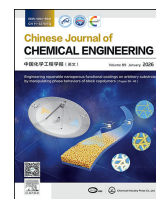




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Full length article

Particle-scale mass transfer characteristics of composite microspheres for cesium ion adsorption in nuclear wastewater within microchannel adsorbers

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ABSTRACT

To efficiently remove radioactive nuclides from nuclear industry wastewater and minimize the generation of radioactive secondary waste, this study proposes the concept of a magnetically controlled microchannel adsorber based on magnetic adsorbents. A novel protocol for achieving high adsorption performance in microchannel adsorbers with periodically distributed particles is developed using the particle-resolved computational fluid dynamics (CFD) method, which addresses the limitations of traditional porous media flow models. To align simulation results more closely with practical scenarios, a typical high-efficiency magnetic adsorbent, magnetic sodium alginate/cobalt-based Prussian blue (M-SA/PB-Co), was synthesized. The M-SA/PB-Co microspheres exhibit a uniform size distribution (300–600 μm), and their Cs^+ adsorption follows the pseudo-second-order kinetic model with a Langmuir saturated adsorption capacity of $124.84 \text{ mg} \cdot \text{g}^{-1}$. The performance parameters of M-SA/PB-Co, obtained from characterization and adsorption experiments, were integrated into CFD simulations. CFD results indicate that as the flow velocity increases, the flow field gradually transitions with vortices expanding in scale and streamline bifurcation points shifting rearward. The Cs^+ concentration decreases progressively along the flow direction, with a more pronounced reduction in the vortex regions downstream of particles. The characteristic velocity and characteristic concentration of specific regions surrounding the particles were extracted based on boundary layer distribution. The amount of concentration reduction of Cs^+ through particle is positively correlated with the characteristic concentration and negatively correlated with the characteristic velocity. The number of microspheres required in the microchannel adsorber was optimized using the response surface method. Compared with industrial fixed-bed adsorbers, microchannel adsorbers exhibit 8–10 times higher processing capacity, demonstrating significant industrial application potential.

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1. Introduction

Nuclear energy, as an efficient and low-carbon energy source, offers significant benefits in its utilization. However, the issue of radioactive contamination has become one of the major environmental challenges faced by numerous countries around the world [1]. In current treatment processes, adsorption technology has

emerged as the preferred method [2] due to its advantages such as lower investment costs, simpler operation, and easier automation [3], compared to evaporation [4], precipitation [5], and membrane separation [6] technologies. Nano-adsorbent materials, owing to their rapid adsorption kinetics and high adsorption capacity, have been extensively employed in adsorption studies [7]. However, nano-adsorbents in fine powder morphology are prone to agglomeration and with high pressure drop during separation applications [8]. To address these issues, they are commonly integrated with structural materials such as activated carbon [9], polyacrylonitrile (PAN) [10], sodium alginate (SA) [11], and chitosan (CTS) [12] to form composite microspheres for practical

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applications [13]. In practical applications, some composite microspheres exhibit insufficient mechanical strength, fragility, and poor material utilization efficiency [14]. Due to these issues, adsorbents often fail prematurely before reaching saturation—especially critical in nuclear wastewater treatment, where extremely low nuclide concentrations demand prolonged and efficient adsorption, thereby exacerbating secondary radioactive waste generation. Therefore, achieving efficient adsorption and minimizing secondary radioactive waste generation during adsorbent application has become the focus of attention [15].

Herein, the concept of magnetically controlled microchannel adsorbents with magnetic composite microsphere adsorbents was innovatively proposed. This design directly addresses the aforementioned issues. Magnetic control mitigates particle agglomeration and reduces mechanical load on microspheres. By dynamically adjusting the adsorption surfaces via magnetic control, it can significantly enhance adsorption efficiency and promote the particles to reach adsorption saturation to the maximum extent, thereby minimizing secondary radioactive waste. While composite microspheres have been studied in fixed-bed adsorbents, their dynamic behavior in microchannels remains poorly understood, which largely due to limitations in microscale flow detection techniques, which hinder investigations into fluid–particle interactions and local flow characteristics.

Computational fluid dynamics (CFD) simulation technology [16], demonstrates unique advantages, enabling precise resolution of multi-dimensional physical field characteristics at sub-millimeter spatial resolution and millisecond-level temporal steps. The porous media flow model is commonly employed to investigate fluid dynamics within adsorption bed layers [17]. Hussein *et al.* [18] and Vera *et al.* [16] investigated simulate the problem of transporting pollutants through a porous media bed in unsteady state case, the results of laboratory work were compared with the results of the CFD simulated model. However, conventional porous media flow models rely on simplified assumptions of fixed-bed geometries [19], sacrificing detailed local structures and flow field characteristics, particularly oversimplifying particle structural features. Such models struggle to accurately describe fluid flow and mass transfer in adsorbents with small tube-to-particle diameter ratios [20]. In contrast, particle-resolved computational fluid dynamics (PRCFD) has emerged as a high-fidelity approach for investigating fluid–solid particle interactions, enabling direct resolution of microscale flow dynamics around single particles [21]. By directly analyzing the microscopic motion of fluids and solid particles, the details of the flow field around the particles can be accurately captured [22]. Bouras *et al.* [23] used PRCFD to predict the pressure drop, liquid saturation, and wettability of different catalyst particle shapes in a trickle-bed reactor. Kutscherauer *et al.* [21] proposed a conjugate heat and mass transfer model for PRCFD simulations, coupling the flow of fluid through the fixed bed with the transfer and reaction in the porous catalyst. In view of the structural particularity and adsorption performance advantages of magnetically controlled microchannel adsorbents, it is necessary for us to establish a particle-scale CFD simulation method to provide a reliable theoretical basis for the optimal design and development of the device.

To better align simulation results with practical scenarios, we selected and synthesized a typical high-efficiency magnetic adsorption material and integrated the performance parameters of the composite microsphere adsorbent obtained from characterization and adsorption experiments into CFD model simulations. Radioactive cesium (^{137}Cs) is one of the primary pollutants in primary loop coolant waste water that draw significant attention in the treatment of nuclear industrial wastewater [24]. Thus, we choose Cs^+ as the typical target. The core adsorbents for Cs^+ are

nano-Prussian blue (PB) [25,26], nano-ammonium phosphomolybdate (AMP) [27], and nano-graphene oxide (GO) [28]. Transition metal-substituted Prussian blue analogs (PBA) also exhibit adsorption properties similar to or even superior to those of PB, and thus have been widely studied [29,30].

This study innovatively integrates magnetic composite microspheres with magnetically controlled microchannel adsorbents. Cobalt-based Prussian blue (PB-Co) was selected as the core material, SA as the base material, and magnetic powder was incorporated to synthesize the composite microsphere adsorbent (M-SA/PB-Co) as the typical adsorption material. The adsorption capacity and adsorption kinetic parameters of this adsorbent were experimentally studied. The PRCFD method was adopted, coupled with the adsorption experimental parameters, to conduct a numerical simulation study on the adsorption characteristics of Cs^+ in nuclear wastewater by microspheres in microchannel adsorbents at the particle scale. The response surface method (RSM) was applied to analyze the interactive effects of process conditions, thereby providing a theoretical basis for the application of composite microsphere adsorbents in microchannel adsorbent systems.

2. Experimental

2.1. Preparation of M-SA/PB-Co

2.1.1. Materials

Potassium ferrocyanide trihydrate ($\text{K}_4\text{Fe}(\text{CN})_6 \cdot 3\text{H}_2\text{O}$) was obtained from Tianjin Guangfu Fine Chemical Research Institute, China. Sodium alginate was sourced from Aladdin Chemical Co., Ltd., China. Cesium standard solution was procured from Merck KGaA., Germany. Cobalt(II) nitrate hexahydrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), calcium chloride (CaCl_2), ammonium hydroxide (25% (mass), $\text{NH}_3 \cdot \text{H}_2\text{O}$), iron(II,III) oxide (Fe_3O_4), and hydrochloric acid (HCl) were purchased from Kemeiou Chemical Reagent Co., Ltd., China. All chemicals were of analytical purity.

2.1.2. Preparation of PB-Co nanoparticles

Based on the optimized preparation methods established in the earlier research of our research group [31], $0.1 \text{ mol} \cdot \text{L}^{-1}$ $\text{K}_4\text{Fe}(\text{CN})_6$ and $0.1 \text{ mol} \cdot \text{L}^{-1}$ $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ solutions were prepared. The two reactant solutions were served as the continuous phase and dispersed phase respectively pumped into the channels of the microreactor. After rapid mixing and reaction, the generation of precipitated product was collected in a beaker, aged at room temperature for 24 h, washed several times by deionized water and finally dried under vacuum at 60°C for 24 h to obtain PB-Co nanoparticles.

2.1.3. Preparation of M-SA/PB-Co

SA (1.0% (mass)) was dissolved in deionized water with vigorous stirring to obtain a SA aqueous solution. Subsequently, PB-Co (10% (mass)) and Fe_3O_4 (5% (mass)) were dispersed into the solution and ultrasonicated for 3 h to obtain a homogeneous dispersion [32]. A 10.0 ml syringe filled with the dispersion was installed on the syringe pump of the electrospray machine. The positive electrode (12 kV) was clamped at the syringe needle. Then, the dispersion was sprayed into 0.5% CaCl_2 aqueous solution to form the microspheres. Finally, the microspheres were entirely solidified for 2 h, washed several times with deionized water, and then freeze-dried to obtain the M-SA/PB-Co microspheres.

2.2. Characterizations

The microscopic morphology of PB-Co nanoparticles and M-SA/PB-Co microspheres was observed using a scanning electron

microscope (SEM, QUANTA 450, FEI, USA) operated at an accelerating voltage of 20 kV. Transmission electron microscopy (TEM) images were acquired using a Tecnai F30 electron microscope (Philips-FEI, Netherlands). X-ray diffraction (XRD) patterns were obtained using a D/MAX-2400 X-ray powder diffractometer (Rigaku, Japan). The particle size distributions were obtained by Litesizer 500 nanoparticle and Zeta potential analyzer (Anton Paar, Austria).

2.2.1. Characterization of PB-Co nanoparticles

Fig. 1(a) presents the PB-Co particles exhibit excellent dispersibility, with a smaller particle size and a narrower size distribution ranging from 80 to 250 nm. Particles with sizes between 100 and 120 nm are the most predominant. From the XRD patterns in Fig. 1(c), it is evident that the diffraction peaks match well with the standard card (JCPDS). The characteristic peaks at 2θ values of 17.5° , 24.5° , 35.0° , 39.5° , and 43.5° are distinct, corresponding to the cubic crystal structure of PB-Co. The high intensity of the diffraction peaks indicates that the PB-Co nanoparticles possess high crystallinity. A slight shift of the main peak towards lower 2θ angles was observed. It speculated to result from a slight lattice expansion caused by the Co^{2+} doping process [33].

2.2.2. Characterization of M-SA/PB-Co

Fig. 2(a) and (b) present the optical images and diameter distribution of wet M-SA/PB-Co microspheres. The results demonstrate that the wet M-SA/PB-Co microspheres exhibit excellent monodispersity, with 92.31% of the microspheres having a diameter distribution between 600–800 μm . As shown in Fig. 2(c), the diameter of dried M-SA/PB-Co microspheres is approximately 500–600 μm , indicating a reduction of about 250 μm compared to their wet state. Furthermore, the homogeneous distribution of these elements in Fig. 2(d) confirms the uniform incorporation of PB-Co and Fe_3O_4 within the M-SA/PB-Co microspheres [34].

2.3. Adsorption experiments

Batch adsorption experiments were conducted to evaluate the adsorption kinetics and isotherms of Cs^+ on M-SA/PB-Co microspheres. Specifically, 10 ml of cesium standard solution with concentrations ranging from 1 to 300 $\text{mg}\cdot\text{L}^{-1}$ was added to vials containing 0.015 g of M-SA/PB-Co microspheres. The vials were then placed in a constant temperature shaker (IKA KS 300 IC control) and agitated at $250\text{ r}\cdot\text{min}^{-1}$ and 30°C . After adsorption, the concentration of Cs^+ in the solution was quantified using an atomic absorption spectrophotometer (AA-Solaar 969 MK11, Thermo Scientific, USA). The amount of Cs^+ adsorbed by the microspheres (Q_e , $\text{mg}\cdot\text{g}^{-1}$) and the removal efficiency (E , %) were calculated using Eqs. (1) and (2) respectively [35].

$$Q_e = \frac{(C_0 - C_e)V_{\text{aq}}}{m} \quad (1)$$

$$E = \frac{C_0 - C_e}{C_0} \times 100\% \quad (2)$$

where C_0 ($\text{mg}\cdot\text{L}^{-1}$) and C_e ($\text{mg}\cdot\text{L}^{-1}$) is the concentrations of Cs^+ before and after adsorption in the solution, V_{aq} (L) is the volume of the solution and m (g) is the mass of the microspheres.

2.3.1. Adsorption kinetics

A series of experiments were conducted to investigate the kinetics of Cs^+ adsorption by removing Cs^+ at different time intervals in 100 $\text{mg}\cdot\text{L}^{-1}$ Cs^+ solution. As shown in Fig. 3(a), the adsorption capacity increased rapidly within the first 10 min, followed by a gradual slowdown, eventually reaching equilibrium after approximately 4 h. To further analyze the kinetic experimental data, pseudo-first-order (PFO) and pseudo-second-order (PSO) kinetics models were applied. The linear forms of these models can be expressed as Eq. (3) [36] and Eq. (4) [37], respectively.

$$\ln(Q_e - Q_t) = \ln Q_e - k_1 t \quad (3)$$

$$\frac{t}{Q_t} = \frac{1}{k_2 Q_e^2} + \frac{t}{Q_e} \quad (4)$$

where Q_t ($\text{mg}\cdot\text{g}^{-1}$) is adsorption amount at t (min) and Q_e ($\text{mg}\cdot\text{g}^{-1}$) denotes adsorption amount at equilibrium. k_1 (min^{-1}) and k_2 ($\text{g}\cdot\text{mg}^{-1}\cdot\text{min}^{-1}$) are the rate constants of the PFO and the PSO kinetics model, respectively. The plots of the two kinetic models are presented in Fig. 3(b) and (c). It is observed that the PSO model exhibits a higher R^2 value of 0.9988, indicating a better fit to the experimental data. Additionally, the calculated adsorption capacity ($Q_{e,\text{pso}} = 61.73\text{ mg}\cdot\text{g}^{-1}$) closely aligns with the experimental adsorption capacity ($Q_{e,\text{exp}} = 60.67\text{ mg}\cdot\text{g}^{-1}$). These findings suggest that the adsorption of Cs^+ on M-SA/PB-Co microspheres is predominantly governed by a chemisorption process, highlighting the strong chemical interactions between Cs^+ ions and the adsorbent surface [38].

2.3.2. Adsorption isotherms

The effect of initial Cs^+ concentration (ranging from 1 to 300 $\text{mg}\cdot\text{L}^{-1}$) on the adsorption capacity of Cs^+ on M-SA/PB-Co microspheres is illustrated in Fig. 4(a). To determine the maximum adsorption capacity, Langmuir and Freundlich isotherm models were employed. The linear forms of these models are expressed as Eqs. (5) and (6) [32], respectively.

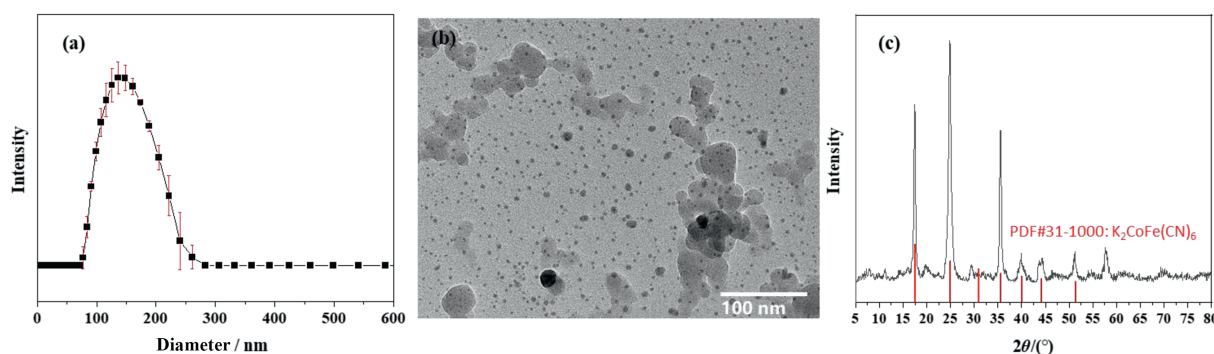


Fig. 1. (a) The particle size distributions (SD, $n = 3$), (b) corresponding TEM images of PB-Co nanoparticles, (c) XRD patterns of PB-Co nanoparticles.

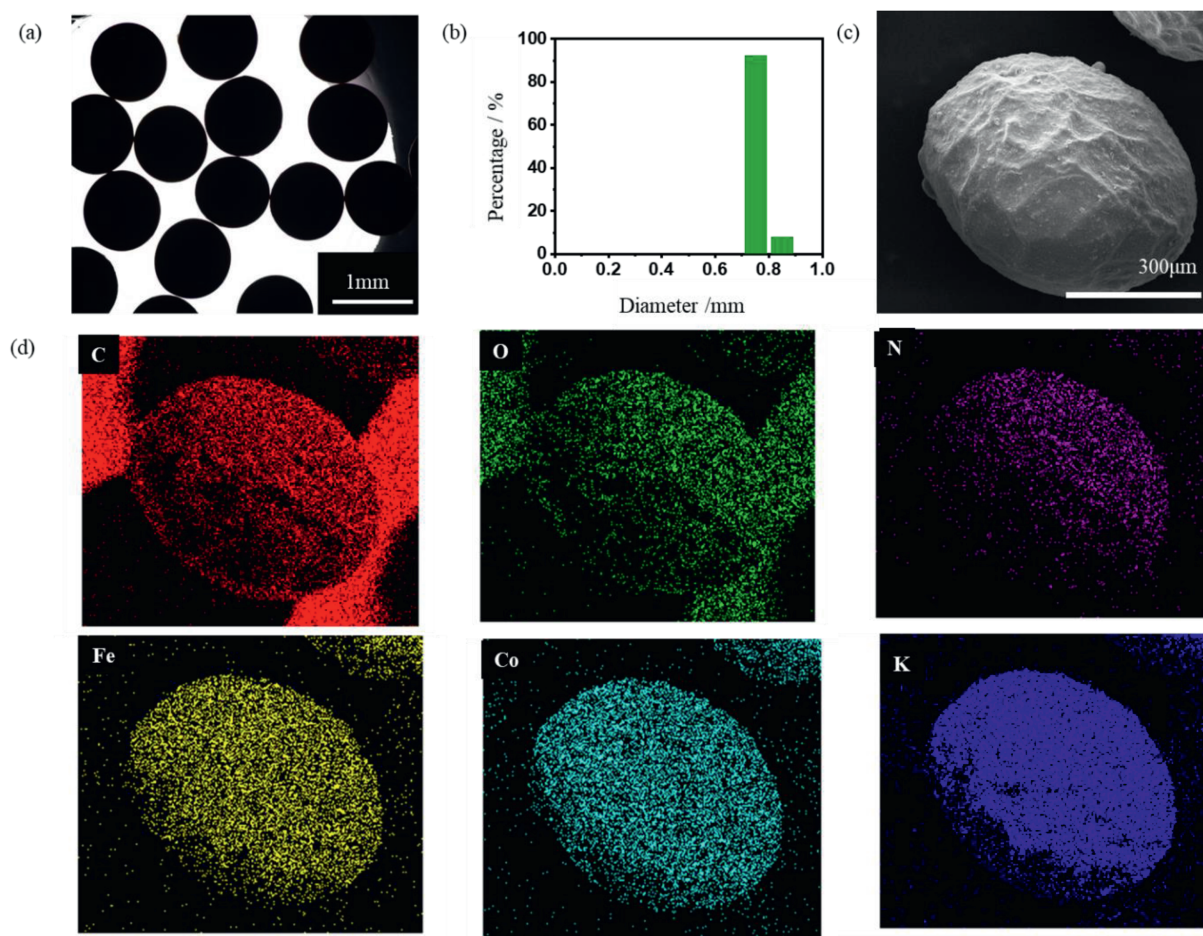


Fig. 2. (a) Optical image and (b) corresponding diameter distribution of wet M-SA/PB-Co microspheres; (c) SEM images and (d) elemental mapping images of dried M-SA/PB-Co microspheres.

$$\frac{C_e}{Q_e} = \frac{1}{bQ_m} + \frac{C_e}{Q_m} \quad (5)$$

$$\ln Q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (6)$$

Here, Q_m ($\text{mg} \cdot \text{g}^{-1}$) represents the adsorption amount at equilibrium, and b ($\text{L} \cdot \text{mg}^{-1}$) is the Langmuir constant associated with the affinity of binding sites [2]. Additionally, K_f ($\text{L} \cdot \text{g}^{-1}$) is the Freundlich constant related to adsorption capacity, and n ($\text{g} \cdot \text{L}^{-1}$) is the Freundlich exponent, which reflects the adsorption intensity and heterogeneity of the adsorbent surface.

From the linear isotherm plots presented in Fig. 4(b) and (c), the higher R^2 indicates that the Langmuir model better describes the adsorption process. This result suggests that the adsorption sites are uniformly distributed on the surface of M-SA/PB-Co microspheres, resulting in monolayer adsorption of Cs^+ . According to the Langmuir model, the maximum adsorption capacity (Q_m) was determined to be $124.84 \text{ mg} \cdot \text{g}^{-1}$.

Table 1 presents a comparative analysis of the adsorption capacities for Cs^+ between M-SA/PB-Co and other microspheres reported in literatures, such as PB-Co on phosphogypsum (PB-Co@PG) [26], ammonium molybdophosphate–polyacrylonitrile

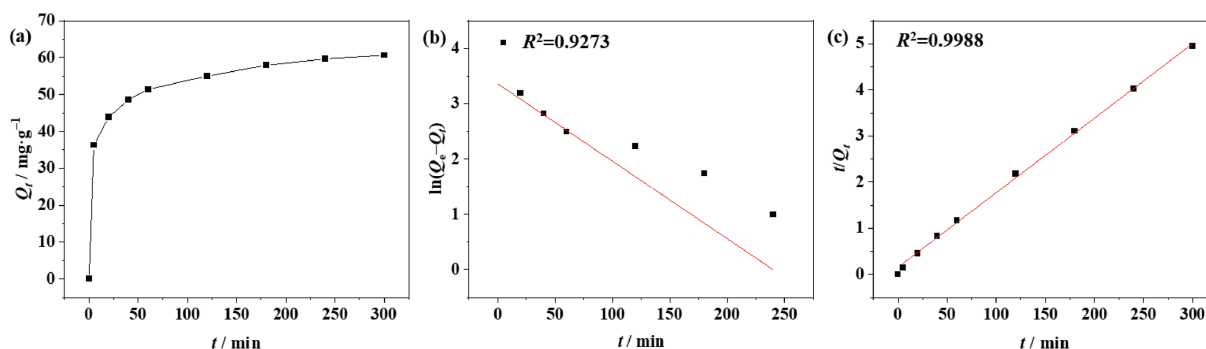


Fig. 3. (a) Effects of contact time on the adsorption capacity of Cs^+ on M-SA/PB-Co microspheres; linear fitting of (b) PFO and (c) PSO kinetic models.

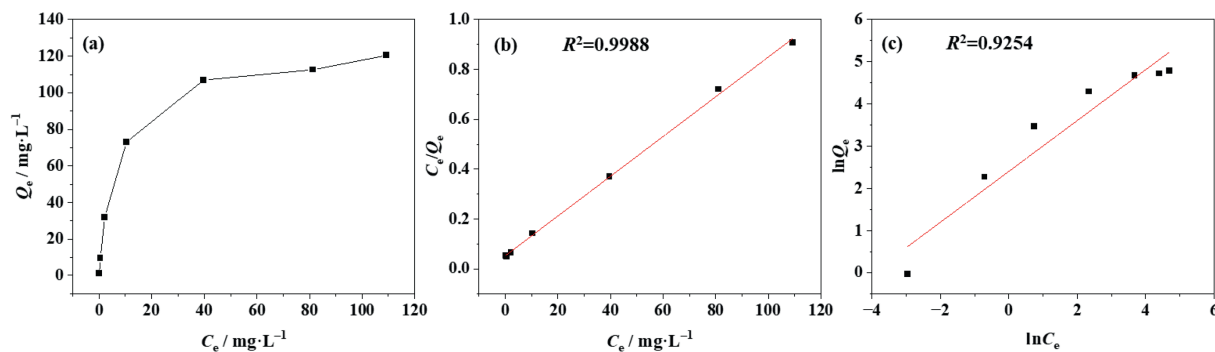


Fig. 4. (a) Effect of Cs⁺ equilibrium concentration on adsorption; adsorption isotherm fittings: (b) linear Langmuir (first two points exhibit part overlap), and (c) Freundlich models.

Table 1

The adsorption capacities for Cs⁺ by different adsorbents.

Adsorbents	$k_2 \times 10^5/\text{g} \cdot \text{min}^{-1} \cdot \text{mg}^{-1}$	$Q_m/\text{mg} \cdot \text{g}^{-1}$	References
M-SA/PB-Co	168	124.84	This work
KNiCF-GAC	97210	163.9	[40]
PB-Co@PG	213.3	135.15	[26]
Expanded perlite/AMP	6070	13.21	[39]
CMC/PB-Cu/PEG	106	149.80	[35]
@CuHCF/P	4.32	131.10	[38]
CMC/PB-La	500	35.20	[34]
AMP-PAN-N20	—	55.05	[41]

on expanded perlite (expanded perlite/AMP) [39], and other PBA composite microspheres. The results demonstrate that M-SA/PB-Co prepared in our work exhibits a high adsorption capacity, highlighting its potential as a highly effective adsorbent for Cs⁺ removal. This superior performance positions M-SA/PB-Co as a promising candidate for applications in Cs⁺ adsorption.

3. Mathematical Model

CFD simulations can provide multi-dimensional quantitative details in terms of temporal and spatial resolutions [42], offering an effective technical approach for investigating microsphere applications in microchannel adsorbents. To obtain the local flow characteristics in microchannels and retain the particle-scale features, a structural form of microchannel adsorbent was proposed, in which microspheres are fixed on the wall in a semi-embedded manner. Based on the PRCFD theory, a two-dimensional symmetric model of the microchannel was established. The model was verified and the grid independence was validated. The mass transfer user-defined function (UDF) source terms, derived from the experimentally parameters, was applied to investigate adsorption performance.

3.1. Basic assumptions and governing equations

To facilitate solving the physical problem, we assume that the dominant flow in the microchannel is laminar and the medium is incompressible (Assumption I). Therefore, the mass conservation equation of the model is shown in Eq. (7).

$$\nabla \cdot \vec{u} = 0 \quad (7)$$

The distribution of Cs⁺ within the phase is governed by the convection-diffusion equation (Eq. (8)).

$$\frac{\partial C}{\partial t} + \nabla \cdot (\vec{u}C) = \nabla \cdot (D\nabla C) \quad (8)$$

where D (m²·s⁻¹) is the diffusion coefficient of Cs⁺ in the liquid phase, C (g·g⁻¹ = 10⁶ mg·L⁻¹) is the mass fraction of Cs⁺, and $\vec{u}$ (m·s⁻¹) is the velocity vector.

During the adsorption process, the particle-liquid interface exhibits no slip velocity. That is, the no-slip wall boundary condition is applied (Assumption II). Additionally, the adsorption mechanism of Cs⁺ on microspheres is mainly ion exchange and surface adsorption [33]. We ignore the pore diffusion inside the particles (instantaneous diffusion). The adsorption can be realized in the form of the mass flux of Cs⁺ on the particle surface. We assume that the adsorption process exclusively occurs within the first layer of grid cells adjacent to the fluid-wall (particle surface) interface, with uniform adsorption capacity $S_i = S$ (kg·m⁻³·s⁻¹) (Assumption III) assigned to each grid, which can be calculated by Eq. (9).

$$\iiint r dV = \bar{r} V_{\text{bead}} = S V_{\text{first layer}} = \iiint S_i dV_i \Rightarrow S = \bar{r} V_{\text{bead}} / V_{\text{first layer}} \quad (9)$$

where V_{bead} and $V_{\text{first layer}}$ (m³) represent the volume of spherical particles and the total volume of the first-layer grid cells, respectively, $\bar{r}$ (kg·m⁻³·s⁻¹) denotes the average adsorption rate.

The linear driving force (LDF) model is typically employed to describe the mass transfer rate of solutes from the fluid phase to the solid phase during the adsorption process. However, its core assumption is that the transfer rate is proportional to the concentration difference between the fluid phase and the solid phase, which does not fully conform to the adsorption kinetics observed in experiment [43]. As shown in our study, the adsorption rate of Cs⁺ by M-SA/PB-Co particles follows the PSO kinetics [2]. Therefore, the adsorption rate in this study was derived from experimentally obtained adsorption isotherm equations and PSO kinetic equations, with the detailed derivation outlined as follows. In the microchannel adsorbent, the solution flows continuously, and for each microsphere particle, the adsorption rate is directly related to the concentration of the solution it contacts. According to the PSO kinetic formula Eq. (4), the initial adsorption rate ($\bar{r}$) is the rate at the time limit $t \rightarrow 0$, as mathematically expressed in Eq. (10).

$$v_0 = k_2 Q_e^2 \quad (10)$$

Previous studies have demonstrated that the Langmuir isotherm and the PSO kinetic model exhibit a mutual derivation relationship under conditions of extremely low equilibrium concentration [44]. In nuclear industry applications, the adsorption of Cs⁺ typically occurs at ultralow equilibrium concentrations C_e . For instance Ref. [45], when the activity concentration of incoming wastewater reaches 10⁶ Bq·L⁻¹, the corresponding mass

concentration of Cs^+ is approximately $9.41 \times 10^{-9} \text{ g} \cdot \text{g}^{-1}$, which aligns with the prerequisite for this derivation. The integral form of the Langmuir isotherm equation is provided in Eq. (11) [46].

$$Q_e = \frac{Q_m b C_e}{1 + b C_e} \quad (11)$$

By substituting Eq. (11) into Eq. (10), and expanding the expression, the initial adsorption rate formula for the microsphere is derived, as presented in Eq. (12).

$$\bar{r} = \frac{k_2 Q_m^2 b^2 C_0^2}{(1 + b C_0)^2} \quad (12)$$

where $C_0 (\text{g} \cdot \text{g}^{-1})$ represents the solution concentration in contact with each microsphere. By incorporating experimentally determined parameters k_2 , Q_m , b , the initial adsorption rate expression of the microspheres is derived and implemented as a UDF source term for numerical simulations.

3.2. Computational domain and boundary conditions

To study the adsorption characteristic of Cs^+ by composite microspheres in the microchannel adsorber, a two-dimensional symmetric model was established, as illustrated in Fig. 5. The typical hydraulic diameter range of microchannel reactors is 0.1 mm to 1 mm [47]. Considering the practical application requirements in nuclear industry wastewater treatment, the microchannel adsorption section in this study is controlled as 0.9 mm. The size of the computational domain is $6.3 \text{ mm} \times 0.75 \text{ mm}$ ($10.5d \times 1.25d$, where $d = 0.6 \text{ mm}$ is the diameter of the microsphere), incorporating four semi-embedded hemispherical microspheres on the channel walls. The distance between the centers of microspheres on the same side is $L_x = 2.5d$. The velocity inlet boundary condition is applied at the $x = 0$ plane; the pressure outlet boundary condition is set at the plane of $x = 6.3 \text{ mm}$; symmetry boundary is imposed at the $y = 0.75 \text{ mm}$ plane; and no-slip wall boundaries are assigned to all remaining surfaces. In addition, Fig. 5 shows the details of the local grid distribution. The size of the first layer of the grid on the surface of the microsphere is $0.5 \mu\text{m}$, the basic grid size is $7.5 \mu\text{m}$, and the total number of grids is 113440.

3.3. Model implementation

The model was implemented using the ANSYS Fluent solver with the coupled algorithm for steady-state simulations, where

the residual convergence criterion was set to 1×10^{-5} for all governing equations. The mass source term representing adsorption on microsphere surfaces was incorporated into the surface grids via UDF. In terms of the discretization schemes, the second-order implicit scheme is adopted for the time term; the second-order central difference is used for the pressure–velocity coupling; and the third-order MUSCL scheme is applied to both the momentum equation and the convective terms.

3.4. Grid independence study

The accuracy of the proposed model was validated through a diffusion problem in a semi-infinite long tube. As shown in Fig. 6 (a), the computational domain and boundary conditions for the validation case were configured with a uniform grid size of $7.5 \mu\text{m}$. Comparative analysis in Fig. 6(b) demonstrates excellent agreement between the transient concentration profiles obtained from CFD simulations and analytical solutions computed via MATLAB, thereby confirming the model's accuracy. On this basis, we further conducted the grid independence verification. As shown in Fig. 6(c), the sampling point is the plane at $x = 3.15 \text{ mm}$ (mid-channel position). The velocity magnitude and concentration are non-dimensionalized based on the inlet velocity and inlet concentration, respectively. The results revealed that the velocity magnitudes calculated using three different grid sizes are almost the same, and there are slight differences in the concentration distributions. Considering both computational efficiency and accuracy comprehensively, the final grid size was determined to be $7.5 \mu\text{m}$.

4. Results and Discussion

4.1. Fluid flow and adsorption mass transfer effects in the microchannel adsorber

Fig. 7 shows the diagrams of the first three microspheres under different flow velocities and initial concentration conditions. In terms of the flow field distribution characteristics, due to the obstruction of the microsphere particles, the fluid velocity near the surface of the particles is significantly reduced. However, in the top area of the particles, the flow velocity relatively increases because of the reduction in the cross-sectional area of the flow channel. The flow velocity reaches its maximum value at the symmetric plane, while the flow velocity in the tail area of the particles and the area near the wall is the lowest.

The formation and evolution of vortices are influenced by variations in flow velocity. The particle Reynolds number ($Re_p =$

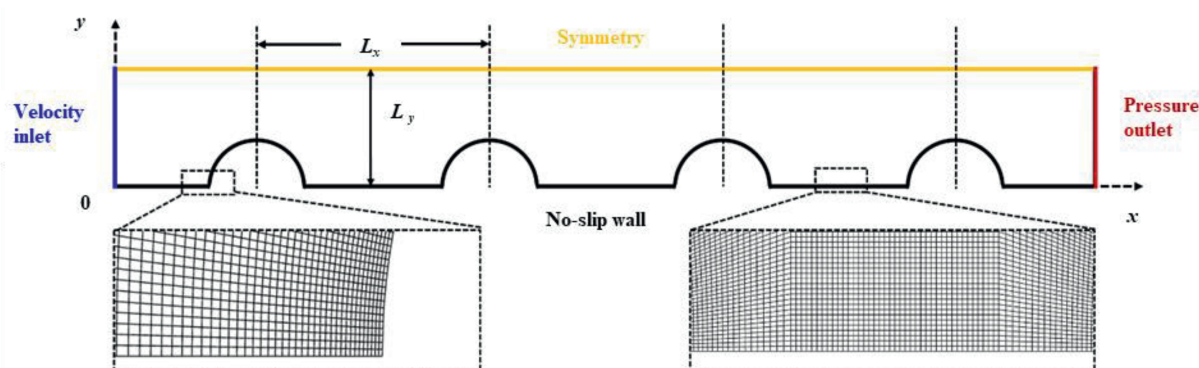


Fig. 5. Schematic of computational domain and boundary conditions.

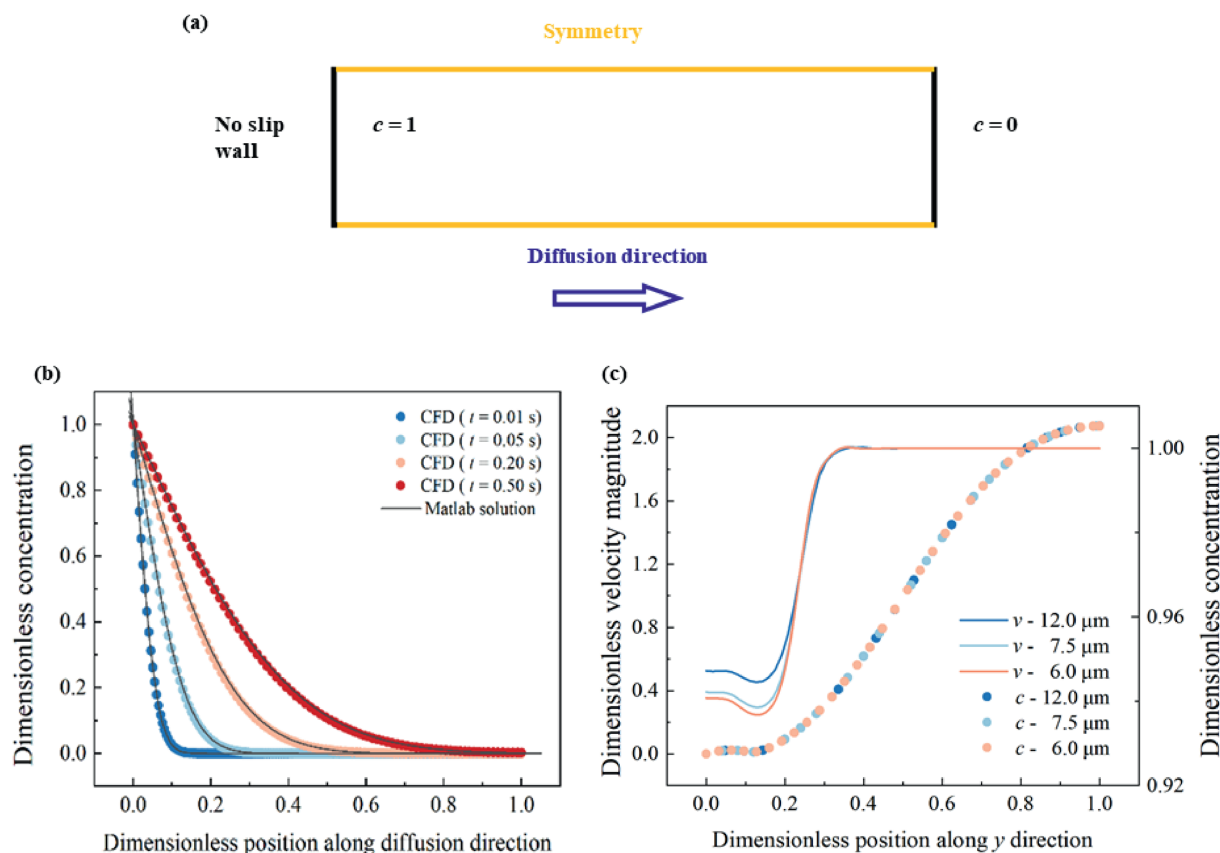


Fig. 6. (a) Computational domain and boundary conditions of verification model, (b) concentration distribution varies with time, (c) grid independence verification of velocity and concentration.

$\rho v d_p / \mu$) is used to characterize the relative motion characteristics between the particles and the fluid. Where, ρ ($\text{kg} \cdot \text{m}^{-3}$) is the fluid density, v ($\text{m} \cdot \text{s}^{-1}$) is the relative velocity between the particles and the fluid, d_p (m) is the diameter of the particles, and μ ($\text{Pa} \cdot \text{s}$) is the dynamic viscosity of the fluid. As can be seen from Fig. 7(a), under the condition of $0.01 \text{ m} \cdot \text{s}^{-1}$ ($Re_p = 6$), the flow exhibits a typical laminar regime. Fig. 8(a) reveals the vortex formation and boundary layer characteristics between particle2 and particle3. Only small-scale clockwise vortices (based on the bottom-view contour) are observed in the wake region of particles. As the flow velocity increases to $0.05 \text{ m} \cdot \text{s}^{-1}$ ($Re_p = 30$), larger-scale vortices formed in the tailing zone. Simultaneously, small clockwise vortices emerge near the flow entrance of the adjacent downstream particle (Fig. 7(b)). As shown in Fig. 8(b), flow recirculation begins to develop on the windward surface of particle3, accompanied by streamline bifurcation at the particle interface and gradual vortex formation. This transition indicates that the fluid flow pattern starts to transition from the advection state to the transitional flow state. When the flow velocity further increases to $0.1 \text{ m} \cdot \text{s}^{-1}$ ($Re_p = 60$), as can be seen from Fig. 7(c), a single clockwise vortex structure is formed between the particles, and the fluid gradually changes to the transitional flow state. Fig. 8(c) reveals the formation of a single clockwise vortex structure between particles, where the fluid in the recirculation zone exhibits cyclic motion, indicating a gradual transition to a transitional flow regime. Higher flow velocities induce a rearward shift of the streamline bifurcation point on the particle surface, signifying that enhanced inertial forces reduce the effective fluid-

particle contact area and consequently diminish particle utilization efficiency.

As can be seen from Fig. 7, as the fluid flows through each particle, the Cs^+ concentration progressively decreases. This reduction is particularly pronounced in the vortex regions downstream of the particles, may due to intensified convective transport under the vortex-induced flow conditions, which accelerates Cs^+ adsorption. In the Y-direction, the concentration distribution of Cs^+ exhibits a general consistency with the velocity profile: in the area with a higher flow velocity, the local concentration of Cs^+ is higher, while at the position with a lower flow velocity, the local concentration of Cs^+ is lower. This phenomenon arises because Cs^+ transport to particle surfaces is governed by external mass transfer mechanisms, forming a distinct concentration boundary layer (δ_c) around the particles. The δ_c and distribution of the boundary layer vary with the initial concentration of the fluid and the degree of turbulence.

4.2. Adsorption characteristics and influencing factors of single particle

The interaction between fluid flow and concentration distribution in microchannel adsorbers is highly complex. Since the particles are arranged periodically in the microchannel adsorber, particle2 and particle3 were selected as the research objects. The influence of the flow field and the concentration field on the adsorption effect of the particles were studied from the perspective of a single particle, so as to further guide the design of the adsorbers.

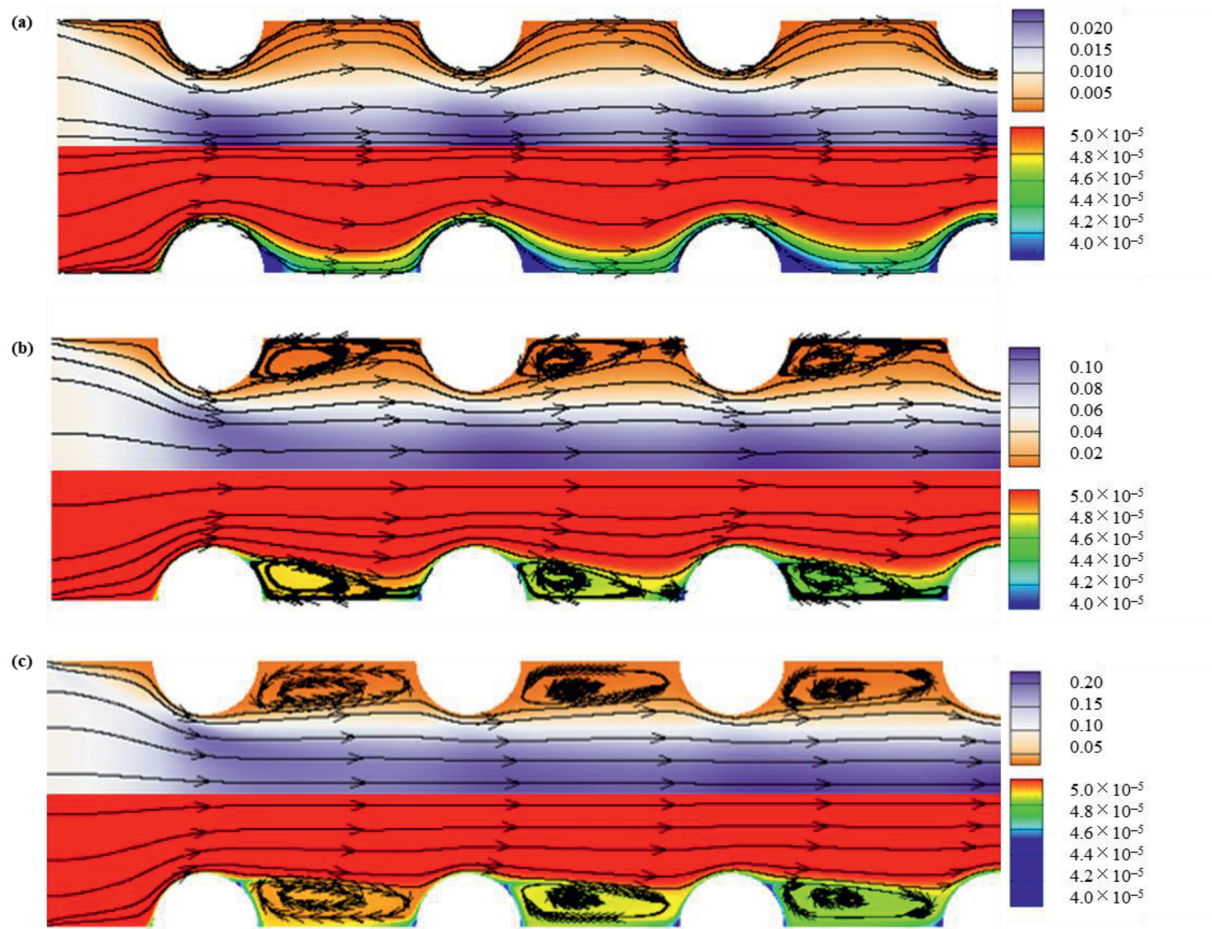


Fig. 7. Flow fields and concentration fields in a microchannel adsorber (lower panel: concentration contour plot, upper panel: velocity contour plot), $C_{in} = 5 \times 10^{-5} \text{ g} \cdot \text{g}^{-1}$: (a) $Re_p = 6$, (b) $Re_p = 30$, (c) $Re_p = 60$.

4.2.1. Characteristic parameters

To comprehensively analyze the variation patterns of single-particle utilization efficiency, it is critical to select appropriate characteristic velocities and concentrations, as the inlet velocity and inlet concentration of the microchannel may not directly govern single-particle adsorption dynamics. The δ_C represents the core region of effective mass transfer, serves as a key indicator for unveiling adsorption behavior under varying operational conditions. Since the δ_C at the top of the particle is between 0.01 and 0.10 mm in this study, a characteristic region with a distance of 0.1 mm from the top of the particle and $0.25d_p$ in front of the particle is selected as the characteristic

region, as shown in Fig. 9(a). The concentration of Cs^+ and the velocity of the fluid in the X-direction within the characteristic region are integrated and averaged, which are respectively defined as the characteristic concentration (C_{avg}) and characteristic velocity (v_{avg}) of the particle. The utilization efficiency (η) of the windward particle surface was quantified by the ratio of the effective adsorption arc angle (from the particle vertex to the flow separation point) to 90° . As can be seen from Fig. 9 (b), η decreases as the fluid flow velocity increases, and the influence of $Re_{p(avg)}$ calculated by the characteristic velocity on η is greater than that of $Re_{p(in)}$ calculated by the inlet flow velocity of the adsorber. Consequently, C_{avg} and v_{avg} are

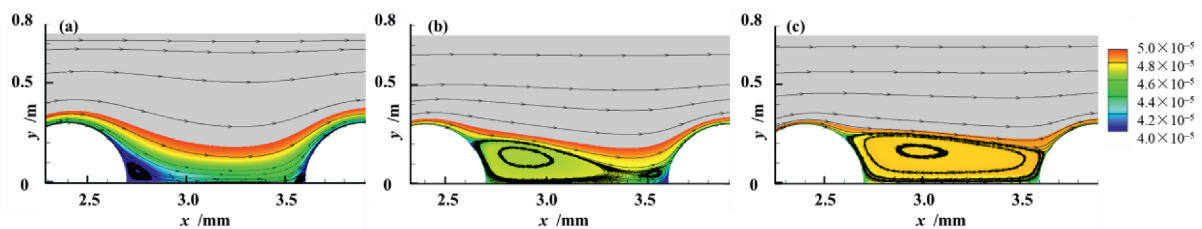


Fig. 8. Particle utilization efficiency and boundary layer profiles, $C_{in} = 5 \times 10^{-5} \text{ g} \cdot \text{g}^{-1}$: (a) $Re_p = 6$, (b) $Re_p = 30$, (c) $Re_p = 60$.

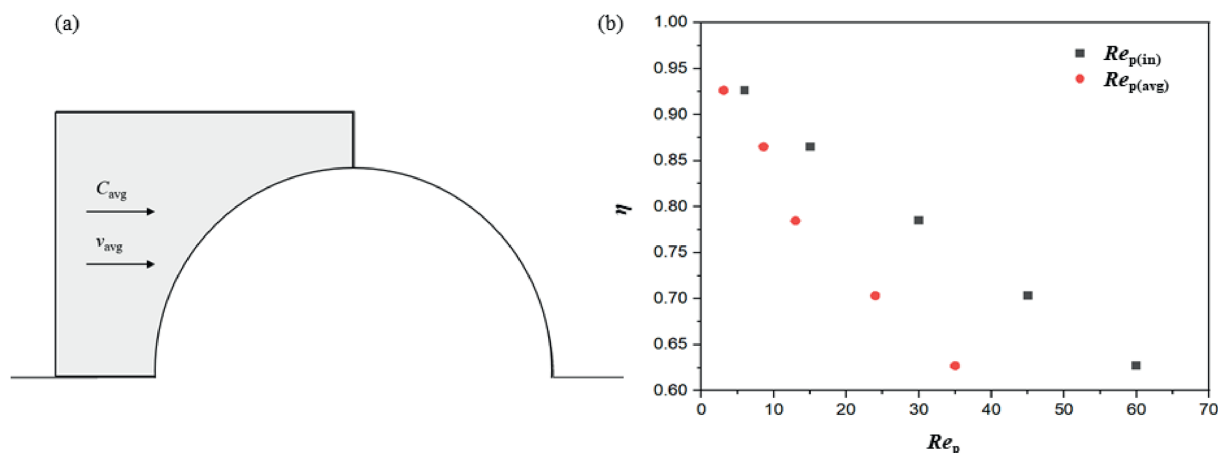


Fig. 9. (a) Schematic diagram of the characteristic region, (b) variation of particle utilization efficiency.

prioritized as governing parameters for single-particle adsorption studies.

4.2.2. Influence of characteristic parameters on δ_c

The influence of characteristic parameters C_{avg} and v_{avg} on δ_c was investigated, as shown in Fig. 10. Under constant Re_p conditions, higher C_{avg} values correlate with thinner δ_c (Fig. 10(a)), which is attributed to enhanced diffusive mass transfer rates driven by elevated initial Cs^+ concentrations. Additionally, Re_p increase under fixed C_{avg} conditions similarly with δ_c reduce, primarily stemming from intensified convective effects that amplify interfacial mass transfer efficiency. In conclusion, it can be seen that δ_c on the particle surface is governed by the synergistic effect of the concentration of Cs^+ around the particle and the convection.

By fitting the relationships between δ_c , C_{avg} and v_{avg} under multiple operational conditions, a predictive model 1 was derived, with detailed analysis of variance (ANOVA) parameters listed in Table 2 and visualized through response surface in Fig. 10(c). Statistical analysis revealed the model's high significance ($p < 0.0001$), demonstrating its capability to explain over 98% of δ_c variability ($R^2 = 0.9840$). It can be seen that convective flow enhancement outweighs concentration-driven diffusion in

thinning the δ_c , which aligns with Damköhler–Péclet scaling laws [48], provides a theoretical basis for the subsequent design and optimization of the microchannel adsorbers.

$$\begin{aligned} \delta_c = & 0.1007 - 1.5067v_{avg} - 615.11C_{avg} + 1129.01C_{avg}v_{avg} \\ & + 8.7975v_{avg}^2 + 2409268C_{avg}^2 \end{aligned} \quad (13)$$

4.2.3. Influence of characteristic parameters on the adsorption performance of a single particle

The influence of characteristic parameters on single-particle adsorption performance was investigated by analyzing the Cs^+ adsorption rate distribution across specific monitoring points (P_A : $x = 21$ mm, $y = 0$ mm; P_B : $x = 24$ mm, $y = 0.3$ mm; P_C : $x = 27$ mm, $y = 0$ mm) on the surface of particle2. As shown in Fig. 11(a), the adsorption rate distribution along the particle surface strongly correlates with variations in local concentration and velocity fields, driven by the velocity field's dominant role in governing Cs^+ transport to the particle interface. Specifically, the adsorption rates follow the order $P_B > P_C > P_A$, with P_B (located at the particle vertex) exhibiting the highest rate due to intensified convective mass transfer and minimized δ_c , while P_A (incident surface) and P_C (wake

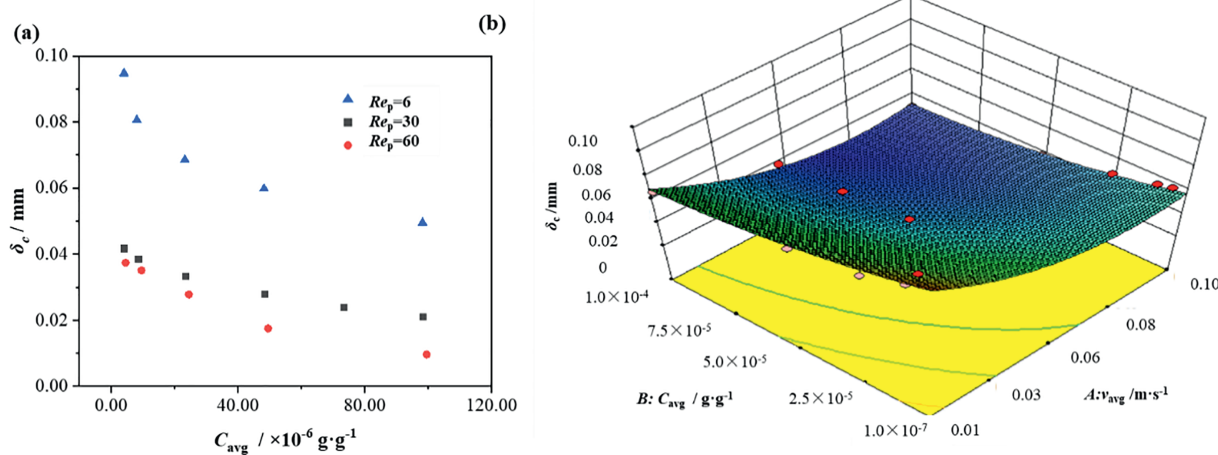


Fig. 10. Variations of δ_c with (a) v_{avg} and (b) C_{avg} . (c) Response surface of δ_c versus C_{avg} and v_{avg} .

Table 2
ANOVA table for the quadratic response surface model.

Source	df	Mean square	F value	p value prob > F
Model	5	0.001740	75.3	<0.0001
A- v_{avg}	1	0.004129	179.0	<0.0001
B- C_{avg}	1	0.001923	83.2	<0.0001
AB	1	3.12×10^{-5}	1.4	0.2699
A ²	1	0.001172	50.7	<0.0001
B ²	1	0.000297	12.8	0.0043
Residual	11	2.31×10^{-5}		
Cor total	16			

region) experience reduced rates owing to vortex-induced flow separation and diminished X-direction velocity. These findings underscore the criticality of optimizing fluid flow patterns, such as enhancing convective mixing via surface microstructures or suppressing recirculation zones through streamlined geometries.

The overall adsorption capacity of a single microsphere was evaluated using the Cs^+ concentration removal (ΔC) as the evaluation index, with systematic analysis revealing two critical trends show in Fig. 11(b). Under constant flow velocity, ΔC increases significantly with rising C_{avg} but plateaus when C_{avg} exceeds 25 $mg \cdot L^{-1}$, attributed to the transition from mass transfer limitation to adsorption saturation. Despite similar ΔC trends across Re_p , the maximum ΔC decreases as Re_p increases. This may be due to vortex-induced reduction in effective surface utilization and shortened residence time collectively suppress interfacial mass transfer efficiency. These findings highlight the need for C_{avg} – Re_p co-optimization in microchannel adsorbers to balance capacity and kinetics.

4.3. Performance prediction of microchannel adsorbers

In nuclear industrial applications, the decontamination factor ($DF = C_{in}/C_{out}$) serves as the critical performance metric for adsorbers, with an industrial threshold typically set at $DF > 100$. This study defines N_{100} as the number of microspheres required to achieve $DF = 100$. As demonstrated earlier, when C_{avg} exceeds 25 $mg \cdot L^{-1}$, adsorption saturation causes stagnation in ΔC , necessitating either increased particle count or flow-channel structural optimization to meet the target DF . Investigating the relationship between N_{100} and inlet flow conditions, as well as the evolution of δ_C from the inlet to outlet of the adsorber, is pivotal for balancing efficiency and scalability in reactor design.

Table 3
Parameters of the fitted relationship between ΔC and C_{avg} .

Re_p	a	b	R^2
3	4.44×10^{-7}	6.26×10^{-6}	0.9843
10	2.30×10^{-7}	2.87×10^{-6}	0.9823
30	1.61×10^{-7}	1.26×10^{-6}	0.9872

The relationship between ΔC and C_{avg} shows a saturation-growth trend, successfully fitted by the Michaelis–Menten model (Eq. (14)), with parameters detailed in Table 3. Particle velocity fields exhibit periodic distribution characteristics, so they all follow the adsorption kinetic equation under the same v_{in} . An iterative computational algorithm (C_{in} (particle n) – $\Delta C = C_{out}$ (particle n) = C_{in} (particle $n + 1$)) was employed to determine N_{100} , yielding the N_{100} – v_{in} – C_{in} regression model (Eq. (15)) with robust statistical validity. Its response surface and fitting diagnosis diagrams are shown in Fig. 12(a) and (b). The model predicts exponential scaling of adsorber size under high v_{in} – C_{in} combinations: N_{100} ranges 100–700 for $C_{in} > 20 \times 10^{-6} g \cdot g^{-1}$, but remains <100 for $C_{in} < 20 \times 10^{-6} g \cdot g^{-1}$, while increasing DF from 100 to 1000 only requires 10–50 additional particles. Boundary layer evolution analysis (Fig. 12(c)) demonstrates progressive thickening along the flow path, necessitating a gradually expanding channel design to balance δ_C gradients and Cs^+ removal efficiency.

$$\Delta C = \frac{aC_{avg}}{b + C_{avg}} \quad (14)$$

$$N_{100} = 9.77 + 1.75 \times 10^3 v_{in} + 1.81 \times 10^6 C_{in} + 4.24 \times 10^7 v_{in}^2 + 2.66 \times 10^9 C_{in}^2 \quad (15)$$

Combining the fitted N_{100} and δ_C , the processing capacity of a microfluidic adsorption unit (dimensions: 0.5 m × 0.1 m × 0.005 m) is calculated as 40–200 bed volumes per hour, 8–10 times higher than conventional industrial fixed-bed systems (5–20) [49,50]. Combined with the directional recovery characteristics of magnetic powder, the microchannel adsorber can achieve integrated dynamic adsorption-separation operation, and the material utilization rate is significantly higher than that of traditional fixed beds. These results demonstrate the superior performance of the M-SA/PB-Co composite microspheres in microchannel adsorbers for Cs^+ removal from nuclear wastewater.

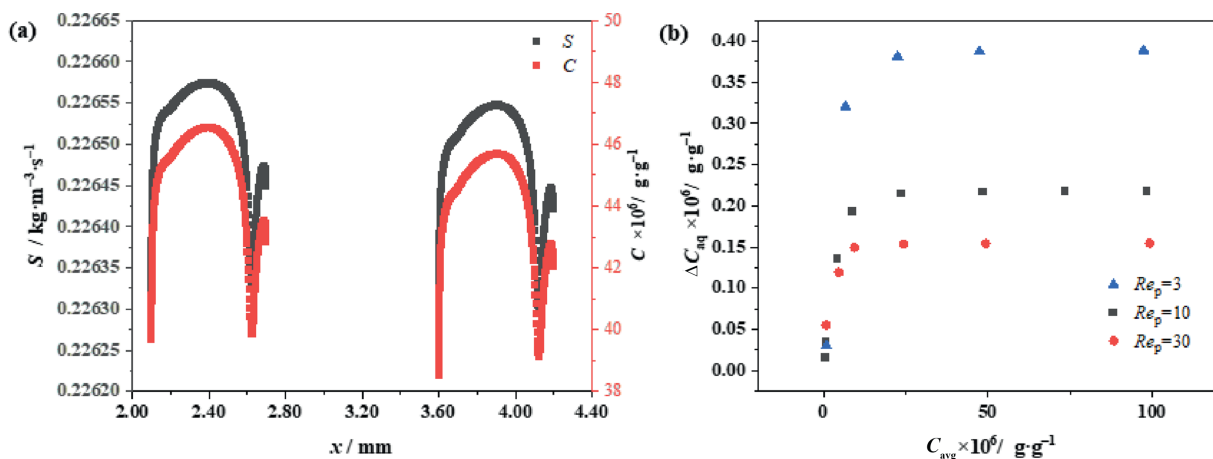


Fig. 11. (a) Adsorption capacity (S) and particle surface Cs^+ concentration (C) profile. (b) ΔC_{aq} versus Re_p and C_{avg} of particle 2.

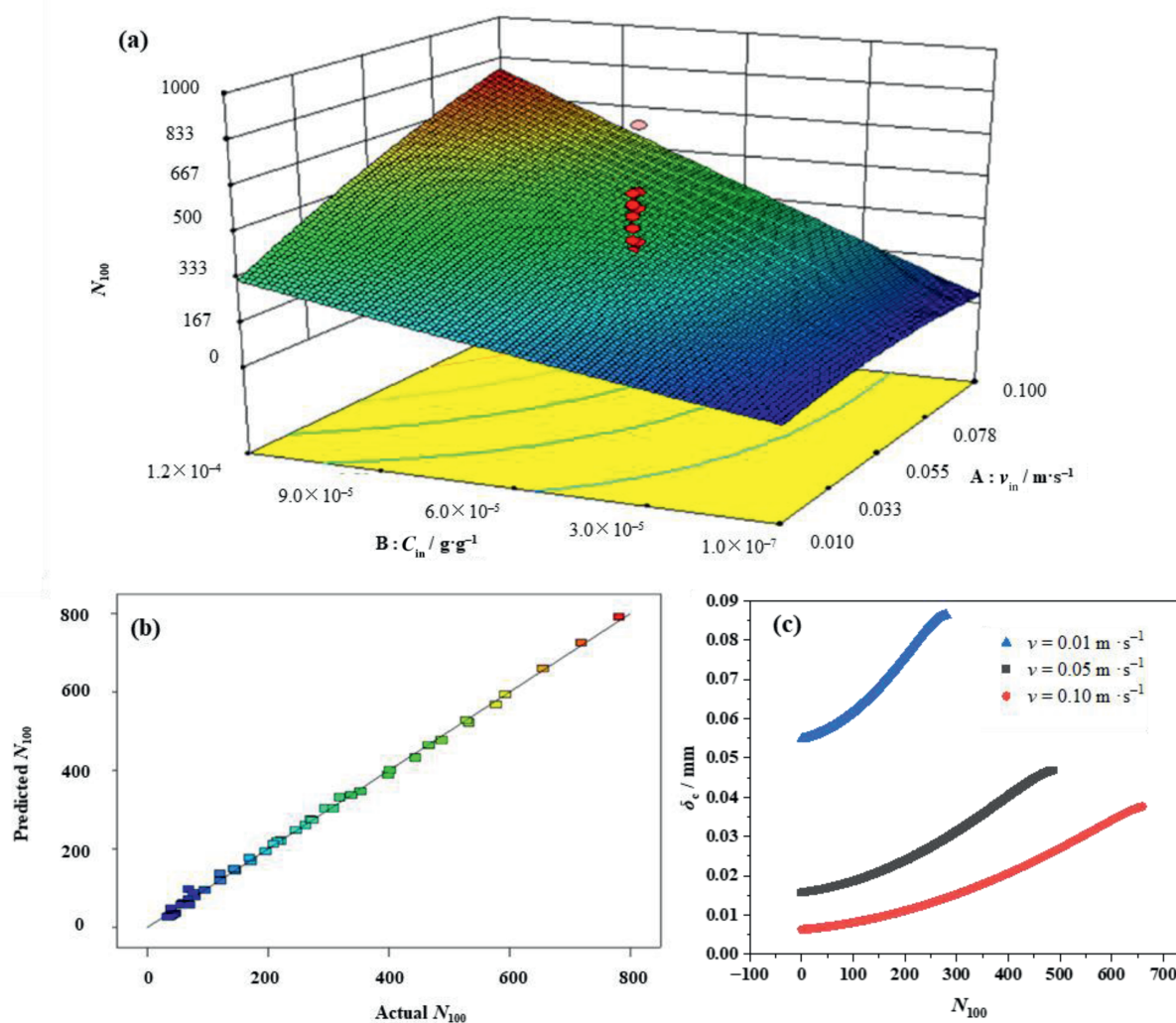


Fig. 12. Response surface (a) and prediction values (b) of N_{100} and the δ_c of particles (c).

5. Conclusions

In this study, the concept of a magnetically controlled microchannel adsorber based on magnetic particles was proposed, and the M-SA/PB-Co composite microsphere adsorbent was successfully prepared for the adsorption treatment of Cs^+ in nuclear wastewater. Adsorption characteristics, CFD simulations, and RSM analyses of the microsphere adsorbents' application in microchannel adsorbers were investigated. The following conclusions were drawn:

- (1) A scheme utilizing magnetically controlled microchannel adsorbers to remove radionuclides from nuclear wastewater was proposed, and the required composite microsphere adsorbents were synthesized. The prepared M-SA/PB-Co microspheres have a uniform size of 300–600 μm and a uniform elemental distribution, demonstrating rapid equilibration (<4 h) governed by PSO kinetics and Langmuir isotherm compliance ($Q_m = 124.84 mg \cdot g^{-1}$).
- (2) A novel protocol was proposed to achieve the higher adsorption performance in a microchannel adsorber with periodically distributed particles. Characteristic velocity and

characteristic concentration of specific regions surrounding the particles were extracted based on boundary layer distribution. The CFD simulation results showed Cs^+ removal capacity ΔC plateaus were at $25 mg \cdot L^{-1}$ ($Re_p = 10$) and ΔC_{max} was reduced as Re_p increased due to vortex-driven residence time shortening.

- (3) The analysis of the concentration boundary layer in the microchannel adsorber flow field indicates a gradually expanding channel form should be considered in the design and optimization of the adsorbers. The processing capacity of the microfluidic adsorption unit is 8–10 times that of industrial fixed beds, showing outstanding advantages in nuclear industrial applications.

CRediT Authorship Contribution Statement

Hongjing Dai: Writing – original draft, Funding acquisition, Data curation, Conceptualization. Ziyi Bo: Writing – review & editing, Supervision, Investigation, Formal analysis. Sifang Wang: Supervision, Resources, Funding acquisition. Shengyang Tao: Supervision, Resources, Investigation. Xuehu Ma: Writing – review & editing, Supervision, Methodology.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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