

INFLUENCE OF HYDRODYNAMIC CONDITIONS ON THE PROCESS OF HEAT AND MASS TRANSFER BETWEEN THE FLOW AND A RASCHIG RING CATALYST

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ABSTRACT

The influence of hydrodynamic conditions on heat and mass transfer for the process of oxidation of SO₂ to SO₃ on a single catalyst pellet – a Raschig ring, is investigated. A set of numerical experiments has been carried out for two different hypotheses of catalyst surface accessibility. A comparison of the obtained results for the temperature and conversion distributions in radial and axial directions in the catalyst pellet is provided.

Keywords: raschig ring catalyst, pellet surface accessibility, heat and mass transfer, numerical modeling.

INTRODUCTION

A wide variety of forms of granulated catalyst pellets (spheres, cylinders, rings, saddles, etc.), packed in layers with various structure and arrangement, have been used in industry. The orientation of the pellets towards the incoming stream, as well as determining the optimal geometrical correlation of their sizes, still continue to be the subject of undying researcher interest. In earlier investigations, for example [1], the influence of geometrical characteristics of a single cylindrical catalyst pellet on the heat and mass transfer between the pellet and the flow was studied theoretically for various kinetics of concrete chemical process and different boundary conditions. In papers [2 - 4] it has been validated experimentally that at fixed geometrical correlation between length, height and thickness for a catalyst Raschig ring, as well as for a special arrangement of these pellets in the layer, a significant influence on the catalyst efficiency for the respective technological process has been observed. It was found that using Raschig rings instead of spheres or other shaped pellets in a number of cases, is more advantageous [5 - 7]. The most essential indices upon which this statement is based are: decreasing of the pressure drop in the packed layer, prolonging of the term of catalyst usage and the high degree of utilization (efficiency η) of the internal catalyst surface [1, 8].

The aim of this work is to investigate the influence of hydrodynamics on the heat and mass transfer between a single catalyst pellet (Raschig ring with finite length and thickness) and the main gas flow. Two principal hypotheses will be considered depending on full or partial accessibility of internal catalyst surface, in respect to the flow. According to the validity of the above-mentioned hypotheses, the boundary conditions for temperature and concentration over the catalyst pellet will be calculated in a different manner. The fields of temperature and concentration are simulated numerically and a comparison will be done between results for both hypotheses at the following conditions: two velocities of gas flow 0,1 and 0,9 (m s⁻¹), flow viscosity $\nu=0,689 \cdot 10^{-4}$ (m² s⁻¹), Raschig ring catalyst pellet with dimensions 12,5 x 12,5 x 3 (mm). According to the hypotheses, the degree of utilization of the catalyst will be investigated too.

Model construction and hypothesis definition

The process of oxidation of SO₂ to SO₃ by diffusion of a gas mixture consisting of 7.5 % SO₂, and 11 % O₂ in a V₂O₅ catalyst layer, carried over the single ring-shaped inert Raschig ring, has been studied. The problem of heat and mass transfer between the gas phase and the single catalyst pellet will be solved at two different working hypotheses about type of boundary conditions – equal internal catalyst surface accessibility and partial accessibility, in respect to the gas flow. For the case of equal

accessibility, it is assumed that the transfer of heat and mass from the flow to the pellet does not depend on the pellet orientation towards the flow. In other words, the fluid velocity will be the same towards each part of the internal catalyst surface.

For the case of non-equal accessibility, the flow velocity which appears in the boundary conditions, implicitly by coefficients of heat and mass transfer - $\alpha, (W.m^2 K^{-1})$ and $\beta, (ms^{-1})$ respectively, will be different for each of the four parts of the external working surface of a ring-shaped granule. In conformity with this, the values of these coefficients will be different for each of the parts and can be calculated with the help of Todes's equation:

$$Nu = 2 + 0.55.Re^{0.5}.Pr^{0.33} \quad (1)$$

In the catalyst surface it has been assumed that heat and mass transfer are due to diffusion and heat conductivity. The influence of the surface accessibility (i.e., the type of boundary conditions) on the distribution of temperature and concentration, as well as on the degree of utilization of the catalyst, is investigated for two different flow velocities.

In cylindrical coordinates (r, l) the mathematical model of the investigated processes of heat and mass transfer between the flow and the catalyst surface, consists of the following system of differential equations and boundary conditions [1], and has the form:

$$D_p \left(\frac{\partial^2 C}{\partial r^2} + \frac{1}{r} \frac{\partial C}{\partial r} + \frac{\partial^2 C}{\partial l^2} \right) + W(C, T) = 0$$

$$\lambda_p \left(\frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial l^2} \right) + Q.W(C, T) = 0$$

$$r = R_1 \quad D_p \frac{\partial C}{\partial r} = \beta(C - C_0);$$

$$\lambda_p \frac{\partial T}{\partial r} = \alpha(T - T_0)$$

$$r = R_2 \quad -D_p \frac{\partial C}{\partial r} = \beta(C - C_0);$$

$$-\lambda_p \frac{\partial T}{\partial r} = \alpha(T - T_0)$$

(2)

$$l = 0 \quad D_p \frac{\partial C}{\partial l} = \beta(C - C_0);$$

$$\lambda_p \frac{\partial T}{\partial l} = \alpha(T - T_0)$$

$$l = H \quad -D_p \frac{\partial C}{\partial l} = \beta(C - C_0);$$

$$-\lambda_p \frac{\partial T}{\partial l} = \alpha(T - T_0)$$

Here (r, l) are the current radius and length of the cylindrical pellet in (m), respectively. It is assumed when working out the model, that the coefficients of internal diffusion $D_p, (m^2 s^{-1})$ and heat conductivity of the catalyst pellet $\lambda_p, (W.m^{-1} K^{-1})$, as well as the coefficients for heat and mass transfer α, β , are constants and do not depend on the direction of heat and mass transfer. This assumption remains valid for the hypotheses of equal accessibility and the value of flow velocity is the same for the whole external surface of the catalyst pellet. The heat effect of the reaction is marked with $Q, kJ mole^{-1}$ and the reaction rate $W, (mole.m^{-3} s^{-1})$ is of first order for the forward and reverse reaction. The considered process is supposed to be an adiabatic one. In the boundary conditions R_1, R_2, H are the internal and the external radii and length of the catalyst pellet, (m) respectively, T, C are the current temperature, ($^{\circ}K$) and concentration, $(mole.m^{-3})$. With indices "0" in Eq. (2) are noted the temperature and concentrations of the reagents in the flow. The system of differential equations and boundary conditions (Eq. (2)) has been transformed in finite differences equations then it has been solved numerically with the well-known method of trial and errors, which is described in [1], and will not be discussed any further here. The fields of dimensionless temperature TITA and conversion X on the catalyst pellet, as well as the degree of utilization of the catalyst surface η , are determined numerically as functions of coordinates. For the hypotheses of equal accessibility, the coefficients of heat and mass transfer in the boundary conditions are calculated by Eq. (1), for the respective value of the Reynolds number.

However, at non-equal accessibility the values of these coefficients for each part of the working surface of the catalyst pellet are different, because of the different values of the flow velocity. This reflects on the

method of their calculation. In this work the following manner is chosen: at the distance equal to the half of the characteristic pellet size in the normal direction from its external surface, one can calculate the mean value of the flow velocity on the external pellet surface:

$$v_{NEQA} = \frac{1}{|V|} \iint_S v(r, l) ds \quad (3)$$

In the common case, the flow velocity is a function of the coordinates (r, l) and can be obtained as follows:

$$|v| = \sqrt{v_r^2 + v_l^2}, \quad v_r = -\frac{1}{r} \frac{\partial \psi}{\partial l}; \quad v_l = \frac{1}{r} \frac{\partial \psi}{\partial r} \quad (4)$$

Here ψ ($m^3 s^{-1}$) is the stream function of the flow, and v, v_r, v_l are the flow velocity magnitude and its radial and axial components, ($m s^{-1}$), respectively.

Data for the stream function included in calculations has been taken from previous works [9, 10], where the numerical solution for a stream-function and vortex of the Navier-Stokes equations at axisymmetric incompressible flow around a hollow cylinder pellet (Raschig ring) with finite length and finite thickness of the wall, has been obtained for Reynolds number from 1 to 100 and at isothermal flow conditions. The area in which that solution was found was double-connected and in order to guarantee the uniqueness of the solution, and to find the unknown value of the stream function on the pellet surface, the criterion for simple quantity of the pressure must be satisfied.

In the case of non-equal accessibility, for the specific value of the Reynolds number and the respective value of the stream function on the catalyst pellet, both components of flow velocity and its magnitude from Eq. (4), as well as the mean integral value by Eq. (3), are calculated. This mean value has been included in Eq. (1) in Reynolds number and for particular input parameters of the process – temperatures, concentrations, viscosity, equivalent diameter, heat conductivity coefficient and so on, the values of heat and mass transfer coefficients in the case of non-equal accessibility of the catalyst surface have been obtained. After their determination, these integrally mean coefficients have been replaced in the boundary conditions in Eq. (2) and the system has been solved as in the case of equal-accessibility. For both considered hypotheses the degree of utilization of the external surface of the Raschig ring catalyst pellet

can be calculated as follows:

$$\eta_1 = \eta \left(1 - \frac{R_1}{R_2}\right)^2 = \frac{\iiint W(C, T)}{V_p x W(C_0, T_0)} \cdot \left(1 - \frac{R_1}{R_2}\right)^2 \quad (5)$$

For determining η_1 - degree of utilization of the hollow catalyst, as well as for the degree of utilization of the external surface of the solid cylinder η , the Thiele modulus has been used:

$$T_i = \frac{V_p}{S_p} \sqrt{\frac{W(C_0, T_0)}{D_p C_0}} \quad (6)$$

Here V_p, S_p are the volume and specific area of the catalyst pellet, in (m^3) and (m^2), respectively.

RESULTS AND DISCUSSION

Numerical solutions for the dimensionless temperature and conversion fields in radial and axial directions over the single catalyst pellet Raschig ring surface with dimensions 0,0125 x 0,0125 x 0,003 (m), at velocities of the gas stream flowing over the pellet 0,1 и 0,9 ($m s^{-1}$), have been obtained. The physico-chemical parameters of the gas mixture flow are as follows: viscosity - $\nu = 0,689 \cdot 10^{-4} m^2 s^{-1}$, density - $\rho = 0,5024 kg m^{-3}$, gas heat conductivity - $\lambda_g = 0,0527 (W m^{-1} K)$, gas specific heat capacity - $c_p = 0,489 (kJ kg^{-1} K^{-1})$, initial temperature in the main flow - $T_0 = 485 K$ and initial concentration of the reagent in the flow - $C_0 = 0,075 kmol m^{-3}$.

The calculated equivalent diameter of the catalyst pellet is 0.00726 m (accounted for Reynolds number) and the catalyst pellet heat conductivity is $\lambda_p = 0.527 (W m^{-1} K)$. The coefficient of internal diffusion in Eqs. (2) has a value - $D_p = 0.419 \cdot 10^{-5} (m^2 s^{-1})$.

The geometry of the considered problem, as well as the manner of integral mean velocity calculation in the case of non-equal accessibility, are presented in Fig. 1. The chosen distance, equal to the half of the characteristic size, corresponds to the area, in which the most essential changes in the distribution of the flow velocity profile in the immediate proximity of the catalyst pellet surface have been observed [9, 10]. Eq. (3) has been solved by numerical integration using the trapezoidal method.

The obtained numerical solution for the radial and axial distributions of temperature and the conversion

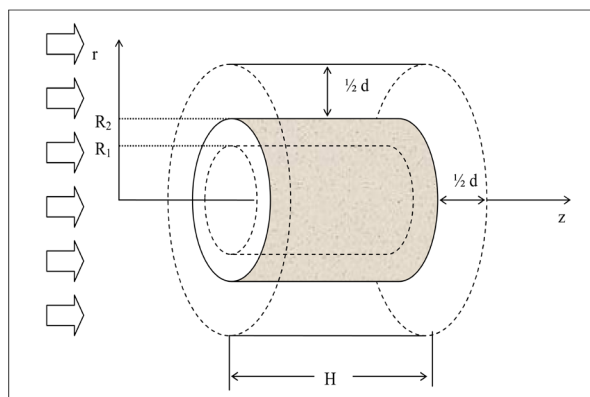


Fig. 1. Geometry of the considered problem and a scheme of the integral mean velocity calculation in the case of non-equal accessibility.

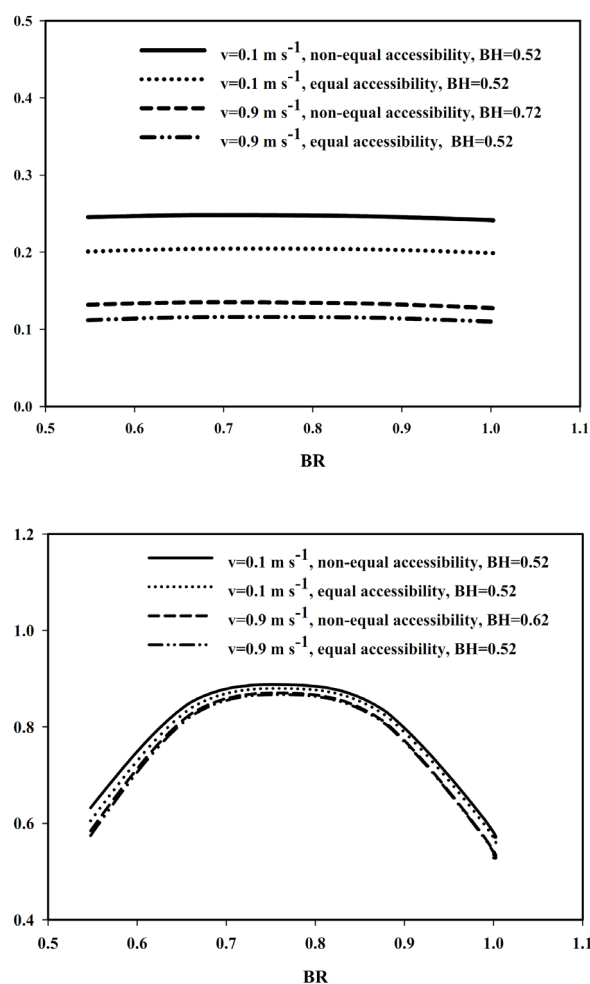


Fig. 2. Radial distribution of dimensionless temperature and conversion distribution for equal and non-equal accessibility of catalyst surface, for axially hottest point, for two flow velocities.

for the catalyst Raschig ring at two flow velocities and for both considered hypotheses are presented in Figs. 2 and 3. The comparison between the results is made at the hottest point radially (BR) and axially (BH), respectively. As seen from Figs. 2 - 3, the external velocity of the flow influences the distributions of temperature and conversion in both directions, since it is expressed stronger for the temperature distributions.

For the conversion, the type of distribution is significantly different in radial and axial directions, while for the temperature the character of distribution in both directions is practically the same. At the distribution of conversion in the radial direction there are well-visible maxima for both hypotheses, while in the axial direction there are sharp changes at the beginning and at end of the conversion profile with a plateau between them.

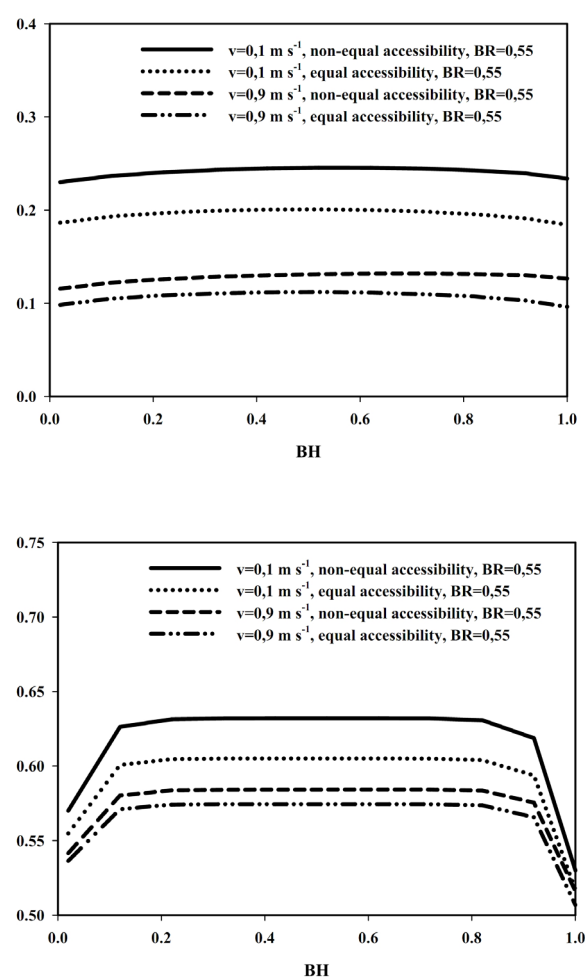


Fig. 3. Axial distribution of dimensionless temperature and conversion distribution for equal and non-equal accessibility of catalyst surface, for radially hottest point, for two flow velocities.

The choice of equal or non-equal accessibility exerts influence on the obtained distributions and the general tendency is that for non-equal accessibility the values of temperature and conversion are higher than in the case of equal accessibility, as was expected. In the radial direction both hypotheses give the same results for conversion for both flow velocities, while in the axial direction (Fig. 3) the increasing of flow velocity leads to decreasing of the conversion for both hypotheses. The degree of utilization of external catalyst pellet surface η_1 for both hypotheses has been calculated by Eq. (5), too. At flow velocity $0.9 \text{ (ms}^{-1}\text{)}$ the value of η_1 is 0.5044 for non-equal accessibility and 0.5075 – for equal accessibility; for flow velocity $0.1 \text{ (ms}^{-1}\text{)}$ 0.4827 and 0.4966, respectively, are obtained. Though very slightly, for the conditions of the considered process and catalyst pellet geometry the change in the velocity influences the degree of utilization of the external catalyst pellet surface for both hypotheses. The future work on this subject will be concerned with determining of optimal geometrical proportion for catalyst as well as identification of those external hydrodynamic conditions, which will guarantee a better utilization of its external surface.

CONCLUSIONS

The influence of the hydrodynamics conditions on the heat and mass transfer for the process of oxidation of SO_2 to SO_3 over a single catalyst pellet (Raschig ring) at two different hypotheses for equal and non-equal accessibility of its surface, has been investigated. The obtained numerical results for the distributions of temperature and conversion over the catalyst pellet surface for both hypotheses have been compared. It is shown that for both hypotheses the external fluid flow velocity depends on the radial and axial distribution of temperature and conversion, since a stronger impact can be observed for the temperature. In both directions (radially and axially) the distribution of temperature is uniform, while for radial conversion distribution one can observe a clearly visible maximum. For the axial direction - there are sharp changes at the beginning and at the end of the conversion profile with a plateau between them. The distributions of temperature correspond to the conditions of considered adiabatic process. The changes of the conversion distribution in both directions are relevant to the reversibility of the considered chemical reaction of first order over the catalyst pellet.

The general tendency for both hypotheses is that for non-equal accessibility the values of temperature and conversion are higher, than for the case of equal accessibility. The degree of utilization of the external catalyst surface depends slightly on the main flow velocity for both hypotheses.

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