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Non-linear mass transfer from a solid spherical particle dissolving in a viscous fluid

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ABSTRACT

Theoretical study of the non-linear mass transfer from a solid particle, suspended in a viscous fluid, is presented. In the cases of intensive mass transfer the processes is completed by the secondary flow as a result of the big concentration gradients and the decrease of the particle radius as a result of the particle dissolution.

1. Introduction

The problems of mass transfer between a solid particle and ambient fluid medium have been intensively studied due to their importance in many chemical and biological processes. The classical general equations modeling mass transport in fluids [1] in the course of these studies are normally simplified and the means of simplification are different for gaseous and liquid medium. For gases, the molar density of the mixture is assumed to depend on pressure and temperature, but not on the concentration. The major part of the works considering diffusion in liquids are based on the assumption of an infinitely diluted solution, which implies that no mechanical properties of the solution, including its density, depends on concentration, while the concentration flux satisfies the first Fick's law. Under this assumption, the velocity field is independent from the concentration, which is determined for a given velocity field from the convection–diffusion equation. The comprehensive review can be found in [2]. In recent papers [3,4] the effect of finite dilution on the mass transfer between a flow and a single inclusion was studied keeping the assumption of a constant density of the mixture. This approach proved to be very effective since it allows the use a known velocity field and it obviously provides a leading order approximation for a solution in the case when the intensity of the flow induced by the mass transfer is small compared to that of an externally imposed flow. However this approach is not applicable in the case of a particle is suspended in the otherwise quiescent fluid and the flow is induced solely by the mass transfer or when the external flow and the natural convection are of comparable intensity. The mass transfer of a solid particle in these situations, which are typical for microgravity conditions and for nearly neutrally buoyant inclusions, is the main subject of the present paper.

The intensive dissolution of spherical particles creates big concentration gradients which induce secondary flow at the liquid–solid phase boundary [5]. As a result the velocity depends from concentration distribution and the convection–diffusion equation becomes non-linear. It leads to the solution of the convection–diffusion equation in conjunction with the hydrodynamic equations. At these conditions the velocity of the spherical particle radius decrease is vastly and must be adding to the fluid velocity at the liquid–solid phase boundary. In these conditions the liquid–solid interphase mass transfer rate will be obtained.

2. Problem formulation

The intensive dissolution problem of spherical particles leads to liquid–solid interphase mass transfer through a mobile phase boundary, where the velocity distribution depends on the secondary flow velocity as a result of the big concentration gradient and the velocity of the solid particle radius decrease. In this case four hierarchical levels of the solution must be used:

1. Linear interphase mass transfer in case of constant radius spherical particle dissolution in immovable fluid.
2. Linear interphase mass transfer in case of inconstant radius (as a result of the particle dissolution) spherical particle dissolution in immovable fluid.
3. Non-linear interphase mass transfer in case of constant radius spherical particle dissolution in movable fluid, when the velocity distribution depends on the secondary flow velocity as a result of the big concentration gradient.
4. Non-linear interphase mass transfer in case of inconstant radius (as a result of the particle dissolution) spherical particle dissolution in movable fluid, when the velocity distribution depends on the secondary flow velocity as a result of the big concentration gradient.

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Nomenclature

u	velocity (m s^{-1})	t	time (s)
u^*	characteristic velocity (m s^{-1})	α	dimensionless parameter
c	concentration (kg m^{-3})	β	expansion parameter
c^*	characteristic concentration (kg m^{-3})	ρ	density (kg m^{-3})
c_0	initial concentration (kg m^{-3})	q	amount of reacted substance (kg s^{-1})
D	diffusivity (m^2 s^{-1})	Q	process efficiency (average mass transfer rate (kg m^{-2} s^{-1})
I	local mass flux (kg m^{-2} s^{-1})	Fo	Fourier number
k	mass transfer coefficient (m s^{-1})	Da	Damkohler number
r_0	radius of the particle (m)	Sh	Sherwood number
l	length scale (m)		

The solution of problem in point 4 is done iteratively and separate iterations are used solutions of points 1–3.

3. Method of solution

The method of problem solution uses the next steps:

1. Linear mass transfer from a solid spherical particle dissolving in a viscous fluid.

1.1. The particle radius is a constant ($r = r_0^{(0)}$).

For a spherical particle the problem satisfy the following equation and boundary conditions:

$$\frac{\partial c}{\partial t} = D\left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r}\right); \qquad t = 0, \quad u = 0; \quad r = r_0^{(0)},$$
$$c = c^*; \quad r \to \infty, \quad c = c_0, \quad c^* > c_0. \tag{1}$$

For the solution of (1) must be used the dimensionless variables:

$$t = t_0 T, \quad r = r_0^{(0)} + lR_0; \qquad u = u_0 U, \quad c = c_0 + (c^* - c_0)C,$$
$$l = \sqrt{Dt_0}. \tag{2}$$

The formulated above problem in new variables takes the form:

$$\frac{\partial C}{\partial T} = \frac{\partial^2 C}{\partial R^2} + \frac{2}{\alpha_0 + R}\frac{\partial C}{\partial R}; \quad \left(\alpha_0 = \frac{r_0^{(0)}}{l}\right), \ T = 0, \ C = 0;$$
$$R = 0, \ C = 1; \quad R \to \infty, \ C = 0 \tag{3}$$

and

$$C = \frac{\alpha_0}{\alpha_0 + R}\operatorname{erfc}\frac{R}{2\sqrt{T}} \tag{4}$$

is an analytical solution of the problem.

1.2. The particle radius decrease ($r = r_0(t)$).

It is means that the particle radius depends from the velocity (v) of the solid particle radius decrease:

$$r_0(t) = r_0^{(0)} + \int_0^t v(t)dt, \quad v = \frac{I}{\rho}, \quad I = -D\left(\frac{\partial c}{\partial r}\right)_{r=r(t)}, \tag{5}$$

where I is a mass transfer rate, ρ is a density of the solid phase, c is the solution of (1).

The dimensionless radius is:

$$\alpha = \alpha(t) = \frac{r_0(t)}{l} = \alpha(t_0 T) = A(T). \tag{6}$$

The dimensionless variables in this case are:

$$t = t_0 T, \quad r = r_0(t) + lR_0; \quad c = c_0 + (c^* - c_0)C_0. \tag{7}$$

In new variables the problem (1) has the form:

$$\frac{\partial C}{\partial T} = \frac{\partial^2 C}{\partial R^2} + \left(A' + \frac{2}{A + R}\right)\frac{\partial C}{\partial R};$$
$$A = \alpha_0 + \frac{c^* - c_0}{\rho}\int_0^T \left(\frac{\partial C}{\partial R}\right)_{R=0} dT,$$
$$A' = \partial A/\partial T = \frac{c^* - c_0}{\rho}\left(\frac{\partial C}{\partial R}\right)_{R=0};$$
$$T = 0, \ C = 0; \quad R = 0, \ C = 1; \quad R \to \infty, \ C = 0. \tag{8}$$

The solution of (8) uses the next iterative approach:

$$\frac{\partial C_i}{\partial T} = \frac{\partial^2 C_i}{\partial R^2} + \left(A'_{i-1} + \frac{2}{A_{i-1} + R}\right)\frac{\partial C_i}{\partial R};$$
$$T = 0, \ C_i = 0; \quad R = 0, \ C_i = 1; \quad R \to \infty, \ C_i = 0;$$
$$A_i = \alpha_0 + \frac{c^* - c_0}{\rho}\int_0^T \left(\frac{\partial C_i}{\partial R}\right)_{R=0} dT,$$
$$A'_i = \frac{c^* - c_0}{\rho}\left(\frac{\partial C_i}{\partial R}\right)_{R=0},$$
$$A_0 = \alpha_0 \frac{c^* - c_0}{\rho}\left(\frac{T}{\alpha_0} + \frac{2\sqrt{T}}{\sqrt{\pi}}\right),$$
$$A'_0 = \frac{c^* - c_0}{\rho}\left(\frac{1}{\alpha_0} + \frac{1}{\sqrt{\pi T}}\right), \tag{9}$$

with iteration stop criterion $\varepsilon < 10^{-3}$:

$$\varepsilon = \frac{\int_0^1 \left[\left(\frac{\partial C_i}{\partial R}\right)_{R=0} - \left(\frac{\partial C_{i-1}}{\partial R}\right)_{R=0}\right]^2 dT}{\int_0^1 \left[\left(\frac{\partial C_i}{\partial R}\right)_{R=0}\right]^2 dT}. \tag{10}$$

In Figs. 1 and 2 are shown concentration distributions (the solutions of (8)) as a function of T and R.

2. Non-linear mass transfer from a solid spherical particle dissolving in a viscous fluid.

2.1. The spherical particle radius is a constant ($r = r_0^{(0)}$).

The velocity of the secondary flow has the form [5]:

$$u(r_0^{(0)}, t) = -\frac{D}{\rho_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}}, \tag{11}$$

where $\rho^* = \rho_0^* + c^*$ is a density of the solution at the particle interface, c^* is a interface concentration. The Navier–Stokes equation and convection–diffusion equation for determining of concentration and velocity profile are used:

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial r} = \upsilon\left(\frac{\partial^2 u}{\partial r^2} + \frac{2}{r}\frac{\partial u}{\partial r} - \frac{2u}{r^2}\right);$$
$$\frac{\partial c}{\partial t} + u\frac{\partial c}{\partial r} = D\left(\frac{\partial^2 c}{\partial r^2} + \frac{2}{r}\frac{\partial c}{\partial r}\right), \tag{12}$$

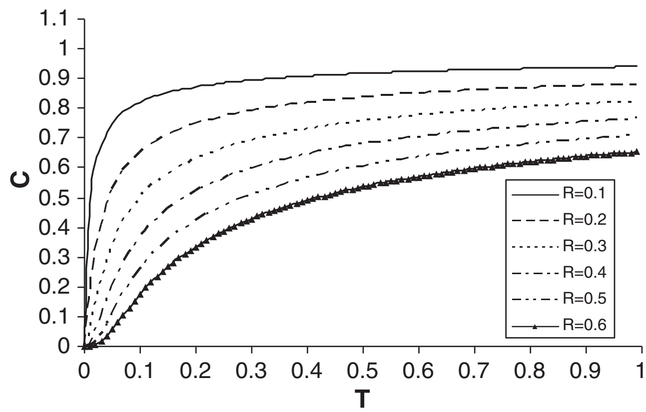

Fig. 1. Concentration profiles (the solutions of (8)), $r = r_0(t)$, α_0 =20, β = 0.1.

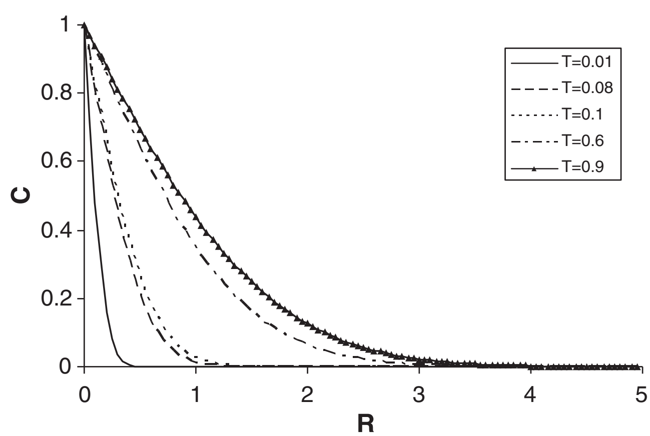

Fig. 2. Concentration profiles (the solutions of (8)), $r = r_0(t)$, α_0 =20, β = 0.1.

under following boundary conditions:

$$t = 0,\ u = 0; \quad r = r_0^{(0)},\ u = -\frac{D}{\rho_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}}; \quad r \to \infty,\ u = 0,$$
$$t = 0,\ c = c_0; \quad r = r_0^{(0)},\ c = c^*; \quad r \to \infty,\ c = c_0. \tag{13}$$

The introduction of the dimensionless variables (2) leads to:

$$\frac{\partial U}{\partial T} + \beta U \frac{\partial U}{\partial R} = Sc\left(\frac{\partial^2 U}{\partial R^2} + \frac{2}{\alpha_0 + R}\frac{\partial U}{\partial R} - \frac{2U}{(\alpha_0 + R)^2}\right);$$
$$\frac{\partial C}{\partial T} + \beta U \frac{\partial C}{\partial R} = \frac{\partial^2 C}{\partial R^2} + \frac{2}{\alpha_0 + R}\frac{\partial C}{\partial R};$$
$$T = 0,\ U = 0; \quad R = 0,\ U = -\left(\frac{\partial C}{\partial R}\right)_{R=0}; \quad R \to \infty,\ U = 0,$$
$$T = 0,\ C = 0; \quad R = 0,\ C = 1; \quad R \to \infty,\ C = 0, \tag{14}$$

where

$$u_0 = -\frac{D(c^* - c_0)}{\rho_0^* l} = \frac{(c^* - c_0)}{\rho_0^*}\sqrt{\frac{D}{t_0}}, \quad \beta = \frac{u_0 t_0}{l} < 1, \quad Sc = \frac{\upsilon}{D} >> 1. \tag{15}$$

The first equation of (14) is possible to be solved in the approximation $0 = Sc^{-1} \ll 1$. In the second equation of (14) $\beta < 1$ and the solution is possible to search in the form:

$$C = C^{(0)} + \beta C^{(1)}. \tag{16}$$

As a result Eq. (14) take the form:

$$\frac{\partial^2 U}{\partial R^2} + \frac{2}{\alpha_0 + R}\frac{\partial U}{\partial R} - \frac{2U}{(\alpha_0 + R)^2} = 0;$$
$$\frac{\partial C^{(0)}}{\partial T} = \frac{\partial^2 C^{(0)}}{\partial R^2} + \frac{2}{\alpha_0 + R}\frac{\partial C^{(0)}}{\partial R};$$
$$R = 0,\ U = -\left(\frac{\partial C^{(0)}}{\partial R}\right)_{R=0},\ C^{(0)} = 1;$$
$$R \to \infty,\ U = 0,\ C^{(0)} = 0; \quad T = 0,\ C^{(0)} = 0. \tag{17}$$

The first order expansion term for concentration, $C^{(1)}(R, T)$ is to be determined by solving the problem:

$$\frac{\partial C^{(1)}}{\partial T} = \frac{\partial^2 C^{(1)}}{\partial R^2} + \frac{2}{\alpha_0 + R}\frac{\partial C^{(1)}}{\partial R} - U\frac{\partial C^{(0)}}{\partial R};$$
$$T = 0,\ C^{(1)} = 0; \quad R = 0,\ C^{(1)} = 1; \quad R \to \infty,\ C^{(1)} = 0. \tag{18}$$

It can see, that problem described by Eq. (17) has analytical solution in following form:

$$U = \left(\frac{\alpha_0}{\alpha_0 + R}\right)^2\left(\frac{1}{\alpha_0} + \frac{1}{\sqrt{\pi T}}\right), \quad C^{(0)} = \frac{\alpha_0}{\alpha_0 + R} erfc \frac{R}{2\sqrt{T}}. \tag{19}$$

The problem formulated above is governed by two parameters α_0 and β. The first one depends of the particle radius, while the second reflects the average value of the concentration gradient at a boundary layer.

In Figs. 3 and 4 are shown concentration distributions as a function of T and R.

2.2. The particle radius decrease ($r = r_0(t)$).

In this case is occasion to be used iterative approach (9) again. The velocity of the solid particle radius (v) decrease and mass transfer rate (J) has the following forms:

$$v = -\frac{J}{\rho} = D\frac{\rho^*}{\rho_0^* \rho}\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)}, \quad J = -D\frac{\rho^*}{\rho_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r(t)}. \tag{20}$$

Effect of secondary flow velocity:

$$u = -\frac{D}{\rho_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)}, \tag{21}$$

leads to a new form of A and A' in the iterative procedure (9):

$$A(T) = \alpha_0 + \frac{c^* - c_0}{\rho}\frac{\rho^*}{\rho_0^*}\int_0^T\left(\frac{\partial C}{\partial R}\right)_{R=0} dT,$$
$$A'(T) = \frac{c^* - c_0}{\rho}\frac{\rho^*}{\rho_0^*}\left(\frac{\partial C}{\partial R}\right)_{R=0}. \tag{22}$$

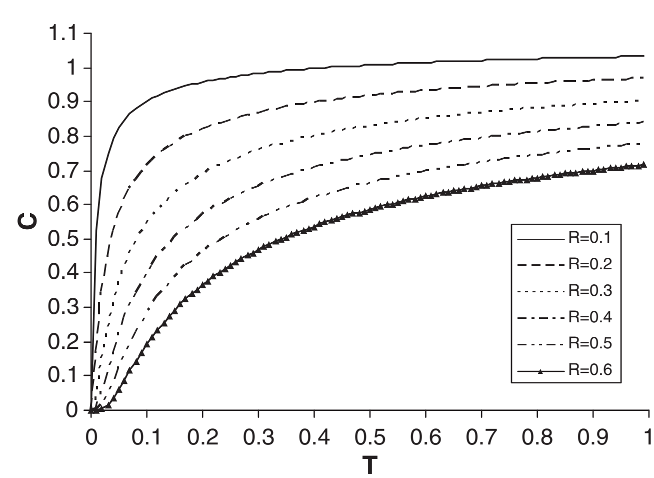

Fig. 3. Concentration profiles $C(T)$, $r = r_0^0$, α_0 =20, β = 0.1.

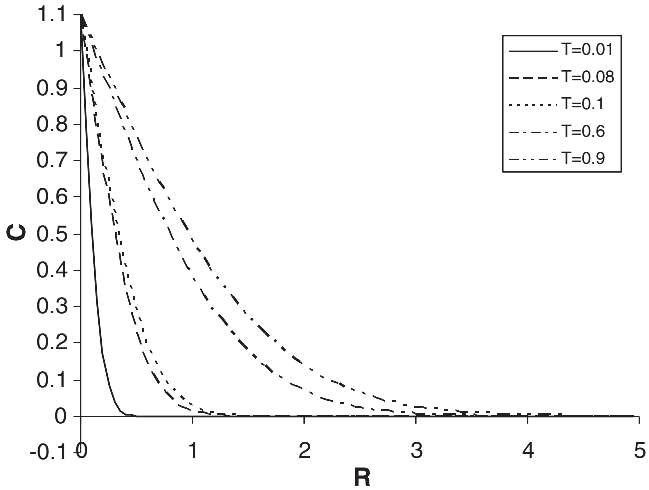

Fig. 4. Concentration profiles $C(R)$, $r = r_0^0$, $\alpha_0 = 20$, $\beta = 0.1$.

In Figs. 5 and 6 are shown concentration distributions $C(T)$ and $C(R)$.

In Figs. 7–12 are shown concentration distributions for different values of R and T. As a result is possible to be make comparison between the concentrations obtained from Eqs. (4), (8), (14) with the concentration obtained from Eq. (14), using iterative procedure (Eq. (9)).

The obtained concentration distributions near the solid particles permit to obtain the effect of the secondary flow velocity Eqs. (11), (21), which increases with the time and as a result of the particle radius decrease in the case of long time mass transfer is compensated.

The local mass transfer rate (local mass flux, kg m^{-2} s^{-1}) I and the amount of the dissolved substance q (kg s^{-1}) is obtained:

$$I = -D\left(\frac{\partial c}{\partial r}\right)_{r=r_0}, \quad q = 4\pi r_0^2 I, \tag{23}$$

where r_0 is the particle radius. As a result is possible to be presented the amount of the substance Q_0 (kg), dissolved for the time t_0 (s) and the average rate of mass transfer (dissolution) Q (kg m^{-2} s^{-1}):

$$Q_0 = \int_0^{t_0} q dt, \quad Q = \frac{Q_0}{4\pi r_0^2 t_0} = k(c^* - c_0), \tag{24}$$

where k is mass transfer coefficient:

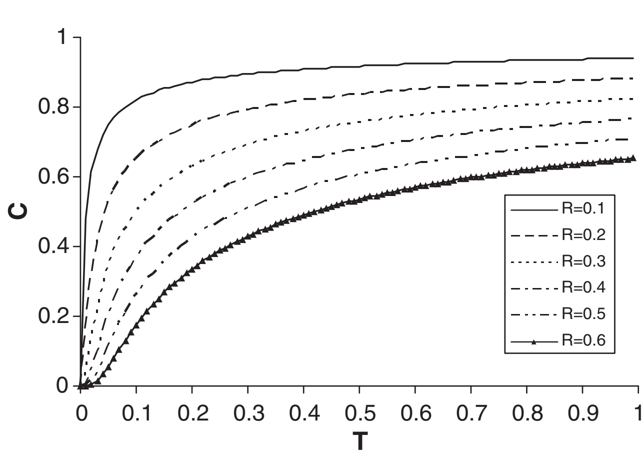

Fig. 5. Concentration profiles $C(T)$, $r = r_0(t)$, $\alpha_0 = 20$, $\beta = 0.1$.

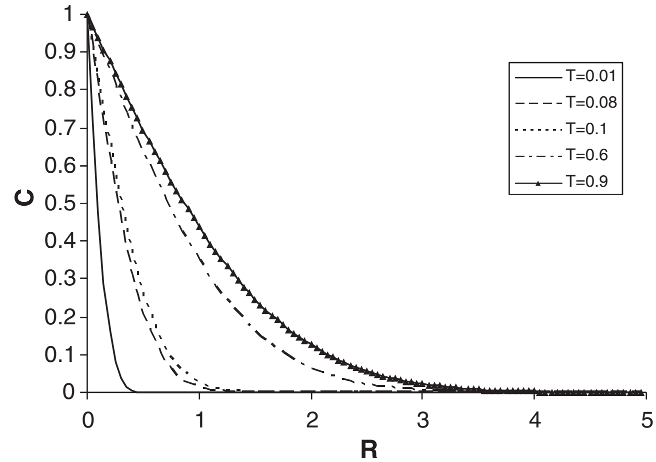

Fig. 6. Concentration profiles $C(R)$, $r = r_0(t)$, $\alpha_0 = 20$, $\beta = 0.1$.

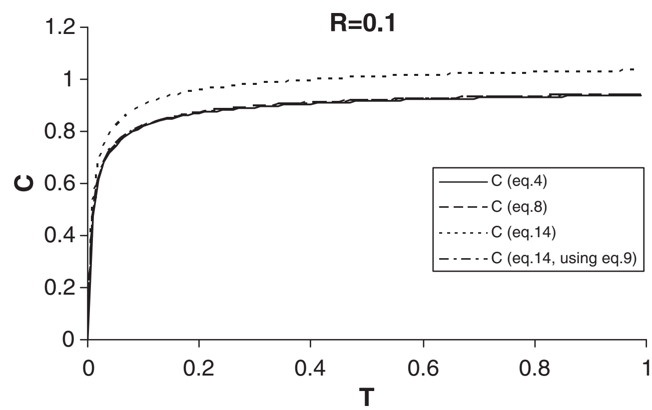

Fig. 7. Comparison of concentration profiles obtained from Eqs. (4), (8), (14) at $R = 0.1$.

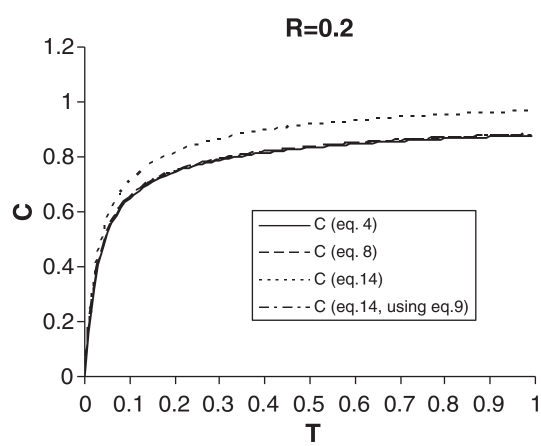

Fig. 8. Comparison of concentration profiles obtained from Eqs. (4), (8), (14) at $R = 0.2$.

$$k = -\frac{D}{(c^* - c_0)t_0}\int_0^{t_0}\left(\frac{\partial c}{\partial r}\right)_{r=r_0} dt. \tag{25}$$

From Eqs. (23)–(25) is possible to obtain Sherwood numbers for four cases, which are analyzed in this paper, and obtained results are shown in Table 1:

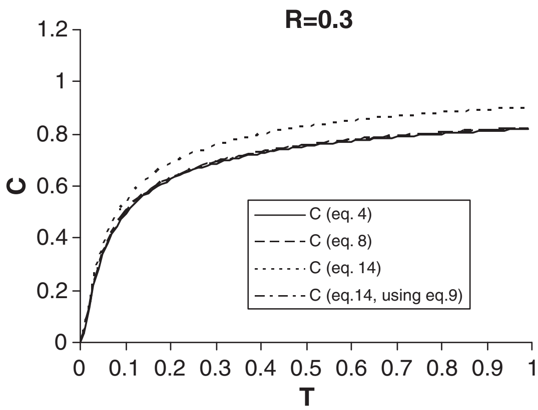

Fig. 9. Comparison of concentration profiles obtained from Eqs. (4), (8), (14) at $R = 0.3$.

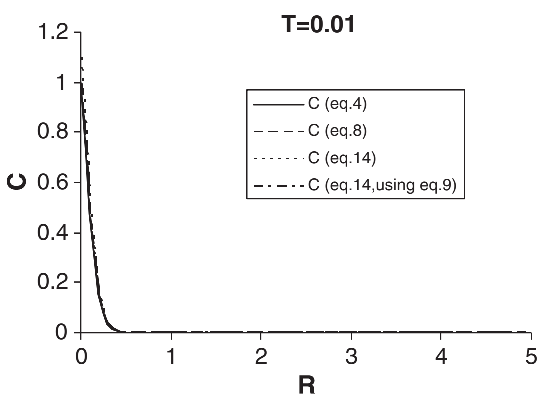

Fig. 10. Comparison of concentration profiles obtained from Eqs. (4), (8), (14) at $T = 0.01$.

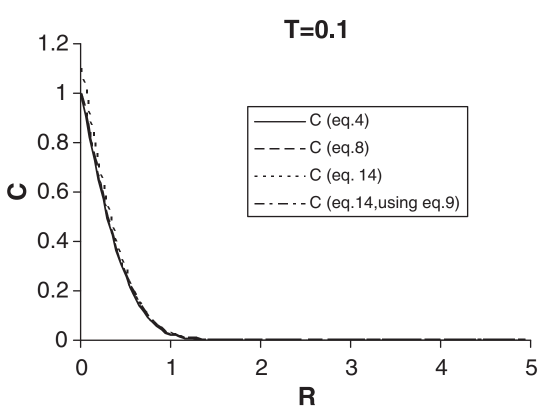

Fig. 11. Comparison of concentration profiles obtained from Eqs. (4), (8), (14) at $T = 0.1$.

1. Short time linear mass transfer ($r_0 = r_0^{(0)} = const$):

$$I = -D\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}}, \quad Q_1 = k_1(c^* - c_0) = -\frac{D}{t_0}\int_0^{t_0}\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}} dt,$$
$$t = t_0 T, \quad r = r_0^{(0)} + lR, \quad l = \sqrt{Dt_0}, \quad c(r,t) = c_0 + (c^* - c_0)C(R,T),$$
$$Sh_1 = \frac{k_1 l}{D} = k_1\sqrt{\frac{t_0}{D}} = -\int_0^1\left(\frac{\partial C}{\partial R}\right)_{R=0} dT. \tag{26}$$

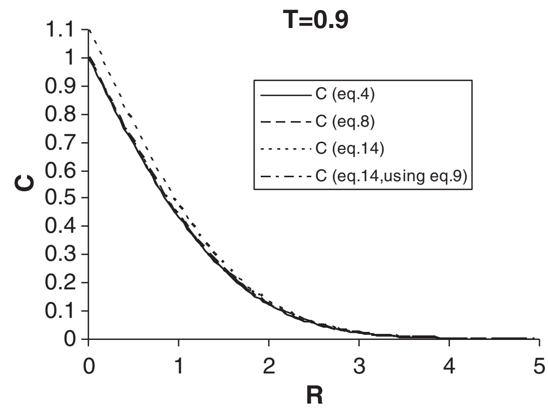

Fig. 12. Comparison of concentration profiles obtained from Eqs. (4), (8), (14) at $T = 0.9$.

Table 1
Sherwood numbers.

	Short time linear mass transfer ($r - 0 = r_0^{(0)} = const$)	Long time linear mass transfer ($r = r_0(t)$)	Short time non-linear mass transfer ($r_0 = r_0^{(0)} = const$)	Long time non-linear mass transfer ($r = r_0(t)$)
Sh	1.1838	1.1658	1.2994	1.1658

2. Long time linear mass transfer ($r = r_0(t)$):

$$r_0(t) = r_0^{(0)} + \int_0^{t_0} v(t)dt, \quad v = \frac{I}{\rho}, \quad I = I(t) = -D\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)},$$
$$t = t_0 T, \quad r = r_0^{(0)} + lR, \quad l = \sqrt{Dt_0}, \quad c(r,t) = c_0 + (c^* - c_0)C(R,T),$$
$$Sh_2 = \frac{k_2 l}{D} = k_2\sqrt{\frac{t_0}{D}} = -\int_0^1\left(\frac{\partial C}{\partial R}\right)_{R=0} dT. \tag{27}$$

3. Short time non-linear mass transfer ($r_0 = r_0^{(0)} = const, u_0^* = u(r_0^{(0)}, t) = -\frac{D}{c_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}}$),

where $c_0^*(\text{kg m}^{-3})$ – concentration of solvent on the surface $r_0 = r_0^{(0)}\rho^* = c_0^* + c^*(\text{kg m}^{-3})$ – density of solution on the surface $r_0 = r_0^{(0)}; u_0^*(\text{m s}^{-1})$ – liquid velocity (of the secondary flow) on the surface $r_0 = r_0^{(0)}$:

$$I = -D\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}} + u_0^* c^* = -D\frac{\rho^*}{c_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}},$$
$$Q_3 = k_3(c^* - c_0) = -\frac{D\rho^*}{t_0 c_0^*}\int_0^{t_0}\left(\frac{\partial c}{\partial r}\right)_{r=r_0^{(0)}} dt,$$
$$Sh_3 = \frac{k_3 l}{D} = k_3\sqrt{\frac{t_0}{D}} = -\frac{\rho^*}{c_0^*}\int_0^1\left(\frac{\partial C}{\partial R}\right)_{R=0} dT,$$
$$C = C_0 + \beta C_1. \tag{28}$$

4. Long time non-linear mass transfer ($r = r_0(t), u_0^* = u(r_0(t), t) = -\frac{D}{c_0^*}\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)}$):

$$v = -\frac{I_0}{\rho}, \quad I_0 = -\frac{\rho^*}{c_0^*}D\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)},$$
$$u^* = u_0^* + v = -\frac{D}{c_0^*}\left(1 - \frac{\rho^*}{\rho}\right)\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)},$$
$$I = -D\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)} + u^* c^* = -\frac{D\rho^*}{c_0^*}\left(1 - \frac{c^*}{\rho}\right)\left(\frac{\partial c}{\partial r}\right)_{r=r_0(t)},$$
$$Q_4 = \frac{1}{t_0}\int_0^{t_0} I dt = k_4(c^* - c_0),$$
$$Sh_4 = \frac{k_4 l}{D} = k_4\sqrt{\frac{t_0}{D}} = -\frac{\rho^*}{c_0^*}\left(1 - \frac{c^*}{\rho}\right)\int_0^1\left(\frac{\partial C}{\partial R}\right)_{R=0} dT,$$
$$C = C_0 + \beta C_1. \tag{29}$$

The results in Table 1 show, that the long time mass transfer rate (k) decreases as a result of the concentration radial gradient decrease with the time. In the non-linear case the mass transfer rate increase, but this effect disappear in the long time case too.

4. Conclusions

The non-linear mass transfer in case of the solid particles dissolution is analyzed theoretically. An iterative method for investigation of the influence caused by a decrease in the radius of particles is presented. The non-linear effect of the big concentration gradient is analyzed too. The comparison analysis shows, that the effect of non-linear mass transfer is bigger than the effect connected with the changing of radius. For the long time case the mass transfer rate decreases and both effects are equal as a result of decrease of the radial concentration gradient.

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