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CFD simulation of a membrane bioreactor for high saline refinery wastewater treatment

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ABSTRACT

In the current research, simulation of chemical oxygen demand (COD) removal process from a high saline refinery wastewater using a membrane bioreactor equipped with a hollow fiber membrane, named as a hollow fiber membrane contactor (HFMC) was investigated using computational fluid dynamics method. A two-dimensional mathematical model was proposed to investigate COD transfer. This model describes the diffusion in the axial and radial directions of the HFMC. The model also examines the momentum transfer toward the tube and the shell. Comparing the model results with the experimental data, a deviation of 7.35% attained which reflects the proper reliability of the proposed model to predict the absorption of COD in the HFMC.

Keywords: Simulation; Hollow fiber membrane contactor; Chemical oxygen demand; Computational fluid dynamics

1. Introduction

With the world population growth and increasing the industrial plants, water consumption around the world has been increased. Since available water resources are limited, reuse of wastewater is receiving significant attention [1]. However, if the recirculated water is not properly treated, harmful compounds could be accumulated [2]. Membrane bioreactors (MBRs) are one of the most important systems in the field of industrial and domestic wastewater treatment in recent years which are preferred to other competing technologies. MBR technology is a combination of activated sludge process with membrane filtration [3]. Hollow fiber membranes have been accepted in different industries due to great advantages. Their high surface to volume ratio provides the possibility to benefit from the required high flux. In addition, the aeration operation of fibers can be easily done in order to prevent the fouling by using the appropriate module design used in wastewater treatment. A hollow fiber membrane immersed in a biological reaction, known as a submerged membrane bioreactor, is a type of hollow fiber membrane contactors (HFMCs) with membrane installed in the aeration tank, has attracted significant attention for municipal wastewater treatment [4–8]. It is preferred as compared with its counterpart which is usually made from the flat sheet type and located outside the biological response system, due to the below advantages [9]:

- Energy consumption and investment costs are low for immersion hollow fiber systems;
- The lower cost of hollow fiber modules compared with the flat sheet ones;
- The ease and low cost cleaning.

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In simultaneous with the growth of HFMC usages, modeling the systems was also developed. Sanaeepur et al. [10] used a two-dimensional mathematical model to study the process of nitrate separation from drinking water by a cylindrical MBR. Their attention has been especially given to develop a mathematical model for transient behavior of an extractive HFMC in drinking water denitrification. Incorporating suitable momentum transfer equations, the effects of lumen and shell velocity or, in turn, flow rates, recirculation system and membrane structural parameters on NO_3^- mass transfer, and consequently, denitrification efficiency was studied. The model resulted in a deviation of 8% compared with the experimental data. Shirazian and coworkers [11–16] presented a series of computational fluid dynamics (CFD) simulation studies in order to model the solute transports in the HFMCs. They considered mass and momentum transfer equations in axial and radial coordinates of the membrane fibers and the shell in laminar modes. It can be said that the main differences between these works are the materials (solvent–solute) that should be transferred through the HFMCs, the wetted or non-wetted modes of operations, and the phases that involved in the operations (solid–liquid, liquid–liquid or gas–liquid phases). These leaded to some differences in the model accuracies in predicting the data. Moreover, Al-marzouqi et al. [17] modeled CO_2 absorption in a membrane contactor. This model compared with the experimental results and showed good predictions in different amounts of liquids and gases at different temperatures. Azari et al. [18] presented a comprehensive two-dimensional mathematical model for CO_2 removal in an HFMC using monoethanol amine as the absorbent solvent. They considered both non-wetted and partially wetted (different wetting values) conditions for a countercurrent gas–solvent flow arrangement. They also investigated the effects of operational conditions and membrane's structural properties on CO_2 removal efficiency and the overall mass transfer coefficients. They concluded that when the gas flows in the fibers and solvent in the shell, a smaller HFMC can be used. In a new work, Ghadiri and Shirazian [19] numerically simulated the recovery of benzoic acid (BA) from aqueous wastewaters flows in the tube side of an HFMC using trioctylamine solvent in the shell side. They concluded that the BA removal does not affected considerably by a change in solvent flow rate and initial concentration of BA. However, an increase in feed flow rate results in enhancing the convective mass transfer in the tube side and finally decreases removal efficiency of BA.

In the present work, it is presented a two-dimensional mathematical model for mass and momentum transfer in an HFMC which can be capable for predicting the results of treating a high salt containing refinery wastewater. The model considers both axial and radial diffusions in the fibers and shell of HFMC. Effect of process parameters on the contaminant removal was also considered. Afterwards, the developed model is validated with the experimental data.

2. Modeling

Modeling can lead to examine the impact of different designing parameters on the system at the least cost. Here, mass and momentum transfer equations for MBR are provided and solved using a finite element method (FEM). The membrane is immersed in the wastewater and mass transfer is carried out from the shell (the reaction medium) toward the tube (hollow fiber membrane).

2.1. Modeling of mass and momentum transfer in the MBR

Two-dimensional mathematical modeling is used to simulate the hollow fiber membrane immersed in the bioreactor which separates the chemical oxygen demand (COD) from industrial wastewater. Here, COD penetrates from the shell into the membrane pores. Fig. 1 shows a schematic for the MBR system used in the case.

The below assumptions were used in the modeling:

- Steady-state conditions and isothermal flow without any considerable frictional losses, stresses and heat of reactions.
- Fully developed and laminar flow velocity profile due to a small radius of the fibers and a negligible end effects in their entrances.
- No reaction in the membrane fibers, because the microorganisms that carried out the bioreaction present only outside the fibers.
- Henry's law for liquid–liquid interface due to an ideal absorption of COD (or the oxygen) in the liquids.

2.1.1. Tube (fiber) equations

Each of the fiber in an HFMC has a cylinder shape that could be modeled in cylindrical coordinates (radius ρ, azimuth ϕ and elevation z). According to the proposed assumptions in the text, a constant azimuth ϕ and also a radial symmetry were the majority of presenting a two-dimensional mathematical model. The continuity equation for COD removal on the tube with cylindrical coordinates using Fick's law of diffusion is as follow:

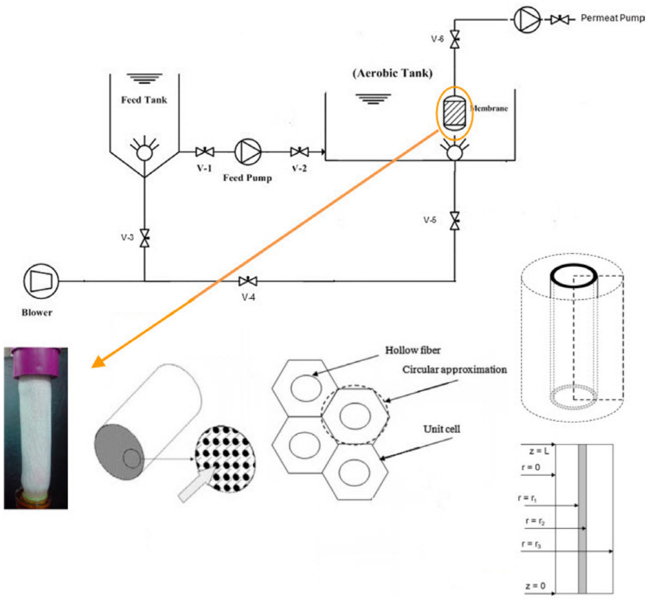

Fig. 1. A schematic representation of the MBR system for COD separation from refinery wastewater.

$$\frac{\partial C_{C-\text{tube}}}{\partial t} - D_{C-\text{tube}}\left[\frac{\partial^2 C_{C-\text{tube}}}{\partial r^2} + \frac{1}{r}\frac{\partial C_{C-\text{tube}}}{\partial r} + \frac{\partial^2 C_{C-\text{tube}}}{\partial z^2}\right] = R_{C-\text{tube}} - V_z \frac{\partial C_{C-\text{tube}}}{\partial z} \quad (1)$$

In this equation, $D_{C\text{-tube}}$, $C_{C\text{-tube}}$, $R_{C\text{-tube}}$ and $V_{z\text{-tube}}$, respectively, are diffusion coefficient, concentration, reaction rate and fluid velocity. $R_{C\text{-tube}}$ is zero because there is no reaction on the tube. Since "z" is much larger than "r", for the numerical solution of the equation a scaling factor is used for "z".

Assuming Newtonian laminar flow, velocity distribution on the tube is obtained from the following equation [13]:

$$V_{z-\text{tube}} = 2u\left[1 - \left(\frac{r}{r_1}\right)^2\right] \quad (2)$$

where u (m/s) is the average velocity on the tube and r_1 is the radius of the fibers.

The boundary conditions for the tube are as follows:

$$C_C = 0 \quad @ \quad Z = 0 \quad \text{Concentration} \quad (3)$$

$$n.(-D_C \nabla C_C = 0 \quad @ \quad Z = L \quad \text{convective flux} \quad (4)$$

C_C (mol/m^3) is COD concentration inside the tube. It is assumed that the convective flux boundary condition can be considered constant in all the fibers. According to this assumption, the permeate flux across the tube is zero.

$$\frac{\partial C_C}{\partial r} = 0 \ @ \ r = 0 \quad (5)$$

$$C_{C-\text{tube}} = C_{C-\text{membrane}} \times m \ @ \ r = r_1 \quad (6)$$

In this case, $C_{C\text{-membrane}}$ (mol/m^3) is COD concentration inside the membrane and m is COD solubility in the liquid solvent.

2.1.2. Membrane equations

Steady continuity equation to transfer COD within the membranes is only defined by penetration. It is as follows:

$$\frac{\partial C_{C-\text{membrane}}}{\partial t} - D_{C-\text{membrane}}\left[\frac{\partial^2 C_{C-\text{membrane}}}{\partial r^2} + \frac{1}{r}\frac{\partial C_{C-\text{membrane}}}{\partial r} + \frac{\partial^2 C_{C-\text{membrane}}}{\partial z^2}\right] = 0 \quad (7)$$

In the above equation, $C_{C\text{-membrane}}$ (mol/m^3) and $D_{C\text{-membrane}}$ (m^2/s) are, respectively, representing COD concentration and COD penetration rate of COD. There is no chemical reaction in the membranes. For this reason, it did not consider in reaction equation. Since mass transfer is only as penetration, Fick's law can be used to penetrate.

The membranes boundary conditions are as follows:

$$n.\text{COD} = 0; \ \text{COD} = -D_{\text{COD}} \nabla C_{\text{COD}} \ @ \ z = 0 \text{ and } z = 1 \quad (8)$$

$$C_{C\text{-membrane}} = C_{C\text{-tube}}/m \quad @ \quad r = r_1 \quad (9)$$

$$C_{C\text{-membrane}} = C_{C\text{-shell}} \quad @ \quad r = r_2 \quad (10)$$

where m is a partition coefficient and it depends on the concentration.

2.1.3. Shell equations

The continuity equation in the steady state to transfer COD on the shell HFMC in cylindrical coordinates by Fick's law diffusion is as follows:

$$\frac{\partial C_{C-\text{Shell}}}{\partial t} - D_{C-\text{Shell}}\left[\frac{\partial^2 C_{C-\text{Shell}}}{\partial C^2} + \frac{1}{r}\frac{\partial C_{C-\text{Shell}}}{\partial r} + \frac{\partial^2 C_{C-\text{Shell}}}{\partial z^2}\right] = R_{C-\text{Shell}} - V_{z-\text{Shell}}\frac{\partial C_{C-\text{Shell}}}{\partial z} \quad (11)$$

where r and z, respectively, are the radial and longitudinal coordinates. The reaction term did not define in the shell, since no reaction happened in HFMC shell. It is noted that chemical reaction will advance biomass in the shell. It is assumed that reactants react with the bacteria and in the biomass (bacteria) environment does not happen to any other chemical changes. So, by assuming a homogeneous environment, velocity distribution on the shell is obtained by solving the equations of motion (momentum) including Navier–Stokes equations. Density and viscosity of the fluid modeling are also assumed constant. Navier–Stokes equation is defined as follows [20]:

$$-\nabla.\eta\left(\nabla V_{z-\text{Shell}} + \left(\nabla V_{z-\text{Shell}}\right)^T\right) + \rho\left(V_{z-\text{Shell}}.\nabla\right)V_{z-\text{Shell}} + \nabla p = F$$
$$\nabla.V_{z-\text{Shell}} = 0 \quad (12)$$

where η, V and ρ are considered for dynamic viscosity of the liquid (kg/m s), the velocity vector (m/s) and density (kg/m^3), respectively. p represents the pressure (atm) and F indicates the force (N). Happel model can be used to estimate the radius of the shell [21]. Free surface radius (r^3) can be defined as follows:

$$r_3 = \left(\frac{1}{1-\phi}\right)^{1/2} r_2 \quad (13)$$

Volume percentage of empty parts (void) is calculated by the following equation:

$$1 - \phi = \frac{nr_2^2}{R^2} \quad (14)$$

where n indicates the number of fibers and R represents inner radius of whole bunch of membrane fibers together.

Happel's free surface model can also be used to determine the velocity of the liquid in the shell. Some studies have used the Happle's free surface model [22], but the Navier–Stokes equations are a more general method to determine the speed on the shell and can be used for all membrane geometries [11].

The boundary conditions for shell are given below:

Continuity equation:

$$C_{C\text{-shell}} = C_{C\text{-membrane}} \quad @ \quad r = r_2 \quad \text{concentration} \quad (15)$$

$$C_{C\text{-shell}} = C_{\text{COD}} \quad @ \quad r = r_3 \quad \text{concentration} \quad (16)$$

$$C_{C\text{-shell}} = C_{\text{COD}} \quad @ \quad z = 0 \quad \text{concentration} \quad (17)$$

$$n.(-D_{C-\text{membrane}} \nabla C_{C-\text{membrane}}) = 0 \ @ \ z = L \ \text{Convective flux} \quad (18)$$

Momentum equation:

$$V_z = 0 \quad @ \quad r = r_2 \quad \text{no slip wall} \tag{19}$$

$$V_z = V_{z,0} \quad @ \quad r = r_3 \quad \text{open boundary} \tag{20}$$

$$V_z = V_{z,0} \quad @ \quad z = 0 \quad \text{inlet} \tag{21}$$

$$p = p_0 \quad @ \quad z = L \quad \text{outlet} \tag{22}$$

where $C_{\text{COD-shell}}$ (mol/m^3), $V_{z,0}$ (m/s) and p_0 (Pa), respectively, are COD concentration, speed at input and output pressure in the shell.

Fig. 2 presents the generated mesh for solving the model equations.

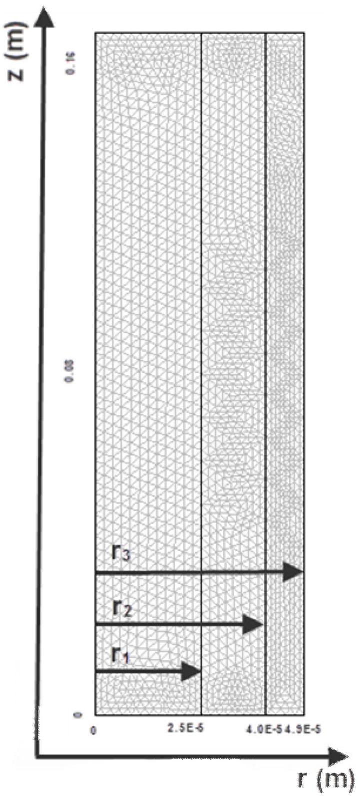

Fig. 2. A generated mesh for the domain considered at the MBR.

2.2. Numerical method

Using the governing equations and boundary conditions, the proposed model will be solved for COD separation from wastewater in the MBR. Physical characteristics and the model parameters are provided in Table 1. The governing equations are solved numerically using the COMSOL (version 4.2) software.

The COMSOL software uses the FEM for the numerical solution. It also uses UMFPACK solver. It is a very powerful direct solver for symmetrical geometries. Practicality, strength and accuracy of the method for membrane processes have proven by some authors [11,23]. A computer (processor Intel Pentium 4, speed CPU = 2,800 MHz) was used to solve the equations.

3. Results and discussion

3.1. Membrane efficiency

The efficiency of the membrane is obtained by subtraction of COD transferred toward the tube, divided by the total COD in the feed which is defined as follows:

$$\text{Yield} = \frac{Q(C_C(0,r,t) - C_C(L,r,t))}{QC_C(0,r,t)} \tag{23}$$

In this equation, "Q" represents the flow rate and "C_{COD}" is the concentration in the area. As Fig. 3 shows, increasing the flow rate within the tube increases the membrane efficiency.

Fig. 4 shows the impact of concentration on the shell side of the membrane on the MBR efficiency. As it can be seen, decreasing the concentration on the shell side will increase the membrane efficiency.

3.2. Model validation

The model predictions were compared with the Razavi's experimental results [24] and the deviation was calculated using the following equation:

$$\text{Deviation} = \left| \frac{\text{experimental} - \text{theoretical}}{\text{experimental}} \right| \times 100 \tag{24}$$

Razavi's data were collected from the removal of COD from a refinery wastewater collected from all units of Imam Khomeini Oil Refinery, Shazand, Iran, with 100 ppm feed concentration. In this regard, a membrane biological reactor equipped with polypropylene hollow fibers was used. Here, the purpose of wastewater treatment is using the recycled water as the makeup water for the cooling towers. A part of the makeup water of each tower is provided by raw water and the rest is provided from the existing recycled water. The amount of experimental COD removal was 81.08% which deviated 7.35% from the value of 87.04% obtained from the present CFD simulation. According to the simplifying assumptions considered in the model, its power in predicting the experimental results is good.

3.3. COD flux in the MBR

Fig. 5 indicates COD flux direction (penetration and translocation) inside the tube, membrane and shell. COD along with water entered the shell ($z = 0$), where COD concentration has its greatest value ($C_{\text{COD-shell}} = 0.02$). COD passed through the membrane and separation was done on the pipe. In the pipe, COD concentration tends toward zero. The maximum COD concentration is in the shell. According to vector directions, the flux of mass transfer in the membrane is in radial direction. However, in the shell and tubes, the fluxes of mass transfers are more in the longitudinal direction.

3.4. Velocity distribution

The velocity profile in the feed side of the membrane module was determined by solving the Navier–Stokes

Table 1
The parameters used for solving the model

Parameter	Value	Description	Reference
r_1	2.5e–5	Fiber inner radius (m)	Measured using SEM
r_2	4.0e–5	Fiber outer radius (m)	Measured using SEM
r_3	4.9e–5	Radius of free surface (m)	Calculated
R	4e–3	Module radius (m)	Measured
n	1,940	Total number of tube	Membrane manufacturer
L	0.16	Model length, L (m)	Measured
$D_{\text{COD-shell}}$ [a]	2e–10	Diffusivity of COD in shell (m^2/s)	Calculated
$D_{\text{COD-membrane}}$ [b]	$D_{\text{COD-shell}}$ (ε/τ)	Diffusivity of COD in membrane (m^2/s)	Calculated
$D_{\text{COD-tube}}$ [a]	3.2e–8	Diffusivity of COD in tube (m^2/s)	Calculated
m	0.78	Partition coefficient	Calculated
ε	0.25	Membrane porosity	Membrane manufacturer
τ	4	Membrane tortuosity	Membrane manufacturer
Q_s	11.442e–7	Water flow rate in shell (m^3/s)	Calculated
p_0	1.01325e–5	Pressure (Pa)	Measured
T_0	298	Temperature (K)	Measured
ρ	1	Density (g/cm^3)	Measured
η	1.0e–6	Viscosity (kg/m s)	Measured
C_{COD}	0.2	COD concentrations in wastewater (kg/m^3)	Measured

[a]Calculated based on Wilke–Chang theory for liquid mixtures.
[b]Effective diffusion coefficient is calculated by considering the effects of porosity (0.25) and tortuosity (4) of the membrane, as provided by the membrane manufacturer.

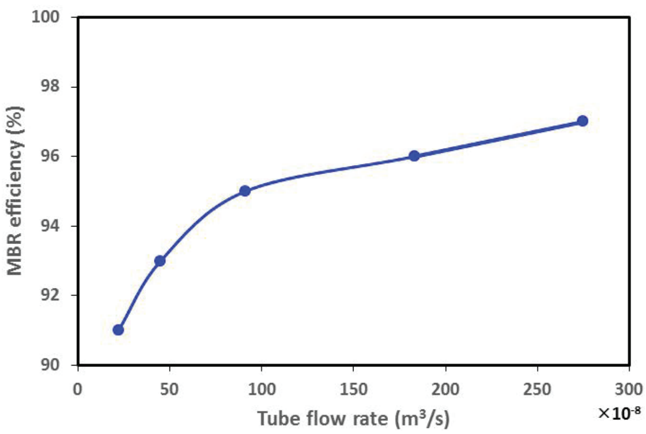

Fig. 3. The impact of tube side flow rate on the membrane efficiency.

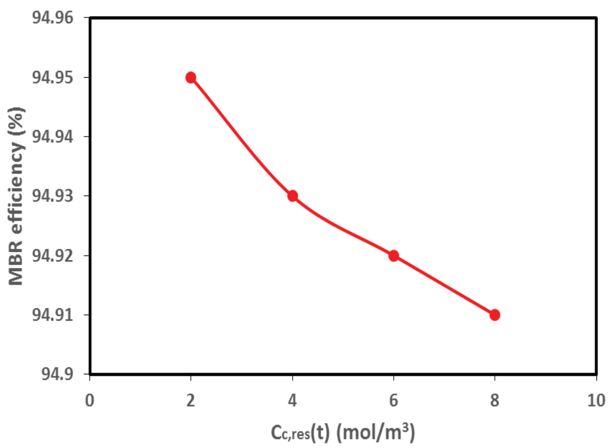

Fig. 4. The effect of concentration in the shell side of the membrane on the MBR efficiency.

equations. The velocity profile along the membrane module is also shown in Fig. 6. It indicates that there is a maximum velocity at a region near the entrance. The maximum velocity increases with increasing feed flow rate in the membrane module.

3.5. Concentration profile of solute along the membrane

Concentration profile (mol/m^3) of solute along the membrane is shown in Fig. 7. The concentration profile is obtained in the lumen (tube) side of the membrane contactor where the aqueous feed flows. It should be pointed out that the flow pattern in the contactor is parallel and countercurrent. The feed phase including aqueous solution of COD flows from one side of the contactor (z = 0) where the concentration of solute is the highest (C_0), whereas the organic phase flows from the other side (z = L) where the concentration of solute is assumed to be zero. As the feed flows through the lumen side, solute (COD) is transferred toward the membrane due to the concentration difference. Fig. 7 confirms the concentration of COD decreases along the extractor. Fig. 7 also indicates that increasing feed flow rate decreases the solute removal in the contactor. The extraction efficiency of solute which is defined as the ratio of the solute transfer from the feed phase to extraction phase to total solute in the initial feed phase is calculated using the numerical simulation.

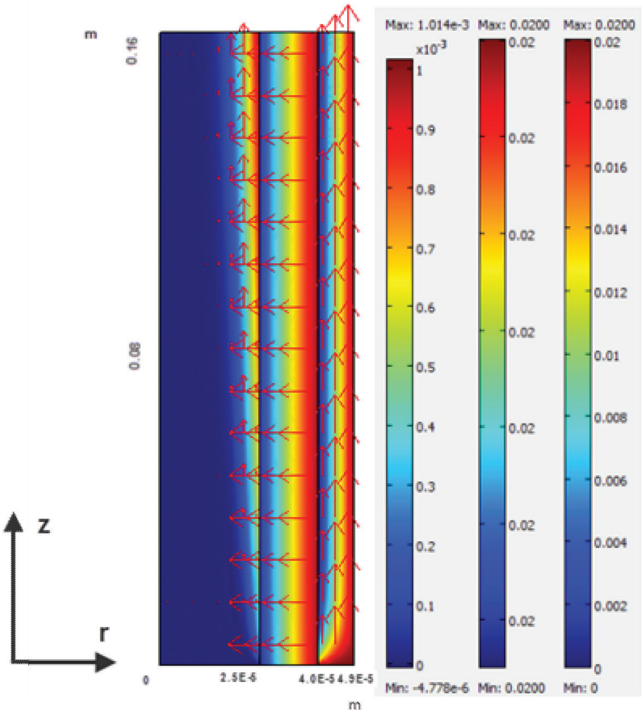

Fig. 5. A representation of total flux in the membrane contactor for r_1 = 2.5e–5 m, r_2 = 4.0e–5, r_3 = 4.9e–5 m, L = 0.16 m, D_{tube} = 2e–10 m²/s, $D_{membrane}$ = 1.25e–11 m²/s, D_{shell} = 3.2e–8 m²/s, m = 0.78 and Q_{shell} = 11.442e–7 m³/s.

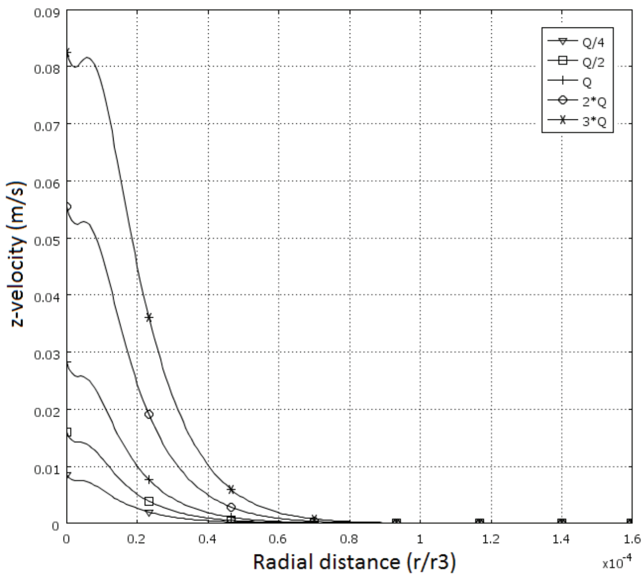

Fig. 6. Velocity profile on the shell at different discharge values.

4. Conclusions

CFD simulation of a hollow fiber MBR used for separation of COD from refinery wastewater was carried out. The model equations were solved using the FEM. Navier–Stokes equations were used to determine the velocity distribution in the shell. The velocity and concentration profiles were evaluated in order to assess the impact of process parameters in the COD removal. The simulation results showed that the proposed model has a fairly good accuracy in predicting the experimental results with a deviation of 7.35%.

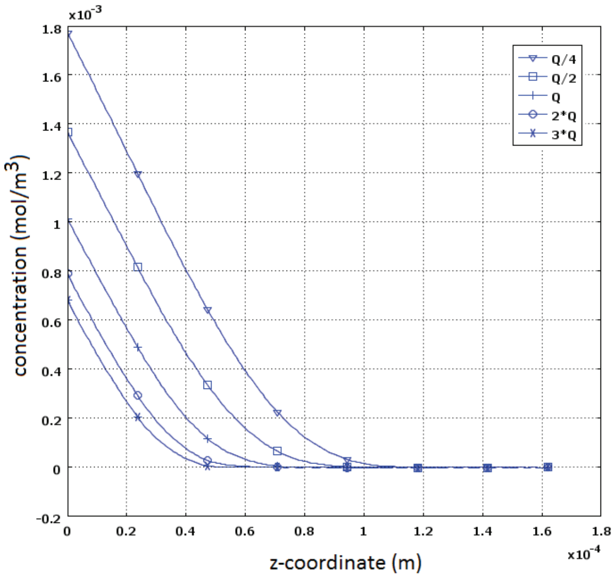

Fig. 7. The concentration on the tube at different flow rates.

Symbols

$C_{COD\text{-}initial}$	—	Initial COD concentration, mol/m³
$C_{COD\text{-}membrane}$	—	COD concentration in the membrane, mol/m³
$C_{COD\text{-}shell}$	—	COD concentration in the shell, mol/m³
$C_{COD\text{-}tube}$	—	COD concentration in the tube, mol/m³
C_i	—	Concentration of any species, mol/m³
C_0	—	Inlet COD concentration, mol/m³
$D_{COD\text{-}membrane}$	—	Diffusion coefficient of COD in the membrane, m²/s
$D_{COD\text{-}shell}$	—	Diffusion coefficient of COD in the shell, m²/s
$D_{COD\text{-}tube}$	—	Diffusion coefficient of COD in the tube, m²/s
D_{gas}	—	Gas phase diffusion coefficient, m²/s
$D_{i\text{-}shell}$	—	Diffusion coefficient of any species in the shell, m²/s
D_{liquid}	—	Liquid phase diffusion coefficient, m²/s
L	—	Length of the fiber, m
m	—	Partition coefficient
N_i	—	Total flux of any species, mol/m² s
r_1	—	Inner tube radius, m
r_2	—	Outer tube radius, m
r_3	—	Inner shell radius, m
R_i	—	Overall reaction rate of any species, mol/s
u	—	Average velocity, m/s
V_z	—	Velocity in the module, m/s
$V_{z\text{-}shell}$	—	Velocity in the shell, m/s
$V_{z\text{-}tube}$	—	Velocity in the tube, m/s
z	—	Axial distance, m
P	—	Pressure, Pa
T	—	Temperature, K
v	—	Fluid velocity, m/s

Greeks

ε	—	Porosity
τ	—	Tortuosity
Φ	—	Module volume fraction, m^3
ρ	—	Density, g/L
η	—	Viscosity (cp)

Subscripts

Shell	—	Space around the fibers in the MBR
Tube	—	Inner part of the fiber

Abbreviations

FEM	—	Finite element method
HFMC	—	Hollow fiber membrane contactor
CFD	—	Computational fluid dynamics
COD	—	Chemical oxygen demand

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