

DRYING OF CERAMIC MATERIALS: NUMERICAL AND EXPERIMENTAL RESEARCH

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Abstract. *This work presents an experimental and mathematical research of ceramics materials. The three-dimensional mathematical modeling assumes to be constant thermo-physical properties and convective boundary conditions at the surface of the solids. The governing equations are discretized using finite-volume method and fully implicit formulation. The experimental tests were done using two clay materials in the production of red ceramics and white ceramic (ball clay). It was used different dimensions and initial moisture content of the material and temperature and relative humidity of air-drying. Numerical and experimental results of the drying kinetics of the samples are shown and analyzed. The diffusion and convective mass transfer coefficients were obtained by comparison between predicted and experimental data using the least square error technique, and good agreement was obtained.*

Keywords. Drying, Numerical, experimental, clays, parallelepiped.

1. Introduction

The drying is a simultaneous phenomenon of heat and mass and shrinkage. Being like this, a realistic approach of the physical-mathematical model for the drying process of a solid, it can be influenced by internal and external conditions and mechanism of moisture migration inside of the material. In this sense, the development of mathematical models to describe the ceramic drying process has been the topic of many research studies in the world, but a major question that still remains is the measurement of the parameters used in models how drying rate and transport properties (Mujumdar, 1995).

According to Itaya and Hasatani (1996), in the case of argils, to promote an understanding of the drying mechanism, a microscopic investigation in the state of the moisture in the product it is requested. In this sense, some authors, such like Elias (1995), Fricke (1981), Ketelaars et all (1992a), Keteraals et all (1992b) and Hasatani and Itaya (1992), Nascimento (2002), Sander et all (2003) reported that during the clay drying, the dominant mechanism of moisture migration is the liquid diffusion transport. However, van der Zanden et all (1996) and van der Zanden (1997), they consider that exist transport of liquid and vapor inside the solid. These authors shown in its works that the vapor concentration is directly proportional to the porosity, in agreement with the experiences of Pukall reported by Medeiros (1977).

In this sense, the goal of this work has is to present a three-dimensional study of the mass transfer in parallelepiped solids and to apply the methodology to the drying of ceramic materials.

2. Mathematical modeling

2.1. Mass transfer model

For simplify the model, the following considerations are adopted:

- the thermo-physical properties are constant, during all the diffusion process;
- the solid is homogeneous and isotropic;
- the moisture content distribution is uniform in the beginning of the process;
- the phenomenon happens under convective condition at the surface of the body, with moisture content dependent of the position and of the time
- the mass transfer coefficient is assumed to be constant in all the process.
- the solid is composed of water in liquid phase and solid material.

Figure (1) illustrates a solid parallelepiped of dimensions $2R_1$x $2R_2$x$2R_3$. For this case, the general differential equation that describes the diffusion phenomenon it is in the way:

$$\frac{\partial M}{\partial t} = \nabla.(D\nabla M) \tag{1}$$

where M is local moisture content (kg/kg), D is diffusion coefficient (m^2/s) and t is the time (s).

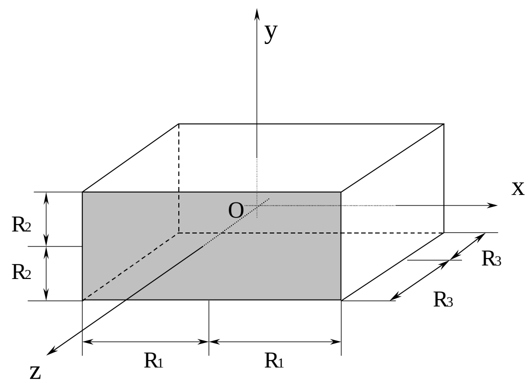

Figure 1. Geometrical configuration of the physical problem.

Due to the symmetry in the solid, in particular, in the planes (x=0,y,z), (x,y=0,z), (x,y,z=0) we consider 1/8 of the volume of the solid. The initial, symmetry and boundary conditions are as follows:

- Initial condition:

$$M(x, y, z, t = 0) = M_o \tag{1a}$$

where M_o is initial moisture content (kg/kg).

- Symmetry condition:

$$\frac{\partial M(x=0,y,z,t)}{\partial x} = \frac{\partial M(x,y=0,z,t)}{\partial y} = \frac{\partial M(x,y,z=0,t)}{\partial z} = 0,\ t>0 \tag{1b}$$

- Boundary condition:

$$-D\frac{\partial M(x, y, z, t)}{\partial x} = h_m(M(x, y, z, t) - M_e) \quad \text{in } t>0 \text{ e } x= R_1 \tag{1c}$$

$$-D\frac{\partial M(x, y, z, t)}{\partial y} = h_m(M(x, y, z, t) - M_e) \quad \text{in } t>0 \text{ e } y= R_2 \tag{1d}$$

$$-D\frac{\partial M(x, y, z, t)}{\partial z} == h_m(M(x, y, z, t) - M_e) \quad \text{in } t>0 \text{ e } z= R_3 \tag{1e}$$

where h_m is convective mass transfer coefficient (m/s).

In order to dimensionless the equation, were adopted the following variables dimensionless:

$$M^* = \frac{M(x,y,z,t) - M_e}{M_o - M_e}\ ,\ x^* = \frac{x}{R}\ ;\ y^* = \frac{y}{R}\ ;\ z^* = \frac{z}{R}\ ;\ t_m^* = \frac{Dt}{R^2}\ ;\ V^* = \frac{V}{R^3} \qquad \text{(2a-f)}$$

where $R = \sqrt{R_1^2 + R_2^2 + R_3^2}$, it represent the diagonal major of the parallelepiped in study (1/8 of the volume presented in the Figure 3.1) and it t_m^* perform the number Fourier for mass transfer.

This way the transient three-dimensional general equation in the form dimensionless can be written:

$$\frac{\partial M^*}{\partial t_m^*} = \nabla^2 M^* \qquad (3)$$

In terms dimensionless the initial and simmetry conditions can be written as follows:

$$M^*(x,y,z,0) = 1\ ;$$
$$-\frac{\partial M^*(x,y,z,t)}{\partial x}\Big|_{x^*=0} = -\frac{\partial M^*(x,y,z,t)}{\partial y}\Big|_{y^*=0} = -\frac{\partial M^*(x,y,z,t)}{\partial z}\Big|_{z^*=0} = 0 \qquad \text{(4a-d)}$$

with the boundary conditions in the following way:

$$-\frac{\partial M^*}{\partial x^*}\Big|_e = \frac{h_m R}{D} M^*;\ \text{ for the x surface.} \qquad \text{(5a)}$$

$$-\frac{\partial M^*}{\partial y^*}\Big|_n = \frac{h_m R}{D} M^*;\ \text{ for the y surface.} \qquad \text{(5b)}$$

$$-\frac{\partial M^*}{\partial z^*}\Big|_f = \frac{h_m R}{D} M^*;\ \text{ for the z surface.} \qquad \text{(5c)}$$

The average moisture content was calculated by (Whitaker, 1980):

$$\overline{M}^* = \frac{1}{V^*} \int_{V^*} M^* dV^* \qquad (6)$$

where $\overline{M}^*$ is dimensionless average moisture content (kg/kg).

3. Experimental Methodology

Two ceramics bricks (ball clay and red ceramic type) were moulded under pressure on the parallelepiped shape under pressing pressure of 2.5 MPa. The bricks was dried by placing then in a oven in specified temperature and relative humidity of the air. The first step of experiment was to weight the sample and to measure the length one in intervals of 10 min during drying process. The mass of the sample was obtained by one electronic balance with accuracy of ±0.01g and the dimensions of the samples were measured using a digital pachimeter. The Table (1) presents all the experimental conditions used in the work. Were: RH is Relative humidity (%), T is air temperature (^{o}C), $2R_{1,}$ $2R_1$ and $2R_3$ are sizes of the samples (m) and v is air velocity (m/s).

In the end of the drying process, the samples were placed in other over under temperature of 100ºC to obtain the equilibrium moisture content.

Table 1. Air and bricks ceramics experimental conditions used in this work

Test	Air			Bricks ceramics					Time
	T (°C)	RH (%)	v (m/s)	M_o (d.b.)	M_e (d.b.)	$2R_1$ (m)	$2R_2$ (m)	$2R_3$ (m)	(s)
1	60.0	10.10	0.1	0.1000	0.00173	0.06045	0.00706	0.02054	16200
2	60.0	10.10	0.1	0.0820	0.00163	0.06064	0.00755	0.02053	13788
3	80.0	4.66	0.1	0.0765	0.00084	0.06081	0.00539	0.02043	3600
4	80.0	5.00	0.1	0.2710	0.009024	0.06026	0.006055	0.02056	11880
5	110.0	2.30	≈ 0.0	0.2270	0.009024	0.06025	0.009056	0.02031	13788
6	110.0	2.20	≈ 0.0	0.2270	0.001810	0.06066	0.005011	0.02048	3600

4. Numerical Solution

In this work we use the finite-volume method to discretize the governing equations. The Figure (2) represents the differential volume of the physical domain, where the nodal points (W, E, N, S, F, T), dimensions and length of the control volume are presented.

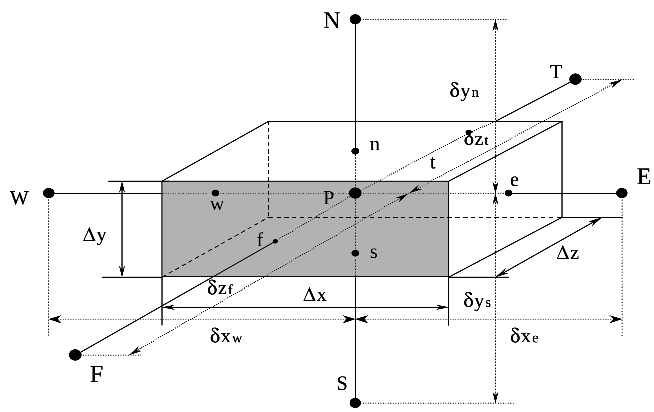

Figure 2 – Control-volume used in this work

Assuming implicit fully formulation, where all terms of are estimated in t+Δt, the Eq. (3) was integrated in the control volume of the Fig. (2) (that correspond to the internal points of the domain), and also in the time, (Maliska, 1995; Patankar, 1980). As results the equation (1) can be written in the linear form as:

$$A_P M^*_P = A_E M^*_E + A_W M^*_W + A_N M^*_N + A_S M^*_S + A_T M^*_T + A_F M^*_F + B \tag{7}$$

with:

$$A_E = \frac{\Delta y^* \Delta z^*}{(\delta x)^*_e} \qquad A_W = \frac{\Delta y^* \Delta z^*}{(\delta x)^*_w} \qquad A_N = \frac{\Delta z^* \Delta x^*}{(\delta y)^*_n} \qquad A_S = \frac{\Delta z^* \Delta x^*}{(\delta y)^*_s}$$

$$A_T = \frac{\Delta x^* \Delta y^*}{(\delta z)_t^*} \qquad A_F = \frac{\Delta x^* \Delta y^*}{(\delta z)_f^*} \qquad A_P^0 = \frac{\Delta x^* \Delta y^* \Delta z^*}{\Delta t_m^*} \qquad B = A_P^0 M^{*0}_P$$

$$A_P = \sum A_K + A_P^0 + \overline{SM}$$

$$\overline{SM} = \begin{cases} 0 & \text{for nodal internal points} \\ \dfrac{\Delta y^* \Delta z^*}{\left(\dfrac{1}{B_{im}} + \delta x_e^*\right)} & \text{for the x surface} \end{cases}$$

$$\overline{SM} = \begin{cases} 0 & \text{for nodal internal points} \\ \dfrac{\Delta x^* \Delta z^*}{\left(\dfrac{1}{Bi_m} + \delta y_n^*\right)} & \text{for the y surface} \end{cases}$$

$$\overline{SM} = \begin{cases} 0 & \text{for nodal internal points} \\ \dfrac{\Delta x^* \Delta y^*}{\left(\dfrac{1}{Bi_m} + \delta z_f^*\right)} & \text{for the z surface} \end{cases}$$

The $A_{E,}$ $A_{N,}$ A_F are zero in boundary nodes and the source term $\overline{SM}$ is associated to the convective mass transfer in the surface of the solid.

The set of equations are solved interactively using the Gauss-Seidel method. The following convergence criterion was used:

$$| M^{*^{n+1}} - M^{*^{n}} | \leq 10^{-8} \tag{8}$$

where n represents the n-th iteration in each time. More details can be encountered in Nascimento et all. (2001a-b) and Nascimento (2002).

The diffusion and mass transfer coefficients were found by varying the D and h_m to minimize the sum of squared deviations between the experimental and predicted data. The relative deviation between experimental and calculated values (relative residuals, ERMQ) and the variance (S^2) are defined as follows:

$$ERMQ = \sum_{i=1}^{m} \left(\overline{M}^*_{i,Num} - \overline{M}^*_{i,Exp}\right)^2 ; \qquad S^2 = \frac{ERMQ}{(m - \hat{n})} \tag{9a-b}$$

where m is the number of experimental points and $\hat{n}$ is the parameters number fitted Figliola and Beasley (1995). The smallest values of ERMQ and S^2 were used as a criterion to obtain the best value of the diffusion and mass transfer coefficients

5. Results and discussions

In order to test the formulation presented in this work, different numerical results of the average moisture content of a ceramics brick along the drying process are compared with experimental results. To obtain the numerical results it was implemented a computational code using the grid 20 x 20 x 20 points and $\Delta t = 1s$. These conditions were obtained by grid and time refines study. Other information about this procedure can be found in the literature (Nascimento et all 2001b, Nascimento, 2002). As the results were obtained of form dimensionless, these possess independence of M_o and $M_{eq.}$

5.1. Moisture content of the ceramics brick

The transport coefficients as well as the variance obtained in the experiments are shown in Tab. (2). The small variances indicate that the model presents good agreement with the experimental data.

Figures (3-8) illustrate the comparison between numerical and experimental data of the average moisture content obtained during the drying. It can be seen in these figures that a good agreement was obtained. The transport coefficients as well as the variance obtained in the experiments are shown in Tab. (2). The small variances indicates that the model presents good agreement with the experimental data

The mass diffusion coefficients were obtained by agreement and increased with the levels of drying temperature, as expected. In relation to the mass transfer coefficients it is observed that it increases with the increase of the drying temperature. However, to 80 ºC and test 3, a highest value this coefficient is observed in comparison with those obtained in 110 ºC, this fact is explained by fact of the experiment have been realized under forced convection, while the tests to the 110 ºC was accomplished under natural convection conditions. Besides the dimensions of the solids are very different, a relationship different area/volume is obtained. This relationship modify the drying rate of the solid.

In a general way, the increase of the convective of mass transfer coefficient with the elevation of the air temperature, present indicate that the shrinkage influences the mass transfer modifying the external structure of the material, in other words, the roughness of the surface.

Table 2. Transport coefficients respect and error and variance, for each drying test.

Test	T (^oC)	D. 10^{+8} (m^2/s)	$h_m.10^{+6}$ (m/s)	ERMQ $(kg/kg)^2$	$\bar{s}^2 .10^{+4}$ %kg/kg
1	60.0	0.231	1.730	0.0013	0.023
2	60.0	0.110	0.258	0.0084	0.130
3	80.0	0.351	4.310	0.0039	0.130
4	80.0	1.086	1.510	0.0036	0.310
5	110.0	1.520	2.000	0.0171	0.580
6	110.0	2.040	2.670	0.0094	0.390

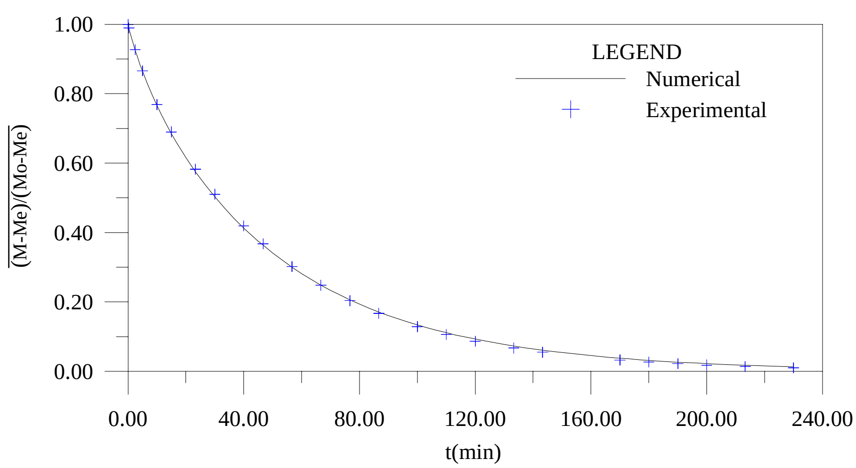

Figure 3. Comparison between predicted and experimental dimensionless mean moisture content during the drying of red ceramics brick.Test 1.

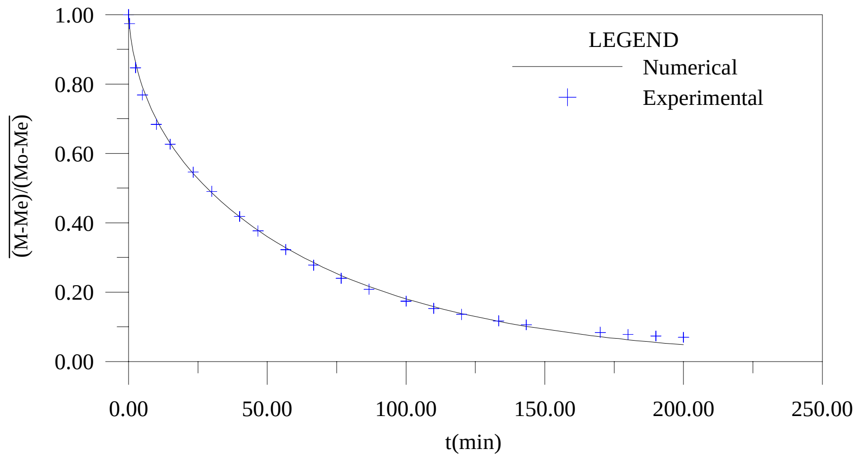

Figure 4. Comparison between predicted and experimental dimensionless mean moisture content during the drying of red ceramics brick. Test 2.

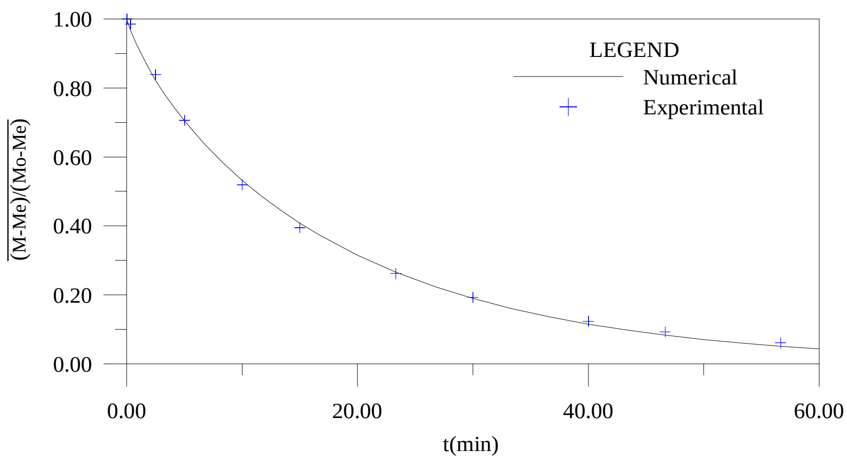

Figure 5. Comparison between predicted and experimental dimensionless mean moisture content during the drying of Ball-Clay ceramics brick. Test 3.

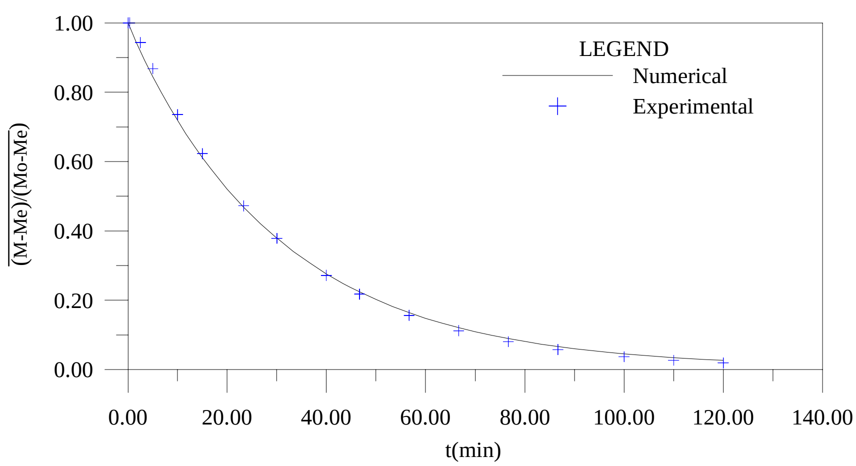

Figure 6. Comparison between predicted and experimental dimensionless mean moisture content during the drying of red ceramics brick. Test 4.

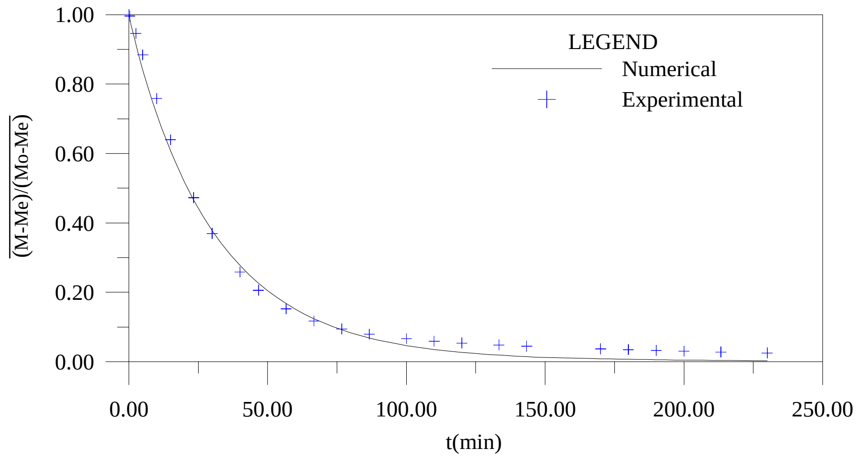

Figure 7. Comparison between predicted and experimental dimensionless mean moisture content during the drying of red ceramics brick. Test 5.

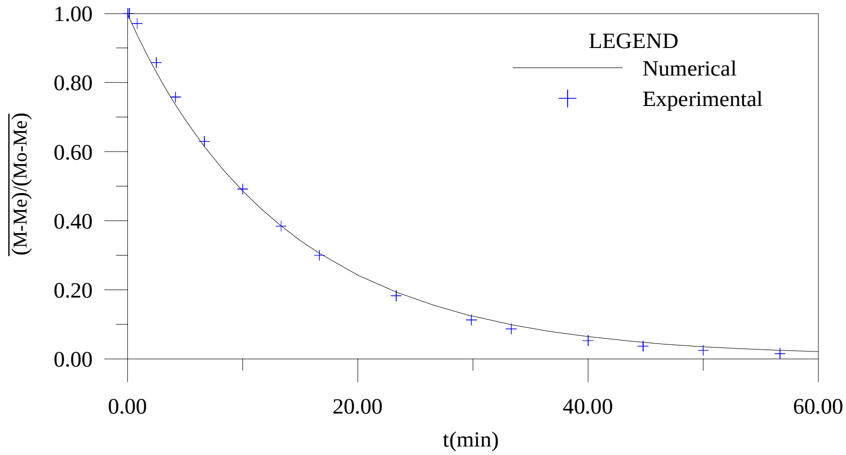

Figure 8. Comparison between predicted and experimental dimensionless mean moisture content during the drying of Ball-Clay ceramics brick. Test 6.

It is important to note that the of mass diffusion and convective mass transfer coefficients obtained using a model that considere neglected the shrinkage effect, are effective values, once they incorporate these and other phenomena that occurs during drying process.

As a comment, given the amount of information supplied, in this work the good agreement obtained in the comparisons and the physical realism present, it can be affirmed that the model and the solution methodology for the problem are satisfactory.

The model is very versatile and the used technique has great potential, being able to be used to describe diffusion processes such as drying, wetting, heating and cooling of solids with geometry that changes of a one-dimensional rod to a parallelepiped, besides plate planes, without restrictions as the nature of the material (fruits, cereals, vegetables, minerals, etc.).

6. Conclusions

Of the analysis of the results obtained it can be concluded that: a) the finite-volume method is a appropriate technique to solve drying problems; b) as larger the time smaller will be the value of the average moisture content, in any point inside the solid; g) the results of the study gave a base of values of transport coefficient of ceramics brick during drying process; h) in this work all equations can be used in many physical problems such as heating, cooling, wetting and drying no problems.

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