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Modeling organics biodegradation and ammonia nitrification by entrapped mixed microbial cell carriers

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ABSTRACT

Entrapped mixed microbial cell (EMMC) process offers good capability to remove organics and nitrogen compounds from wastewater in a single aerobic chamber. This research modeled quantitatively the hydraulic characteristics and biochemical process of immobilized activated sludge process (ASP) for the removal of COD and NH_4^+-N, providing insights to mass and oxygen transfer limitation in EMMC spherical carriers. Based on the conceptual kinetic model and previous experimental results, hydraulic and reaction rate constants were determined for both COD degradation and NH_4^+-N nitrification with the EMMC carrier. The dissolved oxygen (DO) distribution profile along the radius of EMMC carriers was also simulated. The depletion of DO in the EMMC carrier was very rapid resulting from COD removal and ammonia nitrification given the mass transport condition of DO. The anoxic/anaerobic zone developed in the EMMC carrier within 1 cm from its external surface in contact with the bulk water phase. Beyond this anoxic/anaerobic boundary, denitrification of nitrate occurred utilizing the residual COD. The efficiency of organics biodegradation and nitrification was not influenced by the thickness or diameter of the EMMC carriers. EMMC carriers of 1 cm in thickness supported removal of organics by biodegradation and nitrogen compounds via nitrification and denitrification processes. The EMMC carrier enabled combined nitrification and denitrification in the aerobic chamber, which signified the enhancement of a traditional ASP to an anoxic/oxic (AO) or anaerobic/anoxic/oxic reactor system via the EMMC carrier in an aeration tank.

Keywords: Modeling; Entrapped mixed microbial cells; Nitrification; Denitrification

1. Introduction

Biological treatment processes rely on bacteria to reduce discharges of organic wastes into receiving water bodies. The activated sludge process (ASP) involves an aeration basin to convert organics to innocuous inorganics and biomass, with a clarifier to concentrate the newly formed biomass for wastage and recycle of a portion into the aeration basin. Although

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conventional ASPs effectively remove biochemical oxygen demand (BOD), the generation of a large amount of waste sludge and the need to return the activated sludge into the aeration basin has remained as significant issues. In addition, the traditional ASP offers no capability to remove total nitrogen from wastewaters. Advanced processes such as anaerobic, anoxic, and oxic (A2O) incorporate both anaerobic and aerobic conditions to effectively degrade BOD and carry out nitrification and denitrification in separate chambers. The A2O process converts ammonia nitrogen biologically to nitrate in an oxic chamber and the wastewater is recirculated to an anaerobic chamber where the organics-rich wastewater undergoes denitrification. While many biological processes have been modified to improve removal efficiencies of organics and nitrogen, all of these modified processes share disadvantages of requiring additional chambers for separate oxic and anoxic zones as well as recirculation of the nitrified wastewater to the anaerobic/anoxic chamber.

Yang et al. [1] developed entrapped mixed microbial cell (EMMC) to physically simplify the A2O process. Yang tested EMMC treatment for various wastewaters including domestic sewage [1–3], phenol [4], odor producing substance [5], dimethyl sulfoxide [6], and trimethylamine [7] to achieve simultaneous removals of carbon and nitrogen within a single-pass process. The application of EMMC offers several advantages over conventional ASP and A2O including higher biomass density, lower effluent suspended solids, shorter start-up period, and lower operation/maintenance costs. Furthermore, immobilized biomass allows longer sludge retention time, enabling slow-growing bacteria to develop and adapt to complex organic waste streams.

Although the EMMC process has been successfully applied for various wastewaters, the mass transport and hydraulic advection influencing the biochemical reaction kinetics have not been fully investigated. In our previous work, a flat slab of EMMC was prepared to investigate the combined process of advection/diffusion/biological transformation in EMMC [8]. Hydraulic and kinetic parameters were obtained for the plate EMMC system. In this work, the mass transfer and biochemical reactions of carbon organics and nitrogen compounds are modeled for a spherical EMMC using the mass diffusion/advection and reaction kinetic parameters obtained in our previous work.

2. Biochemical reactions and model

2.1. Conceptual model

The mass transport of dissolved oxygen (DO), COD, and ammonia is via advection or diffusion through the EMMC balls, in which immobilized microorganisms utilize oxygen to decompose COD and nitrify NH_4^+-N. Jang et al. [9] indicated that DO and NH_4^+-N concentrations decreased gradually inward from the interface of the balls and water. Further into the pores of the EMMC ball, DO depletes and the anoxic and anaerobic zones develop. As a result, when the transfer of oxygen is limited toward the ball center, the processes gradually shift from nitrification to denitrification; the latter converts nitrate to nitrogen gas utilizing residual COD as the electron donor. For this reason, nitrification occurs only at the outer portion of the EMMC ball (Fig. 1). Fig. 1 provides a conceptual transition of target chemical species (O_2, NH_4^+, NO_2^-, and NO_3^-) and their concentration gradients within the immobilized biomass.

The EMMC ball carrier is an agglomeration of activated sludge with pores of various sizes. Nitrification and denitrification processes occur within a certain depth from the carrier/water interface. Therefore, the mass transfer resistance of solutes is greater at the boundary layer that limits the overall mass transfer. Wang et al. [10] and Yu et al. [11] studied simultaneous carbon and nitrogen removal from wastewater of different compositions, and Ng et al. [12] operated bio-entrapped membrane operation in different modes to treat wastewater of the food and beverage industry. The biochemical processes occurred at the inter macrospores between the carrier balls and the intra microspores within the carrier. The organics and nitrogen removal efficiencies are thus affected by water flowrate and mass transport by advection and diffusion. The conversion of dissolved substrates in the carrier is governed by the advective, diffusive, and reactive processes in the microspores.

This conceptual model provides insights into the transport and biochemical processes. Assuming a steady-state condition, the depletion of oxygen in the EMMC carrier dictates predominant biochemical transformations in it. For instance, if the radius of the EMMC ball is less than the critical distance denoting a certain oxygen deficit, the conversion process would primarily be aerobic or only partially anoxic. On the contrary, if the carrier radius is greater than the critical distance denoting significant oxygen deficit, the conversion processes in this region would be primarily be anoxic or anaerobic. In the proposed model, nitrites and nitrate in this region are assumed to be completely converted to nitrogen gas, that is, no constrains in the denitrification process.

2.2. Mathematical modeling of the EMMC ball

This section provides a mathematical model of the combined nitrification and denitrification reaction in

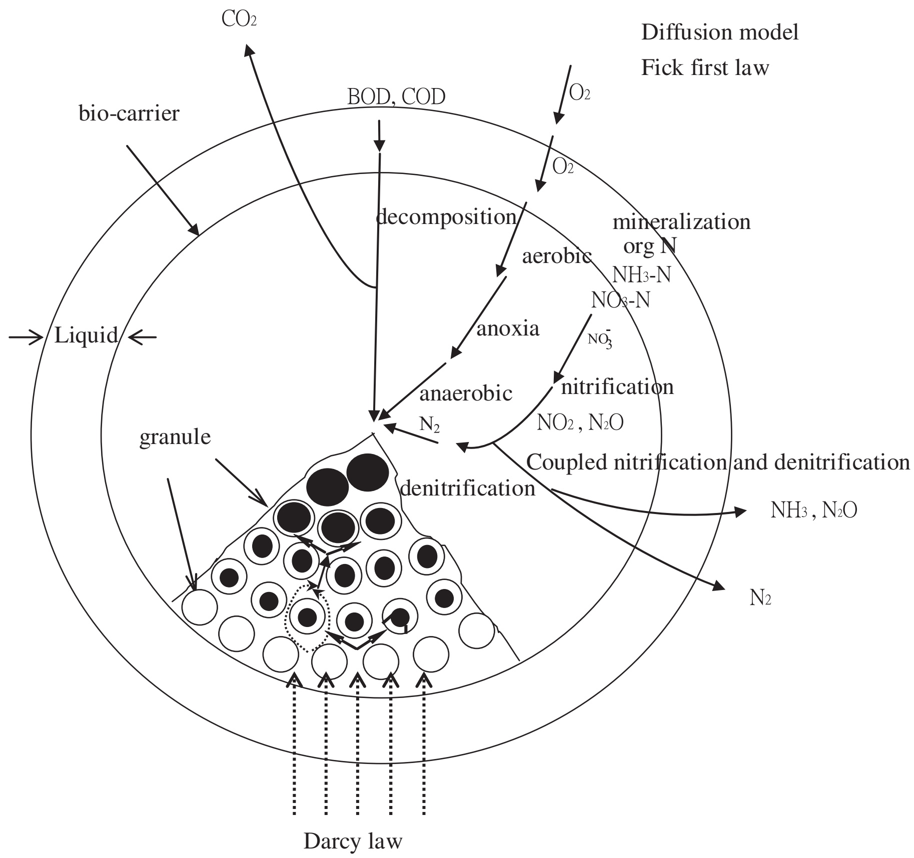

Fig. 1. Schematics illustrating mass transfer in an immobilized microbial cell carrier.

EMMC carrier of spherical shape. The mass transport of DO, NH_4^+-*N*, and COD will be described by one-dimensional advection/diffusion/reaction equations:

$$\frac{\partial C_{\mathrm{COD}}}{\partial t} = -\frac{\partial U C_{\mathrm{COD}}}{\partial x} - k_d\left(\frac{C_{\mathrm{DO}}}{C_{\text{h-s,COD}} + C_{\mathrm{DO}}}\right)C_{\mathrm{COD}} + \frac{\partial}{\partial x}\left(E\frac{\partial C_{\mathrm{COD}}}{\partial x}\right) \quad (1)$$

$$\frac{\partial C_{\mathrm{NH_4-N}}}{\partial t} = -\frac{\partial U C_{\mathrm{NH_4-N}}}{\partial x} - k_n\left(\frac{C_{\mathrm{DO}}}{C_{\text{h-s,NH}_4\text{-N}} + C_{\mathrm{DO}}}\right)C_{\mathrm{NH_4-N}} + \frac{\partial}{\partial x}\left(E\frac{\partial C_{\mathrm{NH_4-N}}}{\partial x}\right) \quad (2)$$

$$\frac{\partial C_{\mathrm{DO}}}{\partial t} = -\frac{\partial U C_{\mathrm{DO}}}{\partial x} - k_d C_{\mathrm{COD}} - \frac{64}{14}k_n\, C_{\mathrm{NH_4-N}} + \frac{\partial}{\partial x}\left(E\frac{\partial C_{\mathrm{DO}}}{\partial x}\right) \quad (3)$$

where C_{COD} = COD concentration (mg/L); $C_{\mathrm{NH_4-N}} = NH_4^+ - N$ concentration (mg/L); C_{DO} = DO concentration (mg/L); U = water flow velocity (m/d); E = diffusion coefficient (m^2/d); $C_{\text{h-s,COD}}$ = deoxygenation half-saturation constant (mg O_2/L); $C_{\text{h-s,NH}_4\text{-N}}$ = nitrification half-saturation constant (mg O_2/L); k_d = deoxygenation coefficient (1/d); k_n = nitrification coefficient (1/d).

At steady-state condition, the equations are rewritten as:

$$0 = -\frac{\partial U C_{\mathrm{COD}}}{\partial x} - k_d\left(\frac{C_{\mathrm{DO}}}{C_{\text{h-s,COD}} + C_{\mathrm{DO}}}\right)C_{\mathrm{COD}} + \frac{\partial}{\partial x} \times \left(E\frac{\partial C_{\mathrm{COD}}}{\partial x}\right) \quad (4)$$

$$0 = -\frac{\partial U C_{\mathrm{NH_4-N}}}{\partial x} - k_n\left(\frac{C_{\mathrm{DO}}}{C_{\text{h-s,NH}_4\text{-N}} + C_{\mathrm{DO}}}\right)C_{\mathrm{NH_4-N}} + \frac{\partial}{\partial x}\left(E\frac{\partial C_{\mathrm{NH_4-N}}}{\partial x}\right) \quad (5)$$

$$0 = -\frac{\partial U C_{\mathrm{DO}}}{\partial x} - k_d\, C_{\mathrm{COD}} - \frac{64}{14}k_n\, C_{\mathrm{NH_4-N}} + \frac{\partial}{\partial x}\left(E\frac{\partial C_{\mathrm{DO}}}{\partial x}\right) \quad (6)$$

Analytical solutions for DO, COD, and NH_4^+-N concentrations are required for the transport-reaction equations but not obtainable because of varying substrate concentrations in the EMMC ball along the radial direction of the EMMC ball due to simultaneous diffusion and consumption of the substrates. Therefore, the numerical approach of a finite segment method was applied to solve for the concentration profiles of DO, COD, and NH_4^+-N along the radius of the EMMC ball. In each segment, the mass transport and biochemical reaction coefficients were defined and substituted in the equations for calculating the DO, COD, and NH_4^+-N concentrations in and out of each segment. The Cartesian coordinate system was used for constructing the general mass balance equation:

$$\frac{\partial C}{\partial t} = -\left(\frac{\partial U_x C}{\partial x} + \frac{\partial U_y C}{\partial y} + \frac{\partial U_z C}{\partial z}\right) + \frac{\partial}{\partial x}\left(E_x \frac{\partial C}{\partial x}\right) + \frac{\partial}{\partial y}\left(E_y \frac{\partial C}{\partial y}\right) + \frac{\partial}{\partial z}\left(E_z \frac{\partial C}{\partial z}\right) + S \quad (7)$$

where C = compound concentration (mg/L); X = x-direction (horizontal) axis (m); Y = y-direction (horizontal) axis (m); Z = z-direction (vertical) axis (m); U_x = velocity in the x-direction (m/s); U_y = velocity in the y-direction (m/s); U_z = velocity in the z-direction (m/s); E_x = diffusion coefficient in the x-direction (m^2/s); E_y = diffusion coefficient in the y-direction (m^2/s); E_z = diffusion coefficient in the z-direction (m^2/s); S = sink or source of the biochemical reactions (mg/L/s).

Assuming uniform one-dimensional flow in the x-direction only with reactions, Eq. (7) was simplified:

$$\frac{\partial C}{\partial t} + \frac{\partial U_x C}{\partial x} = \frac{\partial}{\partial x}\left(E_x \frac{\partial C}{\partial x}\right) + S \quad (8)$$

In Eq. (5), C was the state variable and by using given U_r and E_x, Eq. (9) is obtained with chain rule:

$$\frac{\partial C}{\partial t} + U_x \frac{\partial C}{\partial x} = E_x \frac{\partial^2 C}{\partial x^2} + S \quad (9)$$

For COD degradation and NH_4^+-N nitrification in the EMMC ball, the spherical coordinate system was employed to describe the mass transport and diffusion of COD and NH_4^+-N. The general mass balance equation was expressed as:

$$\frac{\partial C}{\partial t} = -\left(U_r \frac{\partial C}{\partial r} + U_\theta \frac{1}{r}\frac{\partial C}{\partial \theta} + U_\phi \frac{1}{r \sin\theta}\frac{\partial C}{\partial \phi}\right) + \frac{1}{r^2}\frac{\partial}{\partial r} \times \left(E_r r^2 \frac{\partial C}{\partial r}\right) + \frac{1}{r^2 \sin\theta}\frac{\partial}{\partial \theta}\left(E_\theta \sin\theta \frac{\partial C}{\partial \theta}\right) + \frac{1}{r^2 \sin^2\theta}\frac{\partial}{\partial \phi}\left(E_\phi \frac{\partial C}{\partial \phi}\right) \quad (10)$$

where C = compound concentration (mg/L); r = radius (m); Θ = azimuth angle (radian); Φ = polar angle (radian); U_r = radial velocity (m/s); U_θ = azimuth angle tangential velocity (m/s); U_φ = polar angle tangential velocity (m/s); E_r = radial diffusion coefficient (m^2/s); E_θ = azimuth angle tangential diffusion coefficient (m^2/s); E_φ = polar angle tangential diffusion coefficient (m^2/s); S = sink or source due to reactions (mg/L/s).

For one EMMC ball assuming uniform advection, diffusion, and biochemical reactions occurring along the radial direction, Eq. (10) becomes:

$$\frac{\partial C}{\partial t} = -U_r \frac{\partial C}{\partial r} + \frac{1}{r^2}\frac{\partial}{\partial r}\left(E_r r^2 \frac{\partial C}{\partial r}\right) + S \quad (11)$$

where C was defined as a state variable with given U_r and E_x, resulting by chain rule in:

$$\frac{\partial C}{\partial t} + U_r \frac{\partial C}{\partial r} = \frac{2E_r}{r}\frac{\partial C}{\partial r} + E_r \frac{\partial^2 C}{\partial r^2} + S \quad (12)$$

or:

$$\frac{\partial C}{\partial t} + \left(U_r - \frac{2E_r}{r}\right)\frac{\partial C}{\partial r} = E_r \frac{\partial^2 C}{\partial r^2} + S \quad (13)$$

Eq. (13) was solved with the WASP model code [13] in one dimension in terms of the spatial dimension r, with assigned mass transport coefficients U_r and E_r. The S term was quantified via a first-order decay reaction. For COD, the S term characterized the first-order decay of COD in the EMMC ball along the radial direction. Segmentation was achieved by dividing the radius into a finite number of sections, thereby creating finite layers from the surface to the center of the ball. Experimental data were used to derive the hydraulic conductivity for the modeling analysis. Subsequent model runs would eventually finalize the values of the mass transport coefficients.

Mass changes of DO, COD, and NH_4^+-N in the EMMC ball were simulated with boundary conditions at the EMMC ball–water interface: DO = 6 mg/L, COD = 300 mg/L, and NH_4^+-N = 27 mg/L. The kinetic model input was based on experimental results of our previous study [8]. Diffusion coefficients for calculating mass transport and biochemical reaction kinetics for COD degradation (k_d = 6.96 1/day) and ammonia nitrification (k_n = 1.08 1/day) were used in Eq. (13). The radius of the EMMC ball was divided into 10 segments for calculation of mass transport of DO, COD, and NH_4^+-N.

3. Results and discussion

The mass transport with concurrent biochemical reactions involving COD and nitrogen compounds within the EMMC medium is very similar to that of soil medium being irrigated with greywater. In soil environment, biological respiration depletes DO in the inter space of soil aggregates. It creates an anaerobic environment due to advective and diffusive mass transports through the macro and microspaces in the interior of soil aggregates, triggering coupled nitrification–denitrification processes [14]. A mathematical model of nitrification and denitrification has been developed to describe *C* and *N* transformations in a spherical biological carrier [14]. Most aerobic activities occurred at and near the surface of the EMMC balls, that is, COD was consumed and NH_4^+-N nitrified within a short distance into the balls. As the ball size increases, the ball's interior becomes inaccessible to the reach of oxygen; this creates an anaerobic zone that limits aerobic biodegradation and/or nitrification [14].

With mass transport and reaction parameters, biochemical transformations of carbon and nitrogen species along the EMMC ball were simulated. The boundary conditions at the solid–liquid interface were initial COD and bulk DO concentrations. Using the model described in Section 2, the concentration changes of COD, NH_4^+-N, and DO within the EMMC ball are plotted in Fig. 2. The rapid decrease in COD within a short distance (some 1–1.5 cm) from the water-ball interface indicates that anoxic and anaerobic zones develop readily within the EMMC medium. Su and Yu [15] classified aerobic granules of varied size fractions had varied component concentration profiles and respective reaction rates. Biological processes in aerobic granules are determined primarily by concentration gradients of oxygen and many other substrates. Factors that interact with one another such as diffusion, granule size, biomass load, and biodegradation can greatly influence removal of substrates and DO [16]. Mudliar et al. [17] discussed a region existing near the medium surface where liquid velocity was very low that gave rise to a near-stagnant film of liquid, which consequently provided maximum resistance to transfer of substrates to the medium surface. Mudliar et al. [17] reported that the entire biofilm would not be utilized for biochemical transformation when substrate diffusion became a limiting factor. The NH_4^+-N concentration profile is very similar to the COD profile as NH_4^+-N is nitrified in the presence of DO; it follows that nitrifying bacteria are distributed only near the surface of the EMMC ball where DO is available. In the presence of DO and substrates, the EMMC carriers have good contact with them at the outer region, where nitrification process is very active [12]. Our previous experimental results showed that the pH stayed constant around 7.1–7.4 at the influent side and 6.6–6.9 at the effluent side. The slow decrease in pH was attributed to gradual consumption of alkalinity by the nitrifying bacteria.

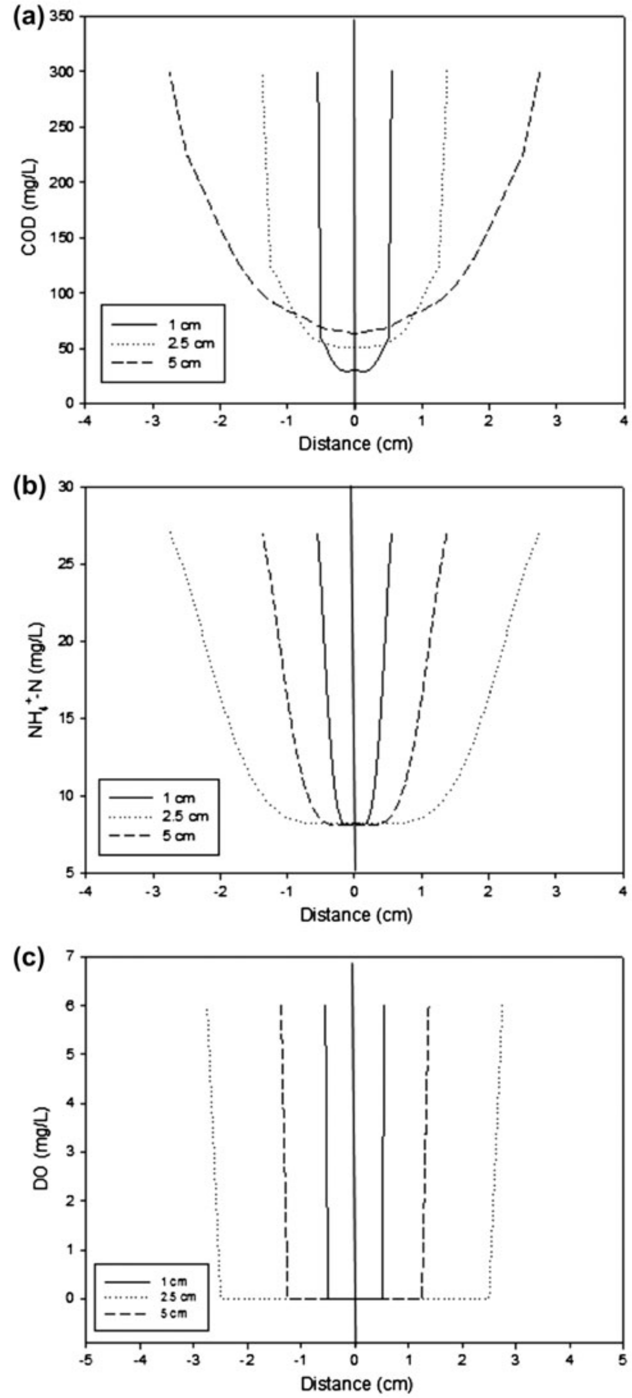

Fig. 2. Simulation of COD, NH_4^+-N and DO concentration profiles along the radius of the EMMC ball.

Fig. 2(c) indicates that the DO concentration decreases sharply to nearly zero. Therefore, it is clear that aerobic condition was predominant at the surface of the EMMC ball, and at several mm into the ball, anaerobic condition appeared due to diffusion limitation of oxygen and its depletion via COD biodegradation and NH_4^+-N nitrification. Jang et al. [9] studied the characteristics of aerobic granules with a DO microelectrode and reported that DO depletion and decrease in ammonia concentration within 1 mm from the water/granule interface. Unfortunately, microelectrodes are not feasible for hard EMMC balls in this study.

Many researchers [1,18–20] suggested that denitrification took place at the inner part of sludge flocs or biofilm under even highly aerated environment because of a very steep DO gradient within the flocs. The explanation was supported by Jang et al. [9] who found ammonia oxidizing bacteria primarily in the upper layers of the granule where DO could reach to support nitrification. The merit of coupling EMMC with traditional ASP is to create an A2O environment in an aeration tank. In other words, with EMMC one can upgrade the AS process into the A2O process without substantial facility changes to the existing aeration chamber. This modeling work shows the distribution of anoxic and anaerobic zones in the EMMC carriers, which can be incorporated to augment the functions of existing AS into those of A2O.

4. Conclusions

This research provided kinetic model simulation based on experimental results and modeling analysis from our previous study in order to examine the effects of immobilized biomass and mass transport on the concentration profiles of COD and NH_4^+-N within the EMMC ball. It also quantitatively modeled the hydraulic characteristics and biochemical reaction of the immobilized ASP for removal of COD and NH_4^+-N, offering insights to mass and oxygen transfer limitation in the EMMC process. The decrease in COD occurs within a very short distance (some 1–1.5 cm) from the water/EMMC ball interface, which indicates that anoxic and anaerobic zones develop readily within the EMMC ball. The change of NH_4^+-N concentration profile is very similar to that of the COD. Nitrification of NH_4^+-N occurs only in the presence of DO, which suggests the distribution of nitrifying bacteria primarily at the outer portion of the EMMC balls. The efficiencies of organics biodegradation and ammonia nitrification were not influenced by the diameter of the EMMC carrier. An EMMC carrier of 1 cm in diameter provides good biodegradation of the organics and nitrification/denitrification of the nitrogen compounds. Evidently, the dominating factor on the efficacy of organic biodegradation and nitrification in an immobilized-cell reactor is the diffusion limitation of DO.

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