UNIVERSITI PUTRA MALAYSIA

APPLICATION OF LAYERED DOUBLE HYDROXIDES AS HOST IN CONTROLLED RELEASE FORMULATION OF LATEX STIMULANT AND METAL CATALYST IN FORMATION OF CARBON NANOTUBE

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ADILA MOHAMAD JAAFAR

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By

ADILA MOHAMAD JAAFAR

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Chairman: Professor Mohd Zobir Hussein, PhD

Faculty : Institute of Advanced Technology

Layered double hydroxides (LDH) have attracted a lot of attention in research area due to its unique properties. LDHs has proven to be useful in various fields of applications and *d*ue to that, LDHs have been chosen to be used in this study as precursor in two applications which are; as host in a latex stimulant controlled release formulation and metal catalyst in the formation of carbon nanotubes. Manipulation of LDHs could lead to the formation of various useful materials and this is contributed by the interesting property of LDHs which are ion exchange and high metal dispersion. From the study, the syntheses of latex stimulant nanocomposites, controlled release of latex stimulant in various aqueous solutions and formation of carbon nanotubes were proven to be successful. It is hoped that in the future the knowledge of LDHs could lead to application of many beneficial materials in various fields.

In this study, Zinc-Aluminium-nitrate-LDH (ZAL) synthesised at different Zn/Al molar ratios, R=2-4 and pH = 7.50-10.00 were successfully synthesised via co-precipitation method under N_2 atmosphere. Latex stimulant agent of 2,4-D and 2-CEPA have been successfully inserted into the lamella of layered double hydroxides via co-precipitation. Various parameters namely Zn to Al molar ratios of the mother liquor which are 2, 3 and 4; concentrations of anion, 2,4-D and 2-CEPA which are 0.08 M- 0.32 M; and pH which are 7.50 - 10.00 have been tested during the synthesis of Zn-Al-2,4-D (ZANDI) and Zn-Al-2-CEPA (ZANE) nanocomposites.

X-ray diffraction patterns showed expansion of the precursor's basal spacing in order to accommodate the 2,4-D and 2-CEPA anions which are larger in size than nitrate. FTIR spectrum of both prepared ZANDI and ZANE nanocomposites resembled a mixture of each parent material, ZALs and guest anions of 2,4-D and 2-CEPA, indicating the presence of both in the resulting materials. Taken together the X-ray diffraction and FTIR analyses, successful formation of ZANDI and ZANE were further confirmed. Further physico-chemical characterisations of the resulting materials including thermal analysis, elemental analysis, surface area and porosity and surface morphology, were also carried out.

The controlled release study of 2,4-D and 2-CEPA were done in aqueous media of sodium chloride, sodium carbonate and sodium phosphate at 0.05 M. Release of 2,4-D and 2-CEPA were also done in water as control. It was found that 2,4-D and 2-CEPA were rapidly released in sodium carbonate and sodium phosphate, respectively.

However, the release percentage of 2,4-D was found to be the highest in carbonate at 99 % and for 2-CEPA in sodium phosphate at 98 % . The kinetic model of pseudo-second order fits well with all of the release profile with $r^2 > 0.9$.

CNT from the calcined CoNiAl-LDH was produced with high yield when compared to calcined FeCoNiAl-LDH and FeNiAl-LDH. However, CNTs from calcined FeCoNiAl-LDH was found in abundance after the purification took place. CNTs synthesised by using the metal foils afforded much smaller diameter and better surface properties which could be due to the well dispersed highly pure metal.

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

APLIKASI HIDROKSIDA BERLAPIS GANDA SEBAGAI PERUMAH DALAM FORMULASI PERLEPASAN TERKAWAL PENGGALAK SUSU GETAH DAN MANGKIN LOGAM DALAM PEMBENTUKAN KARBON NANO TIUB

Oleh

ADILA MOHAMAD JAAFAR

Disember 2010

Pengerusi: **Profesor Mohd Zobir Hussein, PhD**

Fakulti : **Institut Teknologi Maju**

Hidroksida berlapis ganda (HBG) telah menarik perhatian banyak pihak dalam bidang penyelidikan disebabkan oleh cirinya yang unik. HBG telah disahkan berguna dalam pelbagai aplikasi dan kerana itu HBG telah dipilih untuk digunakan di dalam kajian ini sebagai bahan pemula di dalam dua aplikasi iaitu sebagai perumah kepada penggalak susu getah untuk formulasi perlepasan terkawal dan mangkin logam untuk pembentukan karbon nano tiub (KNT). Manipulasi terhadap bahan HBG boleh mencetuskan pembentukan banyak lagi bahan yang berguna dan ini disebabkan ciri istimewa HBG iaitu kebolehan penukargantian ion dan penyerakan logam yang tinggi. Dalam kajian ini, sintesis nano komposit penggalak susu getah, kajian perlepasan terkawal penggalak susu getah dan pembentukan KNT telah berjaya dilakukan. Diharapkan pada masa

hadapan, ilmu berkenaan HBG boleh menghasilkan bahan berguna dalam pelbagai bidang.

Dalam kajian ini, HBG zink-aluminium-nitrat (ZAL) telah dapat disintesis pada nisbah logam Zn/Al yang berbeza iaitu, R=2- 4 dan pH=7.50-10.00 melalui kaedah pemendakan di bawah aliran gas nitrogen. Penggalak susu getah iaitu 2,4-dikorofenoksiasetik asid (2,4-D) dan 2-kloroetilfosfonik asid (2-CEPA) telah berjaya disisipkan ke dalam ruang hidroksida berlapis ganda melalui kaedah pemendakan. Beberapa parameter iaitu nisbah kandungan logam dalam bahan pemula iaitu 2, 3 dan 4; kepekatan anion 2,4-D dan 2-CEPA iaitu 0.08 M-0.32 M dan pH iaitu 7.50-10.00 telah diuji dalam sintesis nanokomposit Zn-Al-2,4-D (ZANDI) dan Zn-Al-2-CEPA (ZANE).

Corak pembelauan sinar-X menunjukkan berlaku pengembangan jarak antara ruang perumah bagi menempatkan anion 2,4-D dan 2-CEPA yang saiznya lebih besar daripada anion nitrat. Spektrum FTIR bagi setiap nanokomposit ZANDI dan ZANE yang telah disediakan menyerupai campuran spektrum bahan pemula, ZAL dan anion tetamu, 2,4-D serta 2-CEPA. Ini menunjukkan kehadiran kedua-dua bahan pemula tersebut dalam nanokomposit yang diperolehi. Gabungan analisis corak pembelauan sinar-X dan FTIR mengesahkan lagi pembentukan nanokomposit ZANDI dan ZANE. Pencirian lain terhadap nanokomposit yang diperolehi telah dilakukan seperti analisis terma, analisis kandungan organik-tak organik, luas permukaan juga keliangan dan morfologi permukaan.

Kajian perlepasan terkawal anion 2,4-D dan 2-CEPA telah dijalankan dalam media akueus seperti natrium klorida, natrium karbonat dan natrium fosfat pada kepekatan 0.05 M. Perlepasan 2,4-D dan 2-CEPA di dalam air juga telah dilakukan sebagai kawalan. Didapati bahawa 2,4-D telah dilepaskan secara pantas di dalam natrium karbonat manakala 2-CEPA di dalam natrium fosfat. Walaubagaimanapun, perlepasan 2,4-D di dapati paling maksimum di dalam natrium karbonat pada 99 % dan 2-CEPA di dalam natrium fosfat pada 98 %. Model kinetik pseudo-kedua telah didapati sesuai dengan semua profil perlepasan dengan $r^2 > 0.9$.

Karbon nano tiub yang dihasilkan daripada kalsin CoNiAl-HBG telah menghasilkan hasil yang tinggi apabila dibandingkan dengan kalsin FeCoNiAl-HBG dan FeNiAl-HBG. Walaubagaimanapun, setelah proses penulenan dijalankan, KNT yang dihasilkan daripada kalsin FeCoNiAl-HBG telah diperolehi dengan banyak. KNT yang disintesiskan menggunakan kerajang lapisan nipis logam telah menghasilkan diameter tiub yang lebih kecil dan ciri permukaan yang lebih baik dan ini adalah kerana penyerakan logam yang baik dan tinggi.

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I certify that a Thesis Examination Committee has met on 29 Disember 2010 to conduct the final examination of Adila Mohamad Jaafar on her thesis entitled "Application of Layered Double Hydroxides as Host in Controlled Release Formulation of Latex Stimulant and Metal Catalyst in Formation of Carbon Nanotube" in accordance with the Universities and University College Act 1971 and the Constituition 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:

Mohd Zaki Abd Rahman, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

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

Sidik Silong, PhD
Associate Professor
Faculty of Science
Universiti Putra Malaysia
(Internal Examiner)

Musa Ahmad, PhD
Professor
Faculty of Science and Technology
Universiti Kebangsaan Malaysia
(External Examiner)

_____

BUJANG BIN KIM HUAT, PhD
Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia

Date: 22nd February 2011

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

Mohd. Zobir Hussein, PhD
Professor
Faculty of Science
Universiti Putra Malaysia
(Chairman)

Asmah Hj. Yahaya, PhD
Assoc.Prof
Agriculture Science Foundation Centre
Universiti Putra Malaysia
(Member)

Zulkarnain Zainal, PhD
Professor
Faculty of Science
Universiti Putra Malaysia
(Member)

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

Date: 10th March 2011

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 concurrent, submitted for any other degree at Universiti Putra Malaysia or other institutions.

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ADILA MOHAMAD JAAFAR

DATE:29 December 2010

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