UNIVERSITI PUTRA MALAYSIA

ADSORPTION OF β-CAROTENE ONTO MESOPOROUS CARBON COATED MONOLITH

MUHAMMAD

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ADSORPTION OF β-CAROTENE ONTO MESOPOROUS CARBON COATED MONOLITH

By

MUHAMMAD

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

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Abstract of thesis presented to the Senate of Universiti Putra Malaysia in fulfillment of the requirement for the degree of Doctor of Philosophy

ADSORPTION OF β-CAROTENE ONTO MESOPOROUS CARBON COATED MONOLITH

By

MUHAMMAD

June 2011

Chairman : Associate Professor Thomas Choong Shean Yaw, PhD

Faculty : Engineering

The mesoporous carbon coated monoliths (MCCM) were developed by dipcoating method using furfuryl alcohol (FA) as a carbon precursor and poly(ethylene glycol) (PEG) as a poreformer. The effect of molecular weight of PEG, carbonization temperature and PEG composition on the synthesis of MCCM were studied. The maximum specific surface area and mesopore area obtained were 61.13 m^2/g and 49.39 m^2/g. The pore size distribution of the carbon coated monolith exhibited two main peaks. One peak was located at 2.0 nm and the other at 3.6 nm. The pore size distribution curve indicated that the porous carbon was bimodally distributed.

The MCCM was utilized as an adsorbent for adsorption of β-carotene from isopropyl alcohol (IPA) and *n*-hexane miscellas. The effect of temperature on the adsorption was

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investigated by batch adsorption experiments. The adsorption quantity increased with increasing temperature. The maximum adsorption capacity of β-carotene obtained was 62.118 mg/g for IPA at 50 ℃. The experimental results were fitted using the Langmuir and Freundlich isotherms. The Langmuir described the adsorption process better. The negative values of Gibbs free energy change suggested that the adsorption was a spontaneous process. The positive values of heat of enthalpy and entropy change confirmed the endothermic nature of the adsorption.

The adsorption kinetics of β-carotene onto mesoporous carbon coated monolith in isopropyl alcohol (IPA) and *n*-hexane solution was investigated, as functions of temperature and β-carotene initial concentration. Adsorption capacity increased as initial β-carotene concentration and temperature increased. In addition, the solvents also play an important role in the adsorption of β-carotene; adsorption kinetic of β-carotene by using IPA is higher than *n*-hexane. Two kinetic models, namely the pseudo-first-order and pseudo-second-order, were used to predict the adsorption kinetics. The rate parameters of the intraparticle diffusion model for adsorption were also evaluated to identify the adsorption mechanisms. The results clearly showed that the adsorption of carotene onto MCCM followed the pseudo-first-order model for IPA, and pseudo-second-order model for *n*-hexane solvent. The energy activation parameters were 11.45 and 9.41 kJ/mol for IPA and *n*-hexane, respectively.

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Sorption kinetics of β-carotene in IPA solution was analyzed at different temperatures and initial concentrations by using the linear driving force (LDF) model. The software MATLAB® was used to solve the LDF model simultaneously with the adsorption equilibrium isotherm at liquid/solid interface. The linear driving force mass transfer coefficient (k_{LDF}) obtained was increased with increasing temperature. However, the LDF model did not describe experimental results satisfactorily at high initial concentrations.

The equilibrium and kinetics of desorption process of β-carotene from MCCM were investigated in a batch system. The MCCM was first saturated with β-carotene from IPA solution. The β-carotene was then desorbed by using *n*-hexane solution. The data of desorption were evaluated by two models *i.e.* linear isotherm and Freundlich isotherm. The desorption was satisfactorily fitted with the Freundlich model. The desorption kinetic was analyzed using a first-order two-compartment three-parameter model. The activation energy obtained was 7.88 and 44.47 kJ/mol for rapid and slow desorption, respectively.

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Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia sebagai memenuhi keperluan untuk ijazah Doktor Falsafah

PENJERAPAN β-KAROTEN KE ATAS MONOLIT DISALUT MESOLIANG KARBON

Oleh

MUHAMMAD

Jun 2011

Pengerusi : Profesor Madya Thomas Choong Shean Yaw, PhD

Fakulti : Kejuruteraan

Mesoliang karbon melapisi monolit (MCCM) telah dihasilkan dengan kaedah pencelupan ke dalam alkohol furfuril (FA) yang bertindak sebagai pelopor karbon dan poli (etilena glikol) (PEG) sebagai pembentuk liang. Pengaruh berat molekul PEG, suhu karbonisasi dan kandungan PEG pada sintesis MCCM telah dikaji. Luas permukaan khusus maksimum dan kawasan mesoliang yang didapati ialah 61.13 m^2/g dan 49.39 m^2/g. Taburan saiz liang dari karbon dilapisi monolit ditunjukkan dengan dua puncak utama. Satu puncak terletak pada 2.0 nm dan yang selainnya ialah 3.6 nm. Lengkungan taburan saiz liang menunjukkan bahawa karbon berliang tersebut ditaburkan secara dwimod.

MCCM ini telah digunakan sebagai penjerap untuk penjerapan β-karoten dari isopropil alkohol (IPA) dan miscellas *n*-heksana. Pengaruh suhu terhadap penjerapan telah diselidiki dengan ujikaji penjerapan secara kelompok. Kuantiti

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jerapan meningkat dengan meningkatnya suhu. Keupayaan maksimum penjerapan β-karoten diperoleh adalah 62,118 mg/g untuk IPA pada suhu 50 °C. Keputusan eksperimen ini dipadankan dengan menggunakan isoterma Langmuir dan Freundlich. Isoterma Langmuir menggambarkan proses penjerapan lebih baik. Nilai negatif dari perubahan tenaga bebas Gibbs menyatakan bahawa penjerapan merupakan suatu proses yang spontan. Nilai-nilai positif haba dari perubahan entalpi dan entropi mengesahkan sifat endotermik penjerapan.

Kinetik penjerapan β-karoten ke atas monolith disalut mesoliang karbon di dalam larutan alkohol isopropil (IPA) dan *n*-heksana telah diselidiki, sebagai fungsi ke atas suhu dan kepekatan awal β-karoten. Keupayaan jerapan akan meningkat setelah kepekatan β-karoten awal dan suhu meningkat. Selain itu, pelarut juga memainkan peranan yang penting dalam jerapan β-karoten; kinetik jerapan dari β-karoten dengan menggunakan IPA lebih tinggi daripada *n*-heksana. Dua model kinetik, iaitu pseudo tertib pertama dan pseudo tertib kedua, telah digunakan untuk menganggarkan kinetik jerapan. Kadar parameter model penyerapan intrapartikel untuk jerapan juga telah dinilai untuk mengenalpasti mekanisma jerapan. Keputusan jelas menunjukkan bahawa jerapan karoten ke atas MCCM mengikuti model pseudo tertib pertama untuk IPA, dan pseudo tertib kedua untuk pelarut *n*-heksana. Parameter tenaga pengaktifan adalah masing-masing 11.45 dan 9.41 kJ / mol untuk IPA dan *n*-heksana.

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Kinetik penjerapan β-karoten dalam larutan IPA dianalisis pada suhu berbeza dan kepekatan awal dengan menggunakan model daya pacu linear (LDF). Peranti perisian MATLAB® telah digunakan untuk menyelesaikan model LDF serentak dengan ekuilibrium isoterma jerapan antara muka cecair/pepejal. Nilai pekali daya pacu linear pemindahan jisim (k_{LDF}) yang diperolehi meningkat dengan peningkatan suhu. Namun, model LDF tidak menjelaskan hasil eksperimen yang memuaskan pada kepekatan awal yang tinggi.

Keseimbangan dan kinetik proses penyahjerapan β-karoten dari MCCM telah diselidiki dalam sistem kelompok. Pada mulanya, MCCM telah ditepukan dengan β-karoten daripada larutan IPA. β-karoten ini kemudian dinyahjerapkan dengan menggunakan larutan *n*-heksana. Data nyahjerapan ini dinilai oleh dua model iaitu isoterma Linear dan isoterma Freundlich. Nyahjerapan itu dikenalpasti sepadan dengan model Freundlich. Kinetik penyahjerapan telah dianalisis menggunakan model tertib-pertama dua-bahagian tiga-parameter. Tenaga pengaktifan yang diperolehi adalah 7.88 dan 44.47 kJ / mol, masing-masing untuk nyahjerapan cepat dan perlahan.

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I certify that a Thesis Examination Committee has met on 24 June 2011 to conduct the final examination of Muhammad on his thesis entitled "Adsorption of β-Carotene onto Mesoporous Carbon Coated Monolith" in accordance with the Universities and University College Act 1971 and the Constitution of the Universiti Putra Malaysia [P.U.(A) .106] 15 March 1998. The committee recommends that the student be awarded the Doctor of Philosophy.

Members of the Thesis Examination Committee were as follows:

Fakhru'L-Razi B Ahmadun, PhD
Professor
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Azni bin Hj Idris, PhD
Professor
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Mohamad Amran bin Mohd Salleh, PhD
Senior Lecturer
Faculty of Engineering
Universiti Putra Malaysia
(Internal Examiner)

Shih-Ming (Steven) Lai, PhD
Professor
National Yunlin University of Science and Technology
Taiwan
(External Examiner)

_____

NORITAH OMAR, PhD
Associate Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 23 Augustus 2011

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This thesis was submitted to the Senate of Universiti Putra Malaysia and has been accepted as fulfilment of the requirement for the degree of Doctor of Philosophy. The members of the Supervisory Committee were as follows:

Thomas Choong Shean Yaw, PhD
Associate Professor
Faculty of Engineering
Universiti Putra Malaysia
(Chairman)

Luqman Chuah Abdullah, PhD
Professor
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Robiah Yunus, PhD
Professor
Faculty of Engineering
Universiti Putra Malaysia
(Member)

Taufiq Yap Yun Hin, PhD
Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

_____
HASANAH MOHD GHAZALI, PhD
Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:

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DECLARATION

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

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Date: 24 June 2011

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