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Numerical investigations and mathematical models of carbon capture by adsorption-A review

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Abstract

The concentration of CO₂ in the atmosphere, which causes a serious impact on the climate, provided a development space for the carbon capture technology. The adsorption technology for carbon dioxide capture has been extensively studied because of its low energy consumption. The experimental study on carbon dioxide capture adsorption technology, which commonly require enough time-consuming and funding, determines the significance of numerical investigation. In this paper, the progress on numerical investigation and mathematical model of heat transfer and mass transfer in the adsorption chamber is reviewed. The simplified method of the adsorption process is analyzed in the following aspects: the mass transfer process is commonly simplified as the adsorption kinetic model and the adsorption equilibrium model, the pressure loss is expressed by the bed pressure drop, and the heat transfer process is analyzed by the temperature distribution in the adsorption chamber. The construction and application of various researchers' models are finally summarized in this paper.

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Keywords: Carbon capture; Adsorption; Mass transfer; Heat transfer; Numerical simulation

1. Introduction

Fossil fuels are still the mainstream of the energy sector in recent year, considering a dominant proportion of cumulative total amount. The annual CO₂ emissions from fossil fuel combustion is about 30Gt, and the deforestation of tropical rainforests results in CO₂ emissions increasing at an annual rate of 4Gt [1]. It is clear that human activity

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leads to an increase in atmospheric CO₂ concentration, and the increase in CO₂ concentration in the atmosphere will lead to an irreversible climate change. It has been argued that this tendency will continue for more than 1,000 years even after the elimination of CO₂ emissions [2], and it is of practical significance to study CCS technology that can reduce atmospheric CO₂ content.

In the large-scale capture of CO₂ industrial technology, the adsorption method attracts much attention as the equipment is easy to control and with low energy consumption. It's also because the low-grade thermal energy can be applied, and the method suitable for large temperature range, the trial of industrial demonstration device is concerned widely [3]. However, with a development of adsorbent, design and optimization of process, a higher research investment and time-consuming would become a challenge to experiment investigation.

The object of the numerical investigation and mathematical model is to clarify the heat transfer and mass transfer in the adsorption volume during the capture process, and thus the equation solving should rely on the control equations formed by the mass equation, the energy equation and the momentum equation [4]. In 2016, Ben-Mansour presented a comprehensive review on the contents of numerical studies and model construction [5]. The construction of the model includes simplifying the mass transfer process as the combination of adsorption kinetic model and the adsorption equilibrium model, representing momentum loss by pressure drop in the adsorption volume, and analyzing the heat transfer process through assuming the distribution of temperature in the adsorption bed. Wang [6] analyzed the adsorption kinetic model including linear driving force model (LDF), local equilibrium model, pore diffusion model. Chen [7] studied a variety of classical adsorption equilibrium models, including the Langmuir model. However, their review work did not show the inspired relationship analysis among these three equations, and thus without a detailed highlight on research methodology on this field.

This paper summarize the existing simplified models on heat and mass transfer of CO₂ adsorption capture with an overview on various application potential and limitation of models. The summary in this paper can be considered as a research methodology guideline for the numerical simulation of carbon dioxide capture by adsorption method. The full-text is shrunken due to page limitation.

Nomenclature

b	Temperature correction coefficient
B	Competitive adsorption correction coefficient
c	Gas concentration in fluid phase
C	Heat capacity
D_L	Axial diffusion coefficient
D_C	Spherical adsorbent diameter
H	Adsorption heat
d_p	Particle diameter
n	Homogeneous temperature
L	Adsorption bed thickness
q	Gas concentration in solid phase
P	Partial pressure of adsorbed gas
T	Temperature
q	Maximum adsorption concentration
u	Fluid velocity
t	Time
v_m	Adsorption capacity
y	Gas phase fraction
θ	Contact angle
ρ	Density
ε	Porosity
μ	Viscosity coefficient
J	Component J
i	Component i
s	Solid

g	Gas
ps	Adsorbat;
pa	Adsorbent

2. Mass conservation

Adsorption volume applied for CO₂ adsorption capture is full of porous adsorbent material. The gas flows in the adsorption volume across the adsorbent material driven by pressure, and flow around the individual adsorbent material particles. The adsorbed gas diffuses to the particle surface, and reach the surface of the adsorbed particles, passing to the pores inside particle, and finally adsorbed by adsorbent material, which is contrary to the desorption process. In this way, take the common axial flow adsorption volume as example, if only consider the bed flow and mass transfer of the flow in the axial direction, the gas mass conservation equation for the whole volume is:

$$-D_L \frac{\partial^2 c}{\partial z^2} + \frac{\partial^2 (vc)}{\partial z} + \frac{\partial c}{\partial t} + \frac{1-\varepsilon}{\varepsilon} \frac{\partial \bar{q}}{\partial t} = 0 \quad (1)$$

Mass transfer rate of adsorbed gas, which is expressed as $\partial q_i / \partial t$, is the source term to be solved in the equation. Thus, the study on how to achieve mass transfer can be considered as an essence study on the adsorption.

2.1. Adsorption kinetics model

The essence of adsorption separation is the mass transfer process between the solid adsorbent particles and external gas, leading to changes in gas quality. The adsorption kinetic model describes the amount of change in the mass of the gas absorbed over time due to the mass transfer between the solid adsorbent particles and the external gas, which is the mass transfer rate of gas adsorption. In order to simplify the model and simulate the adsorption process, many studies focus on the adsorption kinetic model, and common types of models include: local equilibrium model, linear driving force model and role diffusion model. The summary table would be contained in an extended version of this paper.

2.1.1. Local equilibrium model

Ignoring the mass transfer resistance in the transfer process, assume that the main features of the model are the mass transfer between solid adsorbent particles and external gas instantaneous completes. It is a common assumption in the effect between the adsorbent material and the strongly adsorbable component, which means that the amount of change in the mass of the adsorbed gas component over time is equal to the amount of equilibrium adsorption of the adsorbed gas component.

$$\frac{\partial q_i}{\partial t} = \frac{\partial q_i^*}{\partial t} \quad (2)$$

Due to the neglect of mass transfer resistance, the model is limited in practical application, and only is close to the adsorption separation process in the ideal condition. Shendalman [8] used the local equilibrium model for the first time in the PSA process numerical simulation of carbon capture by adsorption, and the application of this model also occurs in Kumar [9], Chou [10] and so on.

2.1.2. Linear driving force model

This model assumes that the internal diffusion rate of the particles is much larger than that of the external convective mass transfer rate, and that the adsorption rate is proportional to the concentration difference of the adsorbed gas component between the interior and external of the adsorbent material, which is the main characteristic of the model. The LDF model is the most widely used model, and its initial model expression as:

$$\frac{\partial q}{\partial t} = k(q^* - q) \quad (3)$$

The proportionality constant k the determination of the adsorption mass transfer coefficient, is the key to the model and can be determined by experiment or by model selection. Glueckauf [11] first proposed the LDF model for a single grain by assuming that the particles with homogeneous interior holes, under isothermal isobaric conditions; Liaw [12] in the solid diffusion conditions, assuming that the concentration of particles is parabolic, has verified the Glueckauf's LDF model; Sircar [13] also validates the effectiveness of the LDF model in simulating the adsorption kinetics, and the application of the model occurs in the work of Diagne [14], Krishnaurthy [15] and so on.

2.1.3. Pore Diffusion Model

Assuming that the microstructure of the spherical adsorbent particles is all micropore crystals, solve the diffusion process of the adsorbed gas from the outer surface of the adsorbent particles to the interior of the adsorbent particles with the solid diffusion condition:

$$\frac{\partial q}{\partial t} = \frac{1}{r^2} D_c \frac{\partial}{\partial r} \left(r^2 \frac{\partial q}{\partial r} \right) \quad 0 \leq r < r_0 \quad (4)$$

The amount of mass change of the adsorbed gas component over time can be calculated as:

$$\frac{\partial q}{\partial t} = 6q^* \sum_{n=1}^{\infty} \frac{D_c}{r_0^2} \exp \left[-(n\pi)^2 \frac{D_c}{r_0^2} \right] \quad (5)$$

The simulated object can be an arbitrary gas component, and the equation is considered an accurate expression of the adsorption rate, but due to the existence of infinite series, mathematical processing is complex, the calculation is difficult, so there are less practical applications. Weber [16] has conducted a detailed analysis of the application of the pore diffusion model, and Brandani [17] evaluated the pore diffusion capacity of the different adsorbed gas, the application also occurs in the work of Doong [18], Mulgundamath [19] and so on.

2.2. Adsorption equilibrium model

Obviously, in order to describe the mass transfer phenomenon during adsorption separation, the amount of adsorption of gas absorbed, q^* , which is used for evaluation of adsorbents' adsorption performance, is a key parameter to numerical investigation. Adsorption equilibrium model, which employed mathematical models to express the equilibrium adsorption amount, mainly reflects in the effect of adsorption isotherm fitting. To choose the adsorption equilibrium model, which can effectively reflect the adsorption characteristics of adsorbents, is of great significance for the numerical simulation of adsorbed carbon dioxide capture. Several models commonly used in the study include Langmuir model, Langmuir-Freundlich model and Toth model.

2.2.1. Langmuir model

Since the establishment of the Langmuir model in 1918, the most widely used model of single component gas-solid adsorption has been developed rapidly. According to the kinetic principle, assuming that only monolayer adsorption exists, and no interaction between molecules, and the adsorbent surface is uniform, with the same adsorption position, Langmuir [20] described the following isotherms with the Langmuir model:

$$\frac{q^*}{q_m} = \frac{KP}{1+KP} \quad (6)$$

The determination of adsorption equilibrium constant is the key of this model, while due to the effect of change of adsorption heat on the change of concentration of gas absorbed, the fitting error caused by this constant increases with the increase of the concentration of adsorbed gas components, which is the main defect of this model. Ruthven DM [21] and Hwang [22] treat adsorption equilibrium constant as a function of temperature, and Kumar [9], Mulgundamath [19] have the application of this model.

2.2.2. Langmuir-Freundlich model

The Langmuir-Freundlich model was proposed by Freundlich to achieve the isotherm of the adsorption amount correction (LRC) of the Langmuir model for multi-component gas-solid adsorption:

$$q_i = \frac{V_m B_i (P y_i)^{b_i}}{1 + \sum_j^n B_j (P y_j)^{b_j}} \quad i = 1, 2, \dots, n \quad (7)$$

The model can be used to calculate the equilibrium adsorption of the adsorbent material of a component gas in a multicomponent gas more accurately, in a non-iterative manner, and its application occurs in Cen [23], Doong [18], Lee [24], and so on.

2.2.3. Toth model

The Toth model is proposed by Toth [25], which is also based on the Langmuir model, to achieve the isotherm taking into account the influence of heterogeneity coefficient on the surface of adsorbent when gas absorbed contact with adsorbent material's surface:

$$q_{Toth} = \frac{q_m b P}{[1 + (bP)^t]^{1/t}} \quad (8)$$

The heterogeneity coefficient introduced by this model can effectively solve the simulation error caused by the heterogeneous surface of the adsorbent material [26]. The application of the model also occurs in Cavenati [27], Dantas [28].

3. Energy and momentum conservation

3.1. Energy conservation

The endothermic and exothermic phenomena in the adsorption and desorption process leads to the energy changes in the adsorption volume, inducing the heat transfer phenomenon, which leads to the temperature change, and then influence the change of mass transfer rate and the equilibrium adsorption capacity. The whole process has important significance of the simulation of adsorption carbon capture. Complex heat transfer phenomenon makes it difficult to solve the simulation, and it is important to simplify the energy conservation equation by some hypothesis.

① Based on the isothermal system, the whole volume temperature is conserved. The equation can be used directly in solving the equations, and the hypothesis was applied in the work of Raghavan [21], Hwang [22], Takamura [29].

② The hypothesis applied by Kumar [9], Cavenati [27], Dantas [28], based on the assumption that the outer wall of the adsorbent volume is adiabatic, the energy conservation equation in the whole volume is:

$$\frac{\partial}{\partial t} [(\varepsilon \rho_g C_g + \rho_s C_s) \theta] \Big|_x = -C_g \frac{\partial}{\partial x} (Q \theta) \Big|_t + \rho_s \sum q_i \frac{\partial n_i}{\partial t} \Big|_x \quad (9)$$

③ Based on the assumption that the energy of the fluid and the adsorbent particles can be equilibrated instantaneously, It is more close to reality and is more widely used, applied in the work of Cen [23], Doong [18], Lee [24], Mulgundmath [19].

$$\frac{\partial u C C_p T}{\partial z} + \varepsilon \frac{\partial C C_p}{\partial t} + \frac{\rho}{V_m} \frac{\partial [q(C_{pa} T - H)]}{\partial t} + \rho C_{ps} \frac{\partial T}{\partial t} + \rho C_{ps} \frac{\partial T}{\partial t} + \frac{2h}{r} (T - T_0) = 0 \quad (10)$$

3.2. Momentum conservation

The loss of momentum caused by resistance when gas is flowing between the adsorbent particles in the porous media area reflects in the drop of pressure in the whole volume. For example, Delgado [14] takes the Ergun model as

the source term This is also the most usual model that be used to simplify the source of momentum equation in many papers.

$$-\frac{\partial P}{\partial x} = \frac{150L\mu(1-\varepsilon)^2}{\varepsilon^3 d_p^2} u + \frac{1.75L(1-\varepsilon)\rho_g}{\varepsilon^3 d_p} u^2 \quad (11)$$

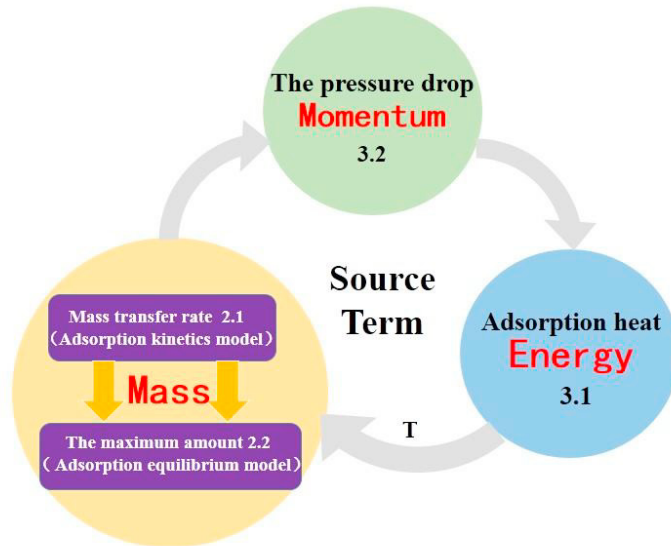


Fig. 1. The source term of control equations

4. Research progress

Shendalman [8] builds the mathematical model of adsorbing and separating CO₂ from He-CO₂ mixed gas with silica gel as adsorbent material, neglecting the adsorption of He on silica and the pressure drop in volume due to momentum loss, with the assumption that the mixture is the ideal compressible fluid and the system is isothermal system, based on which, the model has been built with the selection of local equilibrium model for adsorption kinetic model, and linear equilibrium model for adsorption equilibrium model. The simulation results show an error compared to the experimental results, which is considered caused by the neglect of the adverse effects of mass transfer resistance with the selection of local equilibrium model.

In the mathematical model (Cen, [23]) describing the adsorption of separated CO₂ from the multi-component mixture of the simulation of gasification product gas(H₂ / CO / CH₄ / H₂S / CO₂), with activated carbon as adsorbent material, the drop pressure is also ignored, and it is also assumed that the volume temperature is constant at 40 °C, the wall temperature of the adiabatic adsorption chamber is constant at 20 °C, and the energy of the gas and the adsorbent particles is instantaneously balanced. On the basis of this, select the Langmuir-Freundlich model and the linear driving force model, and the simulation results dovetail the experimental results, which demonstrate the applicability of the Langmuir-Freundlich model for multi-component gas.

Doong [18] built the mathematical model describing the adsorption of separated CO₂ from the multi-component gas mixture (H₂ / CH₄ / CO₂) with activated carbon as adsorbent material by ignoring the drop pressure and assuming the mixture is an ideal fluid. On the basis of this, select the Langmuir-Freundlich model, and compare local equilibrium model with pore diffusion model. The results show the accuracy of the local equilibrium model relative to the pore

diffusion model, prove that the results of pore diffusion model is more suit to the experiment data, it is also proved that the Langmuir-Freundlich model has a higher speed to solve equations than another Langmuir correction model IAS.

Kumar[9] adsorbs separated CO₂ from the multi-component gas mixture (N₂/CH₄/H₂), ignoring the volume pressure drop, assuming that the outer wall of the adsorption chamber is adiabatically, and the energy of the gas and adsorbent particles can equalize instantaneously, with zeolite 5A and BPL carbon as adsorbent materials. On the basis of this and selection of the local equilibrium model and the Langmuir model, The simulation results show that the adiabatic assumption has a greater negative effect on the accuracy of the model than the concentration and pressure.

The model of Diagne[30]adsorbs separated CO₂ from the multi-component gas mixture (CO₂/atmosphere), ignoring the volume pressure drop, assuming the mixture gas is an ideal fluid with zeolite 13X、 zeolite 5A、 zeolite 4A as adsorbent materials. On the basis of this and the selection of the linear driving force model and the Langmuir model, the simulation results are in good agreement with the experimental results to demonstrate the accuracy of the simulation model.

The model of Takamura[29]adsorbs separated CO₂ from the multi-component mixture of simulated boiler flue gas (CO₂/N₂/O₂), ignoring the volume pressure drop, assuming that the mixture gas is an ideal fluid and the system is isothermal, with zeolite NaX and zeolite NaA as adsorbent materials. On the basis of this and the selection of the linear driving force model and the Langmuir model, the simulation results are also in good agreement with the experimental results to demonstrate the rationality of the model.

The model of Dantas[28]adsorbs separated CO₂ from the multi-component gas mixture (CO₂/N₂), ignoring the volume pressure drop, assuming that the mixture gas is an ideal fluid and respectively the outer wall of the adsorption chamber adiabatic / non-adiabatic two cases, with zeolite NaX and zeolite NaA as adsorbent materials. On the basis of this and the selection of the linear driving force model and the Toth model, using the Ergun model to express the pressure drop. The simulation results are in agreement with the experimental results, which proves that the adsorption of CO₂ / N₂ is less competitive, and the Toth model which used for single component adsorption can also be fitted.

5. Summary remarks

In this paper, the basic model construction and research progress of the numerical simulation of carbon capture by adsorption are reviewed. The following conclusions can be drawn:

1. The numerical solution of carbon capture by adsorption depends on the control equations composed of mass, momentum and energy equations. Appropriate assumptions are important for simplifying the equations and reducing the difficulty of solving. This paper lists the assumptions of different researchers, with reference significance.
2. The core of the study is the selection of the adsorption kinetic model and the adsorption equilibrium model: selecting the appropriate mass transfer rate equation to represent the mass transfer process during the carbon dioxide adsorption process, and the simplified method of determining the maximum adsorption amount by adsorbing the equilibrium isotherm to design the quality control Equation source term. This paper lists the selection of different researcher models and analyzes the applicability of different models.
3. In the study of the momentum equation, the bed pressure drop caused by the momentum loss is often neglected and the simulation results are not affected. In the energy equation study, the temperature distribution of the adsorbent bed has an influence on both the heat and mass transfer phenomena.

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Biography

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