the increase of the tensile strength in the material as shown in Fig. 4b. During the following wetting/drying cycle, granulated sugar exhibited a similar behavior, adsorbing moisture during the wetting process and evaporating moisture during the drying process, which led to the increase of the solid bridge radius, and, consequently, the tensile strength of the material. As can be seen, this increase occurred during the drying process, with the strength not changing during wetting. Continuing wetting/drying cycles and subsequent moisture migration would lead to the hardening of the formed solid bridges and the increase in the tensile strength, thus causing the sugar to cake.

It was observed that, because of the small dimensions of the test cell, changes to the exposed surface humidity affected the humidity/moisture/tensile strength along the whole length of the simulated strip, with the sharpest changes occurring in the vicinity of the exposed boundary distance. This result was in agreement with the simulated behaviour of granulated sugar during moisture migration in big bags [16], where the sharpest interaction between environment and material was predicted to occur within the first 0.2 m from the material–environment interface.

5.3. Comparison between predicted tensile strength and experimental data

As experiments of caking processes are time consuming, expensive and often very complicated to carry out, not enough experimental data under carefully controlled conditions are available in literature. Recently, Leaper *et al*. [10] performed tensile strength measurements on granulated sugar in a humidity cabinet under repeated wetting/drying cycles using an experimental tensile test cell (as described in Fig. 1). The 32-cycle wetting/drying simulation described in Section 5.2 reproduced the 32 wetting/drying cycles of Leaper *et al*. [10] using the same initial and boundary conditions (as given in Section 5.1). A comparison between the experimentally measured and predicted values of tensile strength at the sugar–environment interface is shown in Fig. 5. As can be seen, there was excellent agreement between the numerical model and the experiment, within the uncertainty of $\pm 4\%$ of the experimental measurements.

The tensile strength increased sharply during the first few cycles. Further increase of the strength was seen to occur along an almost straight line. This indicated that the first few cycles were critical in the formation and the ultimate strength of the caked material. Further numerical tests were then performed in order to assess the dependence of the caking strength on the boundary conditions, cycling intervals and porosity of the tested material.
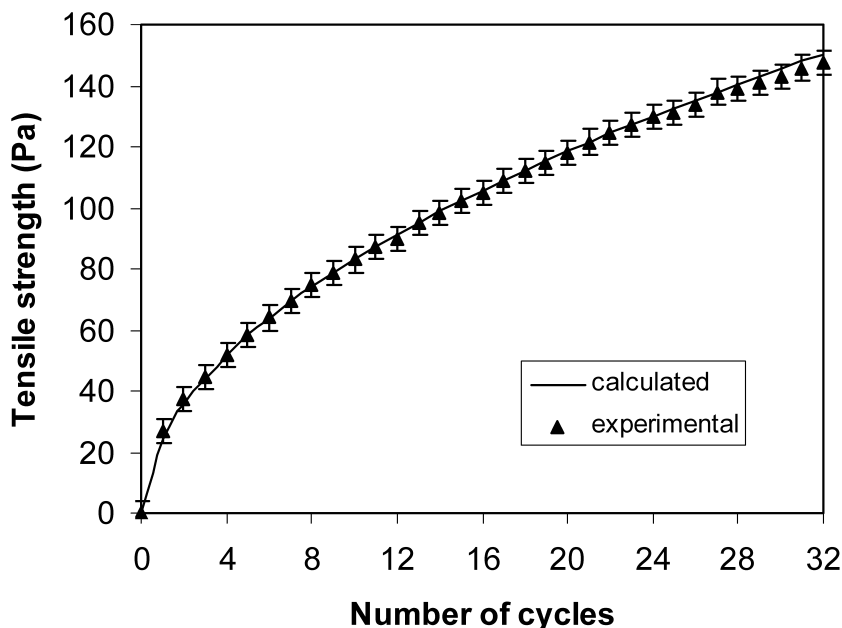
Figure 5. Comparison between predicted and experimentally measured tensile strength after 32 8-h wetting/drying cycles for a granulated sugar sample in a tensile test cell.

5.4. Effect of relative humidity

The effect of different relative humidities at the exposed boundary was studied in a series of simulations, where the minimum environmental relative humidity was kept constant at 20%, and the maximum value was changed to 87.7 and 37.6%. These values of relative humidity correspond to absolute humidities of 0.035 and 0.015 kg/kg, respectively at the environment–sugar interface. A comparison between the predicted caking strengths at the environment–material interface for 32 8-h wetting/drying cycles is presented in Fig. 6. As expected, the tensile strength increased when the boundary humidity increased, since higher humidity would mean an increase in the amount of moisture which the sugar absorbed during wetting. This would lead to an increase in the liquid bridge radius during wetting and, hence an increase in the solid bridge radius during drying, which meant that the tensile strength of the caked material would increase.

5.5. Effect of intervals of oscillation

Different wetting/drying cycle oscillation intervals were employed to make up for the same total time of 256 h of the 32 8-h cycles for which the granulated sugar was exposed to. In the results presented in Fig. 7, the material was subjected to 12-h wetting/drying cycles (6-h half cycle interval) and 16-h wetting/drying cycles (8-h half cycle interval) between 70 and 20% relative humidity. Figure 7 shows that
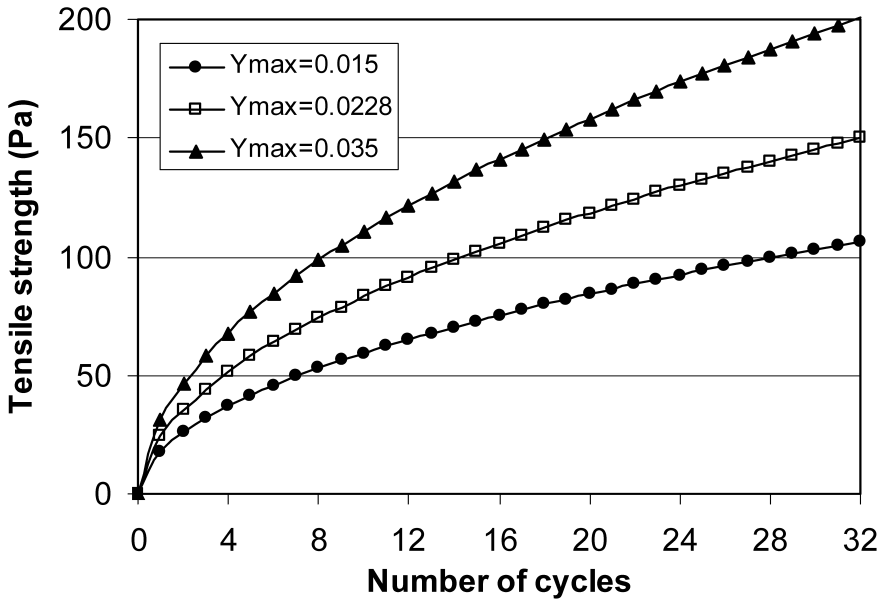
Figure 6. Effect of boundary maximum absolute humidity on tensile strength of caked sugar.
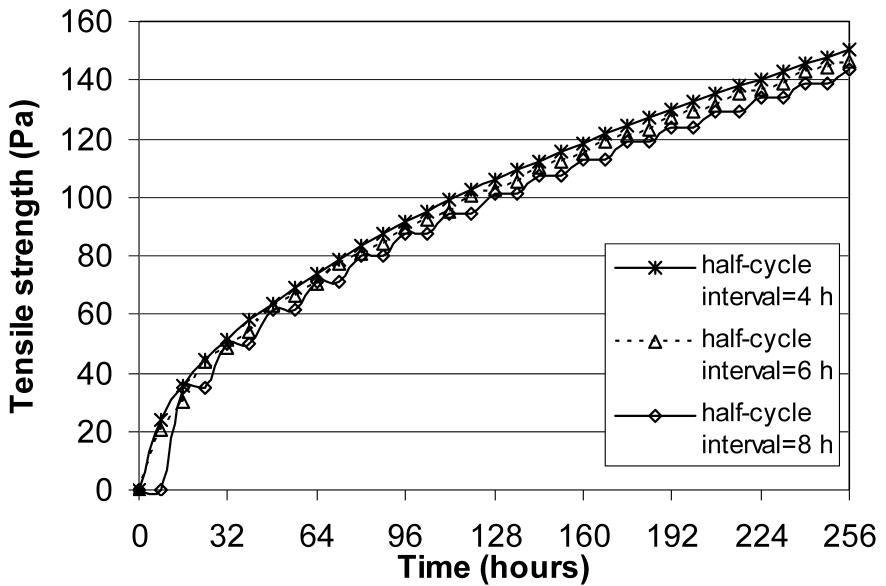
Figure 7. Effect of different cycling intervals on tensile strength over the same overall exposure time to a cycling wetting/drying process.

changes in the tensile strength of sugar at the environment–material interface were not significant for the different cycling intervals employed.
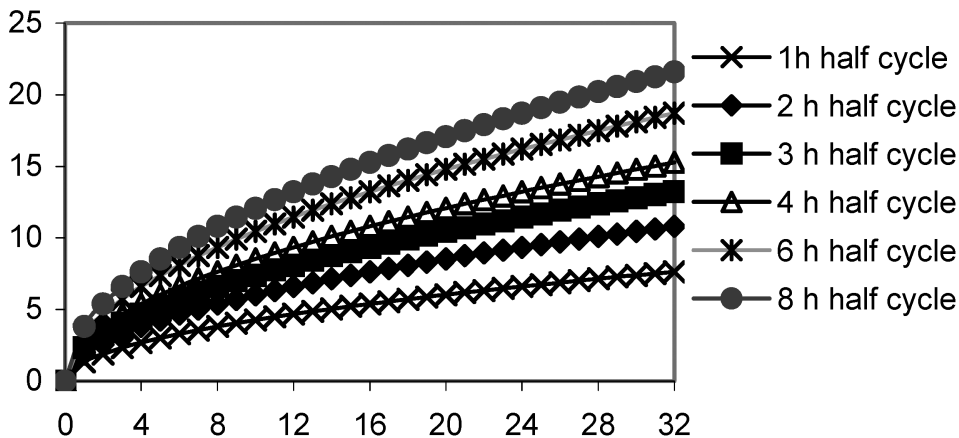
**Figure 8.** Effect of different half-cycle intervals on tensile strength over the same number of cycles.

This was confirmed by Fig. 8, which was a comparison of tensile strengths at the sugar–environment interface for different half-cycle intervals for a total of 32 wetting/drying cycles with the same boundary conditions. Regardless of the frequency of humidity cycling, the critical factor that determined tensile strength was the total time at high humidity.

5.6. Effect of porosity

As deduced by the analysis in Section 3, the tensile strength is highly dependent on the material porosity. Hence, simulations were performed to assess the effect of porosity changes to the predicted strength of the caked sugar. The oscillating boundary humidity conditions were not changed and 32 8-h wetting/drying cycles were simulated. Two values, one lower and one higher than the initial porosity of 0.365, were employed: 0.2 (sugar grains more tightly packed) and 0.5 (sugar grains less tightly packed). Figure 9 presents the results of these simulations. As expected, the tensile strength of the caked sugar increased with decreasing porosity. When the sugar grains were closely packed, the distances between them were smaller; hence, the solid bridges created between the particles would be expected to be more stable and thus, harder to break.

5.7. Tensile strength predictions under varying environmental temperature and humidity conditions

As already has been mentioned, in reality both environmental temperature and relative humidity are expected to vary with time. In order to illustrate the ability of the numerical model to represent moisture migration and caking with both environmental temperature and relative humidity varying, a simulation was performed, where 10 24-h drying/wetting cycles were simulated where both environmental temper-
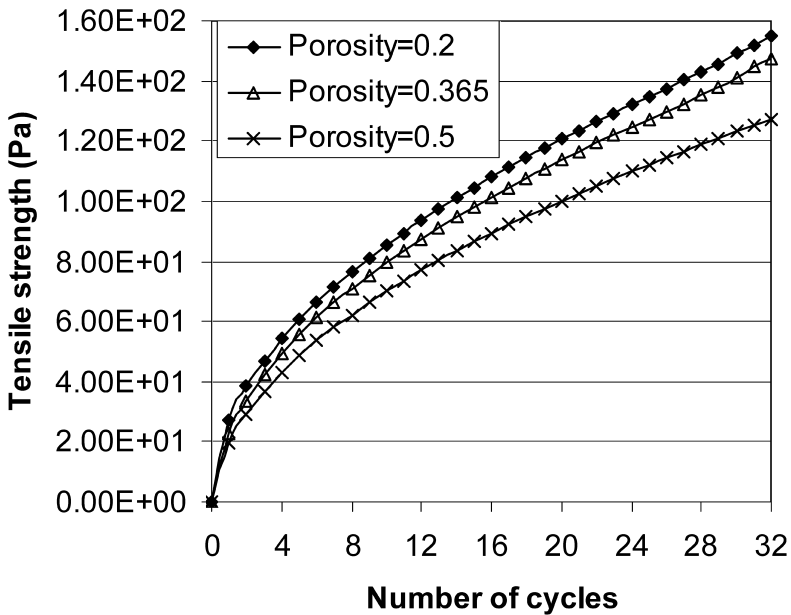
Figure 9. Effect of material porosity on the tensile strength for a 32-cycle wetting/drying process.

ature and relative humidity were allowed to vary periodically. The environmental boundary conditions were chosen in order to represent typical day-time and night-time environmental conditions which would lead to moisture migration phenomena and, possibly, caking. The day-time environmental temperature and relative humidity (which would cause drying) were taken as 27°C and 38%, respectively (corresponding to a boundary absolute humidity of 0.0185 kg/kg). Night-time environmental conditions (wetting) were taken as 16°C and 76% relative humidity (corresponding to a boundary absolute humidity of 0.0193 kg/kg). These conditions were imposed at the exposed boundary of the tensile test cell, and varied periodically between day-time and night-time every 12 h. Initial conditions and material properties were taken as given in Section 5.1 and Table 1. Figure 10 presents the increase in the tensile strength of the caked sugar at the material–environment interface at the end of each cycle. The increase in the tensile strength of caked sugar was lower than in the previously simulated 32 8-h cyclic processes (although the exposure times to the cycling processes were comparable). The boundary absolute humidity values were very close to each other, so that the gradients of absolute humidity in the domain, which would drive moisture migration, would be small. Hence, the moisture uptake/loss by the grains would be less than the 32-cycle process and the solid bridges formed (which depend on the solids moisture content) would be smaller — a fact which would explain the lower values of the tensile strength for this simulation.
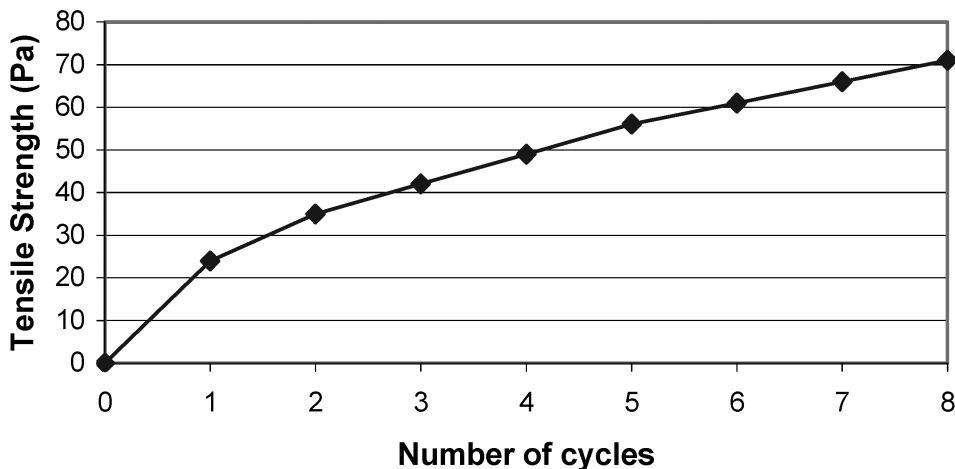
Figure 10. Tensile strength of caked sugar during repeated 24-h wetting/drying cycles. Both environmental temperature and relative humidity vary periodically with time.

6. CONCLUSIONS

In this paper, a numerical model for the mechanism which causes moisture migration caking in hygroscopic powders was introduced, which is based on the formation and growth of solid bridges between neighboring particles. Moisture migration is modeled through the solution of heat and mass transfer equations, implemented with appropriate source terms to represent the moisture uptake/loss by the powder. The analysis performed for the derivation of the equations which describe the behavior of a hygroscopic material at equilibrium is generic and can be easily extended to a range of materials. Sorption isotherms can be obtained via experimental analysis. The process of moisture migration led to formation of liquid bridges (during wetting) which hardened (during drying) and were transformed into solid bridges, thus causing the material to cake. Although the equations for heat and mass transfer were given in one-dimension, the model can easily be extended in two- or three-dimensions.

This study concentrated on granulated sugar and predictions of the tensile strength of the caked material were obtained and compared against experimentally measured values. The model was capable of predicting accurately both quantitatively and qualitatively the moisture migration in the domain and the tensile strength of the caked sugar under varying environmental conditions. The numerical model was then extensively tested under different scenarios and the predictions were consistent with the theoretically expected material behavior. The presented work, which combines heat and mass transfer with moisture migration caking in a continuum mechanics framework, is believed to be unique. Further work is under way to test the response of the model in two- and three-dimensions, and study the behavior of other powders such as laundry detergents, agricultural fertilizers and

pharmaceutical products stored in boxes and hoppers, where interactions with the external environment might lead to moisture migration caking.

REFERENCES

1. T. Tanaka, Evaluation the caking strength of powders, *Ind. Eng. Chem. Prod. Res. Dev.* **17**, 241–246 (1978).
2. T. P. Labuza and C. R. Hyman, Moisture migration and control in multi-domain foods, *Trends Food Sci. Technol.* **9**, 47–55 (1998).
3. K. Rastikian and R. Capart, Mathematical model of sugar dehydration during storage in a laboratory silo, *J. Food Eng.* **35**, 419–431 (1998).
4. M. S. A. Bradley, R. J. Farnish, A. N. Pittman and M. C. Leaper, Caking by moisture migration-an investigation of cuses, *Trans. I. Mech. E* **3**, 39–46 (2000).
5. J. M. Aguilera, J. M. del Valle and M. Karel, Caking phenomena in amorphous food powders, *Trends Food Sci. Technol.* **6**, 149–155 (1995).
6. M. Peleg, Flowability of food powders and methods for its evaluation — a review, *J. Food Process Eng.* **1**, 303–328 (1977).
7. G. I. Tardos, M. I. Kham and P. R. Mort, Critical parameters and limiting conditions in binder granulation of fine powders, *Powder Technol.* **94**, 245–258 (1997).
8. M. C. Leaper, M. S. A. Bradley, J. A. S. Cleaver, I. Bridle, A. R. Reed, H. Abou-Chakra and U. Tüzün, Constructing an engineering model for moisture migration in bulk solids as a prelude to predicting moisture migration caking, *Advanced Powder Technol.* **13**, 414–424 (2002).
9. J. Wang, N. Christakis, M. K. Patel, M. C. Leaper and M. Cross, A computational model of coupled heat and moisture transfer with phase change in granular sugar during varying environmental conditions, *Numer. Heat Transf. A* **45**, 751–776 (2004).
10. M. C. Leaper, M. S. A. Bradley, I. Bridle, A. R. Reed, H. Abou-Chakra and U. Tüzün, Measuring the tensile strength of caked sugar produced from humidity cycling, *Proc. I. Mech. E* **217(E1)**, 41–47 (2003).
11. N. Christakis, Modelling of the microphysical processes that lead to warm rain formation, PhD Thesis, UMIST, Manchester (1998).
12. J. E. Warren and P. J. Root, The behaviour of naturally fractured reservoirs, *Soc. Petrol. Eng. J.* **3**, 245–255 (1963).
13. R. W. Zimmerman, T. Hadgu and G. S. Bodvarsson, A new lumped-parameter model for flow in unsaturated dual-porosity media, *Adv. Water Resourc.* **19**, 317–327 (1996).
14. R. T. Toledo, *Fundamentals of Food Process Engineering*, 2nd edn. Van Nostrand Reinhold, New York (1991).
15. S. V. Patankar, *Numerical Heat Transfer and Fluid Flow*, 1st edn. Hemisphere, New York (1980).
16. M. Cross, Computational issues in the modelling of materials based manufacturing processes, *J. Comp. Aided Mater. Des.* **3**, 100–116 (1996).