UNIVERSITI PUTRA MALAYSIA

PREPARATION AND CHARACTERIZATION OF MONTMORILLONITE MEMBRANE FOR REMOVAL OF LEAD AND CHROMIUM IONS FROM AQUEOUS SOLUTIONS

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By

NURUL AIN BINTI A. TALIB

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

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

PREPARATION AND CHARACTERIZATION OF MONTMORILLONITE MEMBRANE FOR REMOVAL OF LEAD AND CHROMIUM IONS FROM AQUEOUS SOLUTIONS

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Chairman : Associate Professor Mansor Bin Ahmad, PhD

Faculty : Science

Fabrication of membrane from montmorillonite was carried out for removal of metal ions from aqueous solutions. The formation of membrane was optimized based on materials compositions, sintering time and sintering temperature. Montmorillonite KSF (MMT) was used as replacement for the usually expensive materials in membrane fabrication to reduce the cost of production. Each membrane was prepared by suspension of powder materials method. The paste casting was done with different compositions of MMT along with other relatively inexpensive materials which were sand quartz, calcium carbonate, sodium carbonate and boric acid. All membranes were characterized by Fourier transform infrared (FT-IR) spectroscopy, thermogravimetric analysis (TGA), differential thermal analysis (DTA), scanning electron microscopy (SEM), X-ray

diffraction (XRD), nitrogen adsorption-desorption (BET analyzer) and hardness test. Metal ions concentration in solutions was determined by inductively coupled plasma optical emission spectrometry (ICP-OES).

Compared to usually high sintering temperature needed in regular preparation of ceramic membrane, this membrane was fabricated at temperature below 1000 °C, which was considered low for inorganic membrane preparation. TGA and DTA results showed that as the sintering time and temperature increased, the thermal stability of membrane also increased. No more degradation of samples was identified after 5 h sintering. By incorporating higher percentages of MMT, the stability of the membranes increased. For samples containing 35% MMT and above, 850 °C sintering temperature was sufficient to produce membranes stable for application up to 1000 °C.

The porosity of membrane also increased with sintering temperature. However, as the MMT composition in membrane increased, the porosity decreased due to consolidation of materials at high temperature. Sample 35% MMT sintered at 900 °C was identified as suitable microfiltration membrane based on its maximum pore size 2013 Å (201.3 nm), while main pore distribution was centered at 34.3 Å (3.4 nm) as determined by nitrogen adsorption-desorption analysis.

Lead ions were successfully removed from aqueous solution up to 98% by hybridization of adsorption-microfiltration technique at pH 6.1 with 0.8 wt% (w/v) ferric oxide, Fe_2O_3, as the adsorbent. 92% of chromium ion was also successfully removed at pH 6.0 using the same technique. Other than above metal ions removal, there are also possibilities for separation between metal ion such as Pb^{2+} ion and Cu^{2+} ion, and Cr^{2+} ion and Cu^{2+} ion.

The relatively low cost MMT membrane with sufficient membrane properties is a good material for metal ions removal from aqueous solutions. The hybridization of adsorption-microfiltration technique has high potential application for toxic metal ions removal from wastewater system containing metal waste such as electroplating industrial waste.

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

PENYEDIAAN DAN PENCIRIAN MEMBRAN MONTMORILONIT UNTUK PENYINGKIRAN ION PLUMBUM DAN ION KROMIUM DARIPADA LARUTAN AKUEUS

Oleh

NURUL AIN BINTI A. TALIB

April 2013

Pengerusi : Profesor Madya Mansor Bin Ahmad, PhD

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Fabrikasi membran daripada montmorillonit telah dilakukan untuk digunakan dalam penyingkiran ion logam daripada larutan akueus. Pembentukkan membran telah dioptimumkan berdasarkan komposisi bahan, tempoh sinter dan suhu sinter. Montmorilonit KSF (MMT) telah dipilih sebagai pengganti kepada bahan yang biasanya mahal dalam pembikinan membran untuk mengurangkan kos pengeluaran. Setiap membran dihasilkan menggunakan kaedah suspensi bahan serbuk. Pembentukan pes dilakukan dengan komposisi MMT yang berbeza bersama-sama bahan kos rendah yang lain iaitu pasir quartz, kalsium karbonat, natrium karbonat dan asid borik. Pencirian semua membran dilakukan menggunakan spektroskopi inframerah transformasi Fourier (FT-IR), analisis termogravimetri (TGA), analisis pembezaan terma (DTA), mikroskopi

pengimbasan elektron (SEM), pembelauan sinar-X (XRD), analisis luas permukaan dan keliangan (analisis BET) dan ujian kekerasan. Kepekatan ion logam dalam larutan akueus ditentukan menggunakan spektrometri pemancaran optik plasma berganding aruh (ICP-OES).

Berbanding dengan kebiasaan suhu sinter yang tinggi diperlukan untuk penyediaan membran seramik, membran ini dihasilkan pada suhu kurang daripada 1000 °C, yang mana dianggap rendah untuk penyediaan membran bukan organik. Pencirian melalui TGA and DTA menunjukkan bahawa apabila tempoh dan suhu sinter ditingkatkan, kestabilan terma membran meningkat. Tiada lagi proses degradasi dikenalpasti selepas sampel disinter selama 5 jam. Peratusan MMT yang tinggi turut meningkatkan kestabilan membran. Bagi membran yang mengandungi 35% MMT dan ke atas, suhu sinter 850 °C sudah mencukupi untuk menghasilkan membran yang stabil untuk aplikasi sehingga 1000 °C.

Bilangan keliangan membran turut bertambah dengan pertambahan suhu sinter. Walau bagaimana pun, apabila komposisi MMT dalam membran bertambah, bilangan keliangannya berkurangan disebabkan oleh penggabungan bahan pada suhu tinggi. Sampel 35% MMT yang disinter pada 900 °C telah dikenalpasti sesuai sebagai membran mikropenurasan berdasarkan saiz liang maksimumnya iaitu 2013 Å (201.3 nm), manakala taburan liang utamanya berpusat pada 34.3 Å (3.4 nm) seperti yang ditentukan dengan menggunakan analisis penjerapan-penyahjerapan nitrogen.

Ion-ion plumbum telah berjaya disingkirkan dari larutan akues sehingga 98% menggunakan teknik hibrid penjerapan-mikropenurasan pada pH 6.1 dengan 0.8 wt% (w/v) ferik oksida, Fe_2O_3, sebagai penjerap. Sebanyak 92% ion kromium turut berjaya disingkirkan pada pH 6.0 menggunakan teknik yang sama. Selain daripada penyingkiran ion logam tersebut di atas, terdapat juga kebarangkalian untuk pemisahan antara ion, seperti pemisahan antara ion Pb^{2+} dengan ion Cu^{2+}, dan antara ion Cr^{2+} dengan ion Cu^{2+}.

Membran MMT berkos rendah yang cukup dengan sifat membran ini merupakan bahan yang sesuai untuk penyingkiran ion logam daripada larutan akueus. Teknik hibrid ini mempunyai potensi aplikasi yang tinggi untuk penyingkiran logam toksik daripada sistem air buangan yang mengandungi buangan logam seperti bahan buangan industri penyaduran.

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I certify that an Examination Committee has meet on date of viva voce to conduct the final examination of Nurul Ain Binti A. Talib on her Master of Science thesis entitled "Preparation and Characterization of Montmorillonite Membrane for Removal of Lead and Chromium Ions from Aqueous Solutions" in accordance with Universiti Pertanian Malaysia (Higher Degree) Act 1980 and Universiti Pertanian Malaysia (Higher Degree) Regulations 1981. The Committee recommends that the student be awarded the relevant degree.

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

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DECLARATION

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

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NURUL AIN BINTI A. TALIB

Date: 23 April 2013

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