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**A Novel Study of Hydrogen Storage in (Ni-)Mg-Al  
Hydrotalcite-derived Mixed Oxides**

I RUZANNA BINTI IBRAHIM

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A NOVEL STUDY OF HYDROGEN STORAGE IN (Ni-)Mg-Al HYDROTALCITE-  
DERIVED MIXED OXIDES

by

RUZANNA BINTI IBRAHIM

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A NOVEL STUDY OF HYDROGEN STORAGE IN (Ni-)Mg-Al HYDROTALCITE-  
DERIVED MIXED OXIDES

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RUZANNA BINTI IBRAHIM

A Thesis

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CHEMICAL ENGINEERING  
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BANDAR SRI ISKANDAR  
PERAK

SEPTEMBER, 2011

## DECLARATION OF THESIS

Title of thesis

**A Novel Study of Hydrogen Storage in (Ni-)Mg-Al  
Hydrotalcite-derived Mixed Oxides**

I RUZANNA BINTI IBRAHIM

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 UTP or other institutions.

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*Dedicated to my beloved Family*

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## ABSTRACT

Hydrogen was adsorbed using mixed oxides adsorbents derived from (Ni) Mg-Al hydrotalcite-like compounds (HTlcs). This is a novel study on the use of these mixed oxides to adsorb hydrogen as these materials have never been investigated for use as hydrogen storage media. A previous study has only used the as-synthesized HTlcs to adsorb hydrogen and no hydrogen adsorption was observed in the study. In this study the HTlcs were synthesized via the coprecipitation method and were calcined at various temperatures to obtain the corresponding mixed oxides. Characterizations of the as-synthesized and calcined samples were also conducted. XRD characterizations of the as-synthesized samples showed that the hydrotalcite-like phase was obtained and MgO and NiO (only for Ni-Mg-Al HTlcs) were formed after the calcination step. Furthermore, the FTIR showed that the HTlcs contained water and carbonate ions before and after calcination. However, calcination at high temperatures (750°C and 900°C) caused destruction of the hydrotalcite structure which cannot be reconstructed via rehydration. Meanwhile, it was found that the BET specific surface area generally increases after calcinations at moderate temperatures but decreases at higher calcination temperature. Additionally, TPR showed that only a small amount of hydrogen uptake (1.4mmol/g) occurred when the Mg-Al HTlcs were used which may be due to the low reducibility of the material while addition of Ni showed increase in H<sub>2</sub> uptake of Ni-Mg-Al HTlcs (3.3mmol/g) with the same Mg/Al molar ratio. The adsorption of hydrogen following the material reduction in the TPR experiments is indicated by tailings in the reduction profile. Meanwhile, the hydrogen adsorption capacity determined by thermogravimetric analysis conducted at temperatures of 400°C, 200°C and 100°C indicated that Ni-Mg-Al HTlcs with Ni/Mg/Al molar ratio of 1:2:1 adsorbed the highest amount of hydrogen (3 wt% at 100°C). In conclusion, the high reducibility and the hydrotalcite-like structure of the Ni-Mg-Al HTlc make it the most favourable material for hydrogen storage in this study.

## ABSTRAK

Hidrogen telah diserap menggunakan oksida campuran yang diterbitkan dari (Ni)Mg-Al hidrotalsit (HTlcs). Kajian ini adalah pertama kali dijalankan kerana sebelum ini didapati tiada kajian mengenai penggunaan oksida campuran terbitan HTlcs sebagai media penyerapan hydrogen. Penyiasatan sebelum ini hanya menggunakan hidrotalsit yang disintesis untuk menyerap hydrogen tetapi tiada penyerapan hydrogen dilaporkan. Dalam kajian ini, HTlcs telah disintesis menggunakan kaedah pemendakan bersama dan dikalsinasi untuk mendapatkan oksida campuran. Pencirian bahan juga turut dilakukan ke atas sampel sebelum dan selepas kalsinasi. Hasil analisis belauan sinar-X, XRD menunjukkan fasa hidrotalsit telah diperolehi sebelum kalsinasi dan oksida campuran MgO dan NiO (bagi Ni-Mg-Al HTlcs) dihasilkan selepas kalsinasi. Spektroskopi jelmaan Fourier inframerah, FTIR menunjukkan semua HTlcs sebelum dan selepas kalsinasi mengandungi air dan ion karbonat ( $\text{CO}_3^{2-}$ ). Namun, suhu kalsinasi yang tinggi ( $750^\circ\text{C}$  dan  $900^\circ\text{C}$ ) telah memusnahkan struktur hidrotalsit dan tidak dapat dipulihkan melalui rehidrasi. Luas permukaan spesifik BET didapati meningkat setelah sampel melalui proses kalsinasi pada suhu sederhana namun nilai ini berkurang apabila suhu kalsinasi lebih tinggi digunakan. Penurunan berprogram suhu, TPR menunjukkan bahawa hanya sejumlah kecil hidrogen digunakan oleh Mg-Al HTlcs ( $1.4\text{mmol/g}$ ) kemungkinan disebabkan oleh kadar penurunan bahan yang rendah tetapi nilai penggunaan hidrogen ini meningkat dengan penambahan logam nikel ( $3.3\text{mmol/g}$ ). Penyerapan hidrogen dalam kaedah TPR dapat dikesan dari bentuk profil penurunan. Penyerapan menggunakan analisis gravimetri termal, TGA yang telah dilakukan pada suhu  $400^\circ\text{C}$ ,  $200^\circ\text{C}$  dan  $100^\circ\text{C}$  menunjukkan Ni-Mg-Al HTlcs dengan nisbah molar Ni/Mg/Al sebanyak 1:2:1 telah menyerap jumlah hidrogen yang paling banyak (3wt% pada  $100^\circ\text{C}$ ). Kesimpulannya, kadar penurunan yang tinggi dan struktur hidrotalsit yang dimiliki oleh Ni-Mg-Al HTlcs menjadikannya lebih sesuai untuk menyerap hidrogen dalam kajian ini.

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## TABLE OF CONTENTS

	PAGE
STATUS OF THESIS	i
APPROVAL PAGE	ii
TITLE PAGE	iii
DECLARATION	iv
DEDICATION	v
ACKNOWLEDGEMENT	vi
ABSTRACT	vii
COPYRIGHT PAGE	ix
LIST OF TABLES	xiii
LIST OF FIGURES	xiv
LIST OF EQUATIONS	xvii
PREFACE	xviii
Chapter	
1. INTRODUCTION	1
1.1 Background of Study	1
1.2 Problem Statement	4
1.3 Objectives of Study	5
1.4 Scopes of Study	5
1.5 Significance of Study	6
1.6 Thesis Outline	6
2. LITERATURE REVIEW	7
2.1 Introduction	7
2.2 Adsorption	7
2.2.1 Physisorption and Chemisorption	8
2.2.2 Adsorption Forces and Energies	10
2.2.3 Rate of Adsorption and Adsorption Equilibrium	11
2.2.4 Adsorption Isotherms	12
2.2.4.1 IUPAC Classification of Adsorption Isotherms	13
2.2.4.2 Langmuir Adsorption Isotherm	15

2.3	Hydrogen Storage Technologies	15
2.3.1	Physically Bound Hydrogen	16
2.3.2	Chemically Bound Hydrogen	17
2.3.3	Hydrolytic Evolution of Hydrogen	18
2.4	Investigations on Solid Hydrogen Storage	19
2.4.1	Hydrogen Adsorption on Metal Hydrides	19
2.4.2	Hydrogen Adsorption on Activated Carbon	20
2.4.3	Hydrogen Adsorption on Other Carbon Materials	21
2.4.4	Hydrogen Adsorption by Metal Oxides	22
2.4.5	Hydrogen Adsorption by Metal Organic Frameworks (MOFs)	23
2.4.4	Hydrogen Adsorption by Zeolites	24
2.5	Hydrotalcite-like compounds	24
2.5.1	HTlcs Preparation Methods	26
2.5.1.1	Coprecipitation Method	26
2.5.1.2	Hydrothermal Method	27
2.5.1.3	Urea Hydrolysis Method	28
2.5.1.4	Sol-Gel Method	29
2.5.2	Applications of Hydrotalcite-like Compounds	30
2.5.2.1	Applications in Gas Adsorption	30
2.5.2.2	Other Applications of HTlcs	30
2.5.3	Thermal Decomposition of Hydrotalcite-like Compounds	31
2.5.5	Calcination Effects on HTlcs	33
2.6	Properties of Hydrotalcite-like Compounds	33
2.6.1	Phase Determination using X-ray Powder Diffraction	34
2.6.2	Characterization by Fourier Transform Infrared Spectroscopy	35
2.6.3	Morphology Determination by Field Emission Scanning Electron Microscope	35
2.6.4	Elemental Analysis via Energy Dispersive X-Ray	36
2.6.5	Textural Properties Determination by Nitrogen Adsorption	36
2.6.6	Material Reducibility Determination by Temperature Programmed Reduction	37
2.7	Hydrogen Adsorption using Thermogravimetric Analysis	37
2.8	Summary	38
3.	METHODOLOGY	39
3.1	Introduction	39
3.2	Mg-Al HTlcs Synthesis by Coprecipitation	39
3.3	Ni-Mg-Al HTlcs Synthesis by Coprecipitation	40
3.4	Material Characterization	41
3.4.1	Powder X-ray Diffraction (XRD)	41
3.4.2	Fourier Transform Infrared Spectroscopy (FTIR)	44
3.4.3	Field Emission Scanning Electron Microscopy with Energy Dispersive X-ray (FESEM-EDX)	47
3.4.4	Surface Area and Porosity Determination by N <sub>2</sub> Adsorption	48

3.4.5	Temperature-Programmed Reduction (TPR)	49
3.5	Thermogravimetric Analysis (TGA)	51
4.	RESULTS AND DISCUSSION	55
4.1	Introduction	55
4.2	X-Ray Diffraction (XRD) Results	55
4.2.1	XRD of As-synthesized Mg-Al HTlcs	55
4.2.2	XRD of Calcined Mg-Al HTlcs	57
4.2.3	XRD of As-synthesized Ni-Mg-Al HTlcs	62
4.2.4	XRD of Calcined Ni-Mg-Al HTlcs	63
4.3	Fourier Transform Infrared Spectroscopy (FTIR) Results	65
4.3.1	FTIR of As-synthesized Mg-Al HTlcs	65
4.3.2	FTIR of Calcined Mg-Al HTlcs	66
4.3.3	FTIR of As-synthesized Ni-Mg-Al HTlcs	70
4.3.4	FTIR of Calcined Ni-Mg-Al HTlcs	72
4.4	BET Surface Area and Porosity Results	74
4.5	Field Emission Scanning Electron Microscope (FESEM)	76
4.6	Energy Dispersive X-ray (EDX) Results	81
4.7	Temperature Programmed Reduction (TPR) Results	82
4.7.1	TPR of Mg-Al HTlcs	83
4.7.2	TPR of Ni-Mg-Al HTlcs	87
4.7.3	Hydrogen Consumption by the Materials during TPR	90
4.8	Thermogravimetric Analysis Results	92
5.	CONCLUSION	101
5.1	Introduction	101
5.2	Conclusions of the Study	101
5.3	Recommendations	102
	REFERENCES	103
	APPENDICES	119
Appendix 1	Experimental Calculations	119
1.1	Calculations for amount of materials required to synthesis Mg-Al HTlcs	119
1.2	Calculations for amount of materials required to synthesis Ni-Mg-Al HTlcs	122
Appendix 2	BET Isotherms	125
Appendix 3	List of Publications and Presentations	127
3.1	Journal Paper	127
3.2	Conference Papers	127
3.3	Symposium Papers	128

## LIST OF TABLES

Table		Page
2.1	Features of Physisorption and Chemisorption	9
4.1	Textural Properties of the Synthesized Hydrotalcites	75
4.2	Chemical Composition from EDX Analysis of Hydrotalcite-like Compounds	81
4.3	The weight decrease of HTlcs samples after the pretreatment stage	94
4.4	Initial Rates of Adsorption at 400°C for Mg-Al HTlcs	99

## LIST OF FIGURES

Figure		Page
2.1	Gas physisorption isotherms according to IUPAC classification	13
2.2	The structure of hydrotalcite-like compounds	25
3.1	Paths of X-ray beams for Bragg's Law	42
3.2	Diagram of Michelson Interferometer	46
3.3	Reduction Profile	49
3.4	The temperature profile for Temperature-Programmed Reduction experiments	51
3.5	Temperature Profile for Thermogravimetric Analysis of HTlcs	52
4.1	The XRD patterns of Mg-Al HTlcs, Mg/Al molar ratio 2, 3 and 4	56
4.2	The XRD patterns of Mg-Al HTlcs calcined at 450°C, Mg/Al molar ratio 2, 3 and 4	58
4.3	The XRD patterns of Mg-Al HTlcs calcined at 600°C, Mg/Al molar ratio 2, 3 and 4	59
4.4	The XRD patterns of Mg-Al HTlcs calcined at 750°C, Mg/Al molar ratio 2, 3 and 4	60
4.5	The XRD patterns of Mg-Al HTlcs calcined at 900°C, Mg/Al molar ratio 2, 3 and 4	61
4.6	The XRD patterns for the Ni-Mg-Al HTlcs	62
4.7	XRD pattern for Ni-Mg-Al HTlcs calcined at 600°C (top) and 450°C (bottom) with Ni/Mg/Al molar ratio of 1:2:1	64
4.8	The IR spectra of as-synthesized Mg-Al HTlcs with Mg/Al molar ratio of 2 (bottom), 3 (middle) and 4 (top)	65

4.9	The IR spectra of Calcined Mg-Al HTlcs with Mg/Al molar ratio of 2 (bottom), 3 (middle) and 4 (top) calcined at 450°C	67
4.10	The IR spectra of as-synthesized (top) and calcined (bottom) Mg-Al HTlc with Mg/Al ratio of 2	68
4.11	The IR spectra of Calcined Mg-Al HTlcs with Mg/Al molar ratio of 2 (bottom), 3 (middle) and 4 (top) calcined at 750°C	69
4.12	The IR spectra of Calcined Mg-Al HTlcs with Mg/Al molar ratio of 2 (bottom), 3 (middle) and 4 (top) calcined at 750°C	70
4.13	The IR spectra of as-synthesized Ni-Mg-Al HTlcs with Ni/Mg/Al molar ratio of 1:2:1 (bottom) and 2:1:1 (top)	71
4.14	IR spectra of Ni-Mg-Al HTlc (1:2:1) calcined at 450°C and 600°C	73
4.15	IR spectra of as-synthesized (bottom) and 450°C -calcined (top) Ni-Mg-Al HTlc (1:2:1)	73
4.16	FESEM image for Mg-Al HTlc with Mg/Al molar ratio of 2	77
4.17	FESEM image for Mg-Al HTlc with Mg/Al molar ratio of 3	77
4.18	FESEM image for Mg-Al HTlc with Mg/Al molar ratio of 3 calcined at 450°C	78
4.19	FESEM image of Ni-Mg-Al HTlcs with Ni/Mg/Al molar ratio of (1:2:1)	79
4.20	FESEM image of Ni-Mg-Al HTlcs with Ni/Mg/Al molar ratio (2:1:1)	79
4.21	FESEM image of Ni-Mg-Al HTlcs with Ni/Mg/Al molar ratio (2:1:1) calcined at 450°C	80
4.22	The FESEM image of Ni-Mg-Al HTlcs with Ni/Mg/Al molar ratio (2:1:1) calcined at 600°C	80
4.23	Reduction profile for Mg-Al HTlcs calcined at 450°C (the molar ratio of samples are indicated in the bottom legend)	83
4.24	Reduction profile for Mg-Al HTlcs calcined at 600°C	84
4.25	Reduction profile for Mg-Al HTlcs calcined at 750°C	84

4.26	Reduction profile for Mg-Al HTlcs calcined at 900°C	85
4.27	XRD pattern for Mg-Al HTlcs calcined at 600°C (a) before reduction and (b) after reduction	86
4.28	Reduction profile of Ni-Mg-Al HTlcs with Ni/Mg/Al ratio of 1:2:1	87
4.29	Reduction profile of Ni-Mg-Al HTlcs with Ni/Mg/Al ratio of 2:1:1	87
4.30	Reduction profile of Ni-Mg-Al HTlcs with Ni/Mg/Al ratio 1:2:1 calcined at 450°C	89
4.31	Reduction profile of Ni-Mg-Al HTlcs with Ni/Mg/Al ratio 1:2:1 calcined at 600°C	90
4.32	Hydrogen consumption of calcined Mg-Al HTlcs during TPR	91
4.33	Hydrogen consumption of Ni-Mg-Al HTlcs during TPR	92
4.34	TG Curve of as-synthesized Mg-Al HTlc with Mg/Al molar ratio of 2	93
4.35	The adsorption branch of the TG Curve of as-synthesized Mg-Al HTlc with Mg/Al molar ratio of 2	95
4.36	Hydrogen adsorption capacity of the HTlcs samples at various temperatures	97
4.37	Plot of adsorption amount versus time for Mg-Al HTlc with Mg/Al molar ratio of 2	98

## LIST OF EQUATIONS

Equation		Page
2.1	Equation for Adsorption Energy	10
2.2	Equation for Adsorption Energy	11
2.3	Equation for Adsorption Isotherm	12
2.4	Langmuir adsorption isotherm	15
2.5	Reaction of metal hydride formation	17
2.6	Reaction for Hydrolytic Evolution of Hydrogen	19
2.7	Reaction for Hydrolytic Evolution of Hydrogen	19
2.8	Urea Hydrolysis Reaction (Step 1)	28
2.9	Urea Hydrolysis Reaction (Step 2)	28
3.1	Bragg's Law	42
3.2	BET Equation	48
3.3	BET Surface Area for monolayer	48
4.1	First weight loss due to elimination of water	93
4.2	Second weight loss and formation of mixed oxide	93
4.3	Linear Driving force model	97
4.4	Degree of Adsorption	98
4.5	Logarithmic version of Linear Driving force model	98

## PREFACE

In recent years, there has been extensive research on the potential of using hydrogen as an alternative to fossil fuels. However, a major barrier that has to be overcome in order to make hydrogen economy a reality is the lack of efficient hydrogen storage technology. Conventional storage methods such as compressed hydrogen and liquefied hydrogen have limitations in terms of space and safety and high energy consumption. Another potential approach of storing hydrogen is by adsorption in solid adsorbents such as metal hydrides, carbon materials, metal organic frameworks, zeolites and metal oxides but each material has their own limitations for hydrogen storage applications. Hydrotalcite-derived mixed oxides is a new material worth investigating for hydrogen storage potential as these materials have adsorbent characteristics such as high specific surface area and thermal stability. This study involves the synthesis of (Ni-)Mg-Al Hydrotalcite-like compounds and the respective mixed oxides, and characterization and testing for their hydrogen adsorption capability.

