MODELLING OF ADSORPTION OF DYES FROM AQUEOUS SOLUTION BY ACTIVATED CARBON

By

WONG TECK NGIN

Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in Fulfilment of the Requirements for the Degree of Master of Science

March 2004

DEDICATION

To my parents and grandmother

Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of the requirement for the degree of Master of Science

MODELLING OF ADSORPTION OF DYES FROM AQUEOUS SOLUTION BY ACTIVATED CARBON

By

WONG TECK NGIN

March 2004

Chairman: Budiatman Satiawihardja, Ph.D.

Faculty: Engineering

Adsorption process has been gaining popularity as an effective alternative for separation processes. Two fundamental properties that influence the adsorption rate are the adsorption equilibrium and mass transfer limitation. The adsorption isotherm is obtained from batch studies. The mass transfer coefficients obtained from batch studies need to be extrapolated by matching the model with the experimental data. The mass transfer parameters are important in designing a fixed-bed absorber, commonly used in the industry. For dye adsorption on activated carbon, concentration dependent surface diffusivity is the most important mass transfer parameter and must be included in the study. The pore diffusivity should also be included to improve the accuracy of the simulation. Therefore, the film-pore-concentration dependent surface diffusion (*FPCDSD*) model is identified as the best model to describe the adsorption rate of dye onto activated carbon.

In this study, a mathematical model for adsorption rate is developed based on the *FPCDSD* model. The governing partial differential equations (*PDE*s) are transformed to ordinary differential equations (*ODE*s) using orthogonal collocation (*OC*) method. These

sets of *ODE*s are then integrated using the numerical algorithm *DIVPAG* (I*MSL* library subroutine), which is based on variable order, variable step method implementing backward differential formula (Gear's Method) and is suitable for stiff system of first order non-linear *ODE*s. Programs written in FORTRAN 90 are used to extrapolate the mass transfer parameters by matching the simulation data with the experimental data of batch studies. The *FPCDSD* model is sufficiently general and thus can be reduced to describe other simplified models for liquid adsorption easily, *e.g.* the film-concentration dependent surface diffusion (*FCDSD*) model and the film-pore diffusion (*FPD*) model.

Three set of experimental data from Choy *et al.* (2001) based on different masses were selected to test the applicability of the *FPCDSD* model in simulating batch adsorption. Simulation results show that, for acid dye/activated carbon system a single set of mass transfer parameters is able to match the simulation and experimental data using the *FPCDSD* model and the *FCDSD* model. However, ignoring the pore diffusion, there resulting a 30% differences in the surface diffusion. For Methylene Blue/*PKS* (different larger initial concentrations) systems, only the *FPCDSD* model could use a single set of mass transfer parameters.

The *FPCDSD* model is then further extended to model the fixed-bed adsorber. A computer program written in FORTRAN 90 is developed. The *PDE*s for the axial and radial directions are discretised into *ODE*s using *OC* method. Column results showed that the retention time increases with increasing bed length and superficial velocity. Increasing the bed porosity, the residence time will decrease. Using the equilibrium isotherm and mass transfer parameters obtained from batch studies and with a suitable correlation for film mass transfer coefficient, the fixed-bed model can be used to predict the breakthrough curve of column adsorption.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Master Sains

PEMODELAN PENJERAPAN PEWARNA DARIPADA LARUTAN AKUEUS OLEH KARBON TERAKTIF

Oleh

WONG TECK NGIN

Mac 2004

Pengerusi: **Budiatman Satiawihardja, Ph.D.**

Fakulti: **Kejuruteraan**

Proses penjerapan telah mendapat populariti sebagai satu pilihan proses pemisahan yang berkesan. Dua sifat dasar yang menentukan kadar penjerapan ialah keseimbangan penjerapan dan had pemindahan jisim. Isoterma jerapan didapati daripada pengkajian berkelompok. Manakala koefisien pemindahan jisim bagi pengkajian berkelompok diekstrak dengan pemadanan model dengan data eksperimen. Parameter bagi pemindahan jisim penting bagi perekaan turus penjerapan tertetap, terutamanya yang digunakan dalam bidang perindustrian. Bagi penjerapan pewarna pada karbon teraktif, penyandaran kepekatan jerapan permukaan merupakan parameter pemindahan jisim yang paling penting dan mesti dimasukkan dalam pengkajian. Jerapan liang juga mesti dimasukkan untuk meninggikan ketepatan simulasi. Oleh itu, model penjerapan filem-liang-penyandaran kepekatan permukaan (*FPCDSD*) dikenalpastikan sebagai model yang terbaik untuk menghuraikan kadar penjerapan pewarna pada karbon teraktif.

Dalam kerja ini, satu model matematik untuk kadar penjerapan telah diterbit berdasarkan model *FPCDSD*. Persamaan pembezaan separa (*PDE*s) telah ditransfomasikan kepada

persamaan pembezaan biasa (*ODE*s) dengan menggunakan cara kolokasi ortogon (*OC*). Kemudian *ODE*s ini dikamirkan dengan algoritma berangka *DIVPAG* (*IMSL* library subroutine), yang berdasarkan peringkat pemboleh ubah, cara berperingkat yang menampilkan rumus pembezaan terbalik (Gear's Method) dan sesuai untuk *ODE*s tak linear berperingkat pertama yang kecerunannya mendadak. Program yang dituliskan dengan FORTRAN 90 digunakan untuk mendapat parameter pemindahan jisim dengan memadankan data simulasi dengan data eksperimen berkolompok. Model *FPCDSD* adalah umum untuk penjerapan bendalir dan boleh diturunkan untuk menghuraikan model lain yang diringkaskan seperti, model penjerapan filem-penyandaran kepekatan permukaan (*FCDSD*) dan model penjerapan filem-liang (*FPD*).

Tiga set data eksperimen yang terpilih daripada Choy *et al.* (2001) berdasarkan jisim karbon teraktif yang berlainan digunakan untuk menguji keberkesanan model *FPCDSD* untuk menyimulasi penjerapan berkelompok. Keputusan simulasi menunjukkan satu set parameter pemindahan jisim memadankan data simulasi dengan data eksperimen dengan menggunakan model *FPCDSD* dan model *FCDSD*. Bagi sistem *Methylene Blue/PKS* (permulaan kepekatan yang berlainan dengan julat yang besar), hanya model *FPCDSD* dapat menggunakan satu set parameter pemindahan jisim. Namun, pengabaian penjerapan keliangan akan mengakibatkan perbezaan sebanyak 30% dalam penjerapan permukaan.

Seterusnya, model *FPCDSD* dikembangkan untuk mengkaji turus jerapan tertetap. Satu program komputer yang menggunakan FORTRAN 90 telah diterbitkan. *PDE*s dalam arah paksi dan jejari didiskretasikan kepada *ODE*s dengan cara *OC*. Keputusan turus menunjukkan masa mastautin meningkat dengam peningkatan ketinggian turus dan kelajuan superficial. Peningkatan keliangan turus akan memendekkan masa mastautin. Dengan keseimbangan isoterma dan parameter pemindahan jisim daripada pengkajian berkelompok dan korelasi yang sesuai bagi koefisien pemindahan jisim berfilem, peramalan model turus penjerapan tertetap boleh digunakan untuk meramal lenkungan pecah-terus bagi turus penjerapan.

ACKNOWLEDGEMENT

I wish to express my deepest gratitude to my main supervisor, Dr. Thomas Choong Shean Yaw and co-supervisor Dr. Chuah Teong Guan for their inspiring guidance, comments, encouragements and patient supervison of my work.

I am also very grateful to Dr. Quek Siew Young for her help, suggestions and encouragement which have contributed to the success of this work.

I am indebted to Dr. Thomas Choong Shean Yaw and Dr. Ngoh Gek Cheng for introducing me to Universiti Putra Malaysia, and to the Ministry of Science and Technology for the financial support.

Finally, I would like to thank my parents and grandmother for their spiritual support.

I certify that an Examination Committee met on 30th March 2004 to conduct the final examination of Wong Teck Ngin on his Master of Science thesis entitled "Modeling of Adsorption of Dyes From Aqueous Solution by Activated Carbon" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the candidate be awarded the relevant degree. Members of the Examination Committee are as follows:

Budiatman Satiawihardja, Ph.D.
Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Medyan Riza, Ph.D.
Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Tey Beng Ti, Ph.D.
Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Abdul Wahab Mohammad, Ph.D.
Associate Professor
Faculty Engineering
Universiti Kebangsaan Malaysia
(Independent Examiner)

____

GULAM RUSUL RAHMAT ALI, Ph.D.
Professor/Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

This thesis submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Master of Science. The members of the Supervisory Committee are as follows:

Thomas Choong Shean Yaw, Ph.D.
Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Azni bin Haji Idris, Ph.D.
Professor
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Chuah Teong Guan, Ph.D.
Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Quek Siew Young, Ph.D.
Lecturer
Faculty of Food Science and Biotechnology
Universiti Putra Malaysia
(Member)

____

AINI IDERIS, Ph.D.
Professor/Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

DECLARATION

I hereby declare that the thesis is based on my original work except for quotations and citations which have been duly acknowledged. I also declare that it has not been previously or concurrently submitted for any other degree at UPM or other institutions.

____

WONG TECK NGIN

Date:

TABLE OF CONTENTS