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PERIODIC MODES IN AN ISOTHERMAL TUBULAR REACTOR

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Nonstationary periodic conditions in an isothermal tubular reactor are considered. A study is made of the effects of the amplitude, frequency, and waveform of the input concentration signals on the average throughput.

A reactor with nonlinear kinetics is often better operated in nonstationary cyclic mode than in a stationary one. This was first pointed out in [1, 2], and then in [3] variational methods were used to formulate the optimization conditions. Since then, there have been very extensive studies on periodic modes of various types: for example, in [4] a study was made of the quasistationary state, while in [5] positive feedback was considered, in [6, 7] the study concerned systems was distributed parameters, in [8] periodic relaxation oscillations were examined, and [9] dealt with adiabatic reactors. In [10], very effective estimates were obtained by means of the π criterion, while in [11] differential inequalities were used for similar purposes. The results and problems in this area have been surveyed in [12, 13].

The published data relate primarily to completely mixed systems, or sometimes to models for ideal displacement, but very little is known about models characterizing intermediate mixing states. Here we may mention [7], in which an axial dispersion model was used to examine isothermal tubular reactors for the case of a second-order reaction, but only for a sinusoidal input signal and for parameters varying over a very narrow range.

Here we present some more general results for isothermal reactors, which can be described by means of an axial dispersion model. We consider irreversible reactions of order n and give data for limiting cases of mixing. Also, the results obtained previously for second-order reactions are extended to the effects of the frequency of the periodic input signals on the throughput, and we discuss the determination of the optimum period for sinusoidal inputs. The calculations were performed with an analog computer using perturbation methods.

Formulation. Consider an isothermal tubular reactor in which there is an irreversible reaction of order n, which is described mathematically by an axial dispersion model:

$$\frac{\partial c(x,\ t)}{\partial t} = L_a^n c(x,\ t),\ x \in (0,\ 1),\ t > 0; \tag{1}$$

$$L_a^n = \frac{1}{\mathrm{Pe}} \frac{\partial^2(\cdot)}{\partial x^2} - \frac{\partial(\cdot)}{\partial x} - R\tau(\cdot)^n \tag{1a}$$

with the boundary conditions

$$c(x,\ t) - \frac{1}{\mathrm{Pe}} \frac{\partial c(x,\ t)}{\partial x} = u(t),\ x = 0,\ t > 0; \tag{2a}$$

$$\frac{\partial c(x,\ t)}{\partial x} = 0,\ x = 1,\ t > 0, \tag{2b}$$

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and the initial condition

$$c(x,\ 0) = 0. \tag{3}$$

We assume that the function is periodic with period T > 0, i.e., u(t) = u(t + T), $t \geqslant 0$, while c_s is the stationary solution to (1)-(3) when u_s is constant:

$$L_a^n c_s(x) = 0, \quad x \in (0,\ 1); \tag{4}$$

$$u_s = c_s(x) - \frac{1}{\mathrm{Pe}} \frac{dc_s(x)}{dx}, \quad x = 0; \tag{5a}$$

$$\frac{dc_s(x)}{dx} = 0, \ x = 1. \tag{5b}$$

Then the treatment for the periodic state can be formulated as follows: let the time average of the periodic function u be equal to the constant u_s. We compare the time average for the function c in the steady state with the value for c_s taken at the same point. If it is less, the periodic state gives higher conversion, and therefore under otherwise equal conditions may be better than the corresponding stationary state. We then have to consider the character of the periodic functions giving this result and which of these functions is the best. We first consider the quasistationary state.

Quasistationary State with Reaction of Order n. If there are slow oscillations at the input (period greatly exceeding the mean residence time or time constant of the reactor), in which case the reaction dynamics have only a small impact, then we have a quasistationary state. Then the reactor is considered to operate by passing to working points with different stationary states no matter what the dynamic effects, i.e., the response is completely determined by the static characteristic. This means that if A_s is the mapping $u_s \to c_s$ defined by (4) and (5), the response of the reactor at time t is given by the following equation for some function slowly varying in time:

$$c_q(t) = A_s(u(t)). \tag{6}$$

As the mapping $c_s \to c_s^n$, n > 1, is convex, the mapping A_s is concave and monotonically increasing [11]. According to the inequality of [14] applicable to concave functions, we get that

$$\overline{A_s(u(t))} \leqslant A_s(\overline{u(t)}), \tag{7}$$

where the integral

$$\overline{u(t)} = \frac{1}{T} \int_0^T u(t)\, dt \tag{8}$$

denotes the value averaged over time.

Then if $|u_s| = \overline{u(t)}$ we get from (7) that

$$\overline{A_s(u(t))} \leqslant A_s(u_s), \tag{9}$$

i.e., slowly propagating oscillations usually raise the throughput of a tubular reactor as described by the model of (1)-(3) and never reduce it.

Limiting Cases of Mixing with a Reaction of Order n. We now consider the problem of (1)-(3) in the limiting cases Pe → 0 and Pe → ∞, which reflect the corresponding cases of limiting mixing in the reactor. Then in addition to the above qualitative arguments we apply the method of approximate solution given in [2] to derive quantitative estimates.

Case Pe = 0. The reactor is completely mixed. If we write the input function in the form $u(t) = u_s + u'(t)$, where $\overline{u'(t)} = 0$, then (1)-(3) become a problem with the initial value

$$\frac{dz(t)}{dt} + z(t) + \alpha z^n(t) = \delta u(t) + 1, \ z(0) = 0, \tag{10}$$

where

$$z = c/u_s, \ \delta u = u'/u_s, \ \alpha = R\tau u_s^{n-1}. \tag{11}$$

We represent z as the sum of the stationary solution z_s and the deviation y from it ($z = z_s + y$) and substitute this into (10). Then we get the differential equation

$$\frac{dy(t)}{dt} + (1+\beta)\,y(t) = \delta u(t) - \alpha \sum_{k=2}^{n} a_k y^k(t), \tag{12}$$

where

$$a_k = \frac{n! z_s^{n-k}}{k!\,(n-k)!}, \quad \beta = u\alpha z_s^{n-1}.$$

We introduce the artificial parameter μ with the nonlinear term on the left. The solution to (12) is sought as

$$y = y_0 + \mu y_1 + \mu^2 y_2 + \cdots \tag{13}$$

We substitute (13) into (12) with parameter μ and equate the coefficients to identical powers of μ on the left and right in (12) up to terms of the second degree to get a system of equations:

$$\frac{dy_0(t)}{dt} + (1+\beta)\,y_0(t) = \delta u(t); \tag{14}$$

$$\frac{dy_1(t)}{dt} + (1+\beta)\,y_1(t) = -\alpha \sum_{k=2}^{n} a_k y_0^k(t); \tag{15}$$

$$\frac{dy_2(t)}{dt} + (1+\beta)\,y_2(t) = -\alpha \sum_{k=2}^{n} k a_k y_0^{k-1}(t)\,y_1(t). \tag{16}$$

If $\delta u(t) = A \sin \omega t$, then

$$y_0(t) = B \sin(\omega t - \varphi), \tag{17}$$

where

$$B = \frac{A}{\sqrt{(1+\beta)^2+\omega^2}}; \quad \varphi = \operatorname{Arc\,tg}\frac{\omega}{1+\beta}; \quad \overline{y_0(t)} = 0.$$

The expressions for approximations above the zeroth order are very complicated, but we are interested only in the time means. For example,

$$\overline{y_1(t)} = -\frac{\alpha}{1+\beta} \sum_{j=1}^{[n/2]} \binom{n}{2j} \binom{2j}{j} z_s^{n-2j} \frac{B^{2j}}{2^{2j}}; \tag{18}$$

$$\overline{y_2(t)} = -\frac{\alpha! \overline{y_1(t)}}{1+\beta} \sum_{j=2}^{[n/2]} (2j-1) \binom{n}{2j-1} \binom{2j-2}{j-1} z_s^{n-2j+1} \frac{B^{2j-2}}{2^{2j-2}}. \tag{19}$$

The mean solution in the case μ = 1 takes the form

$$\overline{z(t)} \sim z_s + y_1(t) + y_2(t) \tag{20}$$

and according to (18) and (19) is less than z_s. Note that for n = 2 (18) coincides with the solution obtained in [2].

Case Pe = ∞. The reactor is an ideal-displacement system, and the model is described in the above symbols by the following boundary-value problem:

$$-\frac{\partial z(x,t)}{\partial x} - \alpha z^n(t) = \frac{\partial z(x,t)}{\partial t}; \tag{21}$$

$$z(0,\ t) = 1 + \delta u(t); \tag{22}$$

$$z(x,\ 0) = 0. \tag{23}$$

If the solution is sought in the same way as for an ideal-mixing reactor, the first-order approximation for the mean is

$$\overline{z(x,\ t)} \sim z_s(x) - \frac{1-e^{-\beta x}}{\beta} \alpha \sum_{j=1}^{[n/2]} \binom{n}{2j} \binom{2j}{j} z_s^{n-2j} \frac{c^{2j}}{2j}, \tag{24}$$

where $c(x) = Ae^{-\beta x}$.

Here it is clear that the reduction measure is independent of frequency and is a function only of the amplitude.

Axial Dispersion Model with Second-Order Reaction. We consider the model of (1)-(3) for the case n = 2. The boundary-value problem for the deviation from the stationary solution takes the form

$$\frac{\partial y(x,t)}{\partial t} = L_y^1 y(x,t) - \alpha_2 y^2(x,t),\ x\in(0,1),\ t>0; \tag{25}$$

$$L_y^1 = \frac{1}{\mathrm{Pe}}\frac{\partial^2(\cdot)}{\partial x^2} - \frac{\partial(\cdot)}{\partial x} - \beta_2(\cdot); \tag{25a}$$

$$\delta u(t) = y(x,t) - \frac{1}{\mathrm{Pe}}\frac{\partial y(x,t)}{\partial x},\quad x=0; \tag{26a}$$

$$\frac{\partial y(x,t)}{\partial x} = 0,\quad x=1, \tag{26b}$$

with the initial condition y(x, 0) = 0, where $\alpha_2 = R\tau u_s$ and $\beta_2 = 2\alpha_2 z_s$.

In the way described above, we use parameter μ from the approximations of zeroth and first orders to get the following boundary-value problems:

$$\frac{\partial y_0(x,t)}{\partial t} = L_y^1 y_0(x,t),\ x\in(0,1),\ t>0; \tag{27}$$

$$\delta u(t) = y_0(x,t) - \frac{1}{\mathrm{Pe}}\frac{\partial y_0(x,t)}{\partial x},\quad x=0; \tag{28a}$$

$$\frac{\partial y_0(x,t)}{\partial x} = 0,\ x=1; \tag{28b}$$

$$\frac{\partial y_1(x,t)}{\partial t} = L_y^1 y_1(x,t) - \alpha_2 y_0^2(x,t),\ x\in(0,1),\ t>0; \tag{29}$$

$$0 = y_1(x,t) - \frac{1}{\mathrm{Pe}}\frac{\partial y_1(x,t)}{\partial x},\quad x=0; \tag{30a}$$

$$\frac{\partial y_1(x,t)}{\partial x} = 0,\ x=1. \tag{30b}$$

In the case δu(t) = A sin ωt, the solution of (27)-(28) is obtained in the form

$$y_0(x,t) = \sum_{k=j}^{\infty} A_k \sin(\omega t - \varphi_k), \tag{31}$$

where

$$A_k = \frac{2A\exp\left(\frac{\mathrm{Pe}\,x}{2}\right)}{\sqrt{\left(\frac{\gamma_k^2}{\mathrm{Pe}} + \frac{\mathrm{Pe}}{4} + \beta_2\right)^2 + \omega^2}} \cdot \frac{\gamma_k\cos\gamma_k x + \frac{\mathrm{Pe}}{2}\sin\gamma_k x}{\gamma_k^2 + \left(\frac{\mathrm{Pe}}{2}\right)^2 + \mathrm{Pe}}; \tag{32a}$$

$$\mathrm{tg}\,\varphi_k = \frac{\omega}{\frac{\gamma_k^2}{\mathrm{Pe}} + \frac{\mathrm{Pe}}{4} + \beta_2}, \tag{32b}$$

and the quantities γ_k (k = 1, 2, ...) are the positive roots of the transcendental equation

$$\mathrm{tg}\,\gamma_k = \frac{\mathrm{Pe}\,\gamma_k}{\gamma_k^2 - \left(\frac{\mathrm{Pe}}{2}\right)^2}. \tag{33}$$

The mean value of the solution to (29)-(30) is

$$\overline{y_1(x,t)} = -\alpha_2 \sum_{k=1}^{\infty} B_k(x)\left[\sum_{l=1}^{\infty}\sum_{j=1}^{\infty}\int_0^1 \exp\left[-\frac{\mathrm{Pe}}{2}\xi\right] A_l(\xi)A_j(\xi)\left(\gamma_k\cos\gamma_k\xi + \frac{\mathrm{Pe}}{2}\sin\gamma_k\xi\right)d\xi\right], \tag{34}$$

where

$$B_k(x) = \frac{\exp\left(\frac{\mathrm{Pe}}{2}x\right)\left(\gamma_k \cos\gamma_k x + \frac{\mathrm{Pe}}{2}\sin\gamma_k x\right)}{\left(\frac{\gamma_k^2}{\mathrm{Pe}} + \frac{\mathrm{Pe}}{4} + \beta_2\right)\left(\gamma_k^2 + \left(\frac{\mathrm{Pe}}{2}\right)^2 + \mathrm{Pe}\right)}, \quad k = 1, 2, \ldots \tag{35}$$

Expression (34) is too complicated for one to draw any direct conclusion on the changes, and in what follows it will be used for detailed calculations.

Analog Computer Simulation. A detailed study was made of (1)-(3) for the case n = 2 with an analog computer. Periodic input functions with various periods were used.

We examined the following input functions:

$$u(t) = u_s + u_A \sin\omega t; \tag{36}$$

$$u(t) = \begin{cases} 2u_s, & nT \leqslant t < nT + 0.5T, \\ 0, & nT + 0.5T \leqslant t < (n+1)T, \quad n = 0, 1, 2, \ldots; \end{cases} \tag{37}$$

$$u(t) = \frac{2u_s}{T}t, \quad nT \leqslant t < (n+1)T, \quad n = 0, 1, 2, \ldots \tag{38}$$

Equation (1) was approximated on a discrete network in the variable x as follows:

$$\frac{dc_i}{dt} = \frac{1}{\mathrm{Pe}}\frac{c_{i+1} - 2c_i + c_{i-1}}{h^2} - \frac{c_{i+1} - c_{i-1}}{2h} - R\tau c_i^2, \quad i = 1, 2, \ldots, N, \tag{39}$$

where h = 1/N; $c_i(t) = c(ih, t)$, i = 0, 1, ..., N.

The interval [0, 1] was split up into five equal parts (n = 5), which gives us the following system of equations on the basis of the boundary conditions:

$$\frac{dc_1(t)}{dt} = k_1 c_2(t) + k' c_1(t) - R\tau c_1^2(t) + k'' u(t); \tag{40a}$$

$$\vdots$$

$$\frac{dc_i(t)}{dt} = k_1 c_{i+1}(t) - k_3 c_i(t) + k_2 c_{i-1}(t) - R\tau c_i^2(t), \quad i = 2, 3, 4; \tag{40b}$$

$$\frac{dc_5(t)}{dt} = k_3[c_4(t) - c_5(t)] - R\tau c_5^2(t); \tag{40c}$$

$$c_1(0) = \cdots = c_5(0) = 0, \tag{40d}$$

in which the constants are given by

$$k_1 = \frac{25}{\mathrm{Pe}} - 2.5; \quad k_2 = \frac{25}{\mathrm{Pe}} + 2.5; \quad k_3 = \frac{50}{\mathrm{Pe}};$$
$$k' = \frac{k_2}{1 + 5\mathrm{Pe}} - k_3; \quad k'' = \frac{5k_2\mathrm{Pe}}{1 + 5\mathrm{Pe}}. \tag{41}$$

The MEDA analog computer at the Central Laboratory of the Theoretical Principles of Chemical Engineering, Bulgarian Academy of Sciences, was used to solve this problem. Figure 1 shows the solution to (40) for the input functions (36)-(38), where the function c_5 are the output functions (i.e., $c_5 = c_{out}$) for the initial reactant.

Discussion. We examined the dependence of the average conversion on the amplitude and frequency for a sinusoidal input function. The mean conversion increased with the amplitude as shown in Fig. 2. In essence, the same conclusion can be drawn from (34) and in the limiting cases from (24) and (18)-(20).

The effects of frequency are not so pronounced. At frequencies large by comparison with the reciprocal of the mean residence time, periodic concentration oscillations at the input have virtually no effect, since the inertia in the system effectively filters them out. The effect appears as the period of the oscillations increases at first, then again approximates to zero for very slow oscillations; there is therefore an optimum frequency. This is shown

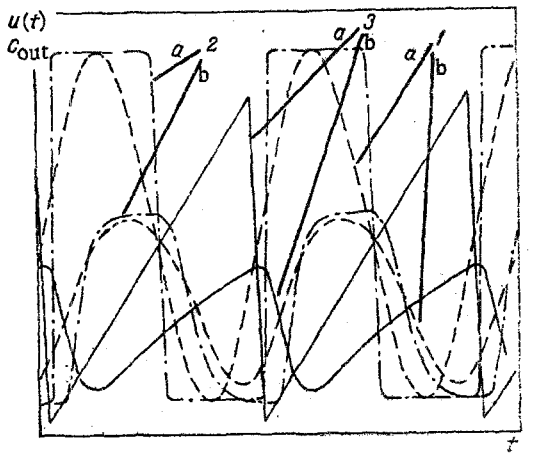

Fig. 1. Solution to (1)-(3) obtained with an analog computer for various input functions in the case n = 2; Pe = 10; $R\tau$ = 10; 1) sinusoidal, u_S = 1, u_A = 0.5, T = 6.285; 2) square wave, u_S = 1, T = 6.285; 3) sawtooth, u_S = 1, T = 6.285; a) input function u(t); b) output c_{out}.

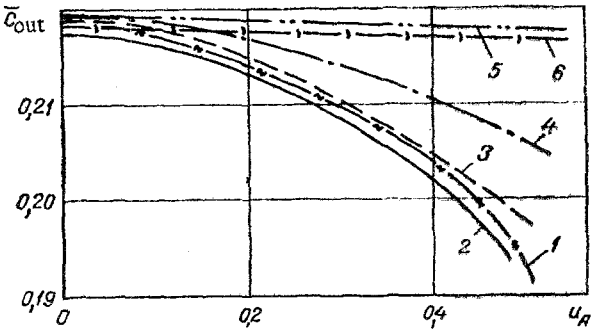

Fig. 2. Effects of amplitude on productivity for a sinusoidal input function with various frequencies (Pe = 1, $R\tau$ = 10, u_S = 1): 1) ω = 0.5; 2) 1; 3) 5; 4) 10; 5) 50; 6) 100.

in Fig. 3, where the individual points up to ω = 0.5 were obtained by solution with the analog computer. At lower frequencies (with given values for the parameters), the processes were too slow by comparison with the period of the analog computer, so the calculations could be performed only with considerable error.

To overcome these difficulties, the calculations were performed with a digital computer by means of (34). We used the 12 first positive roots of (33), which have been given in [15] in tabular form. Curve 2 of Fig. 3 shows the results, which indicates that the approximate solution of (34) gives a good description of the tendencies, but there are considerable differences between the absolute values.

To find more accurate values for frequencies $\omega \leq 0.5$, the boundary-value problem of (1)-(3) was solved numerically in the case n = 2 with the digital computer by quasilinearization [16]. The nonlinear term was linearized by means of a Newton–Raphson formula, while the differentiation operator was approximated as a Frank–Nicholson difference one. The resulting algebraic system with a three-diagonal matrix was solved by Thomas's method [16]. The program was written in Algol-1204 and an Odra-1204 computer was employed. Curve 3 of Fig. 3 shows the results, which supplement the information obtained with the analog machine.

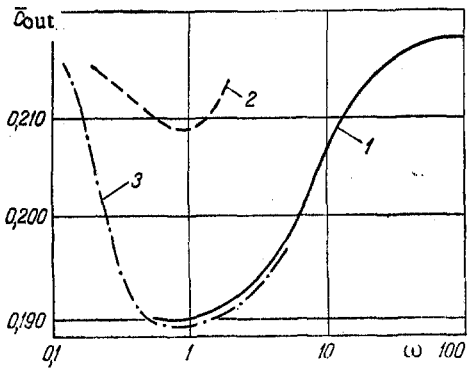

Fig. 3

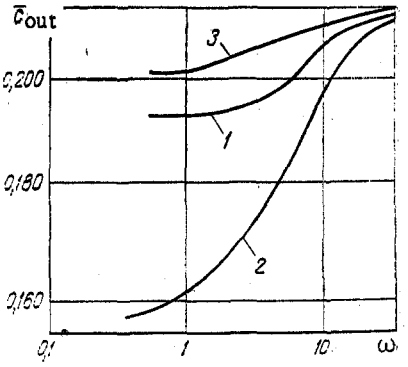

Fig. 4

Fig. 3. Effects of frequency on reactor productivity for a sinusoidal input (Pe = 1, Rτ = 10, u_s = 1, u_A = 0.5): 1) solution obtained with analog computer; 2) approximate solution; 3) numerical solution from digital computer; ω in 1/sec.

Fig. 4. Effects of the input concentration waveform on reactor productivity (Pe = 1, Rτ = 10): 1) sinusoidal, u_s = 1, u_A = 0.5; 2) square wave, u_s = 1; 3) sawtooth, u_s = 1.

The analog computer was used to examine the effects of input-signal shape on the solution, which is equivalent to frequency analysis. Figure 4 shows that the best results were obtained for waves of the rectangular form of (37) at the frequencies examined, followed by the sinusoidal function of (36) and the sawtooth of (38). It should be noted that an input signal of square form is the most suitable for practical realization, although it is impossible to obtain this waveform absolutely exactly because of the inertia.

NOTATION

a_k, constant; A_s, stationary solution operator; A_k, amplitude of (32a) (k = 1, 2, ...); A, B, C, amplitudes; c, concentration function of initial reactant; c_q, quasistationary concentration function; c_i, discrete concentration function; h, step along x; k_1, k_2, k_3, k', k", constants in (40)-(41); L_a^n, L_y^i, differential operators; Pe, Peclet number; R, reaction constant; t, dimensionless time; T, period time; u, input concentration function; u', periodic function with zero mean; u_A, amplitude; x, dimensionless length; y, deviation from the stationary solution; y_i, parameter series functions (i = 1, 2, ...); γ_k, positive roots of (33); φ_k, phase angle of (32b); μ, perturbation parameter; ω, frequency; τ, mean residence time; subscript s, stationary value.

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THERMAL-PARAMETER DETERMINATION FOR THIN-WALLED DRUM-TYPE CRYSTALLIZERS

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A two-dimensional heat-transfer problem has been solved with boundary conditions of the first and third kinds for a rotating hollow thin-walled cylinder. An example of the results in use is given.

Crystallizers of drum type are used in metallurgy [1], in the chemical industry [2], and in the production of ice [3], so calculations on their thermal conditions are of interest.

Studies have been made [4-6] of the thermal fields in a rotating hollow cylinder with boundary conditions of the first kind.

In [4], a thin-walled cylinder was envisaged, with a temperature difference only around the perimeter. In [5], on the other hand, the temperature change along the cylinder director was not incorporated. In [6], the problems were solved with temperature variation along the radial and angular coordinates. The solution was presented in terms of Kelvin functions, which makes for certain difficulties in using it.

If the radius of the cylinder is greater than the wall thickness by a factor of 50 or more, the problem can be treated in Cartesian coordinates, which simplifies it considerably. This formulation may be applied to a two-dimensional plate of finite length with a conjugation condition at the ends.

In [7], the two-dimensional problem was solved for a rectangular plate with a temperature distribution on one of the surfaces varying in a specified fashion with time, while there was zero temperature at the other surfaces and a nonzero initial temperature distribution.

A real crystallizer usually works in a quasistationary state, where the initial temperature distribution is unimportant and the ends of the plate have identical nonzero but unknown temperature distributions over the thickness, while Newton's law applies to the heat transfer at the cooled surface. The problem is formulated mathematically as

$$\frac{1}{a}\frac{\partial\theta}{\partial\tau}=\frac{\partial^2\theta}{\partial x^2}+\frac{\partial^2\theta}{\partial y^2}, \quad \begin{matrix}0\leqslant x\leqslant\delta,\\ 0\leqslant y\leqslant l=2\pi R,\end{matrix} \tag{1}$$

$$\theta(x,\ y,\ 0)=0, \quad \theta(x,\ 0,\ \tau)=\theta(x,\ l,\ \tau), \tag{2}$$

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