

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

Bi-Fe DOP DAN RAWATAN MEKANOKIMIA MANGKIN VANADIUM FOSFAT UNTUK PENGOKSIDAAN TERPILIH *n*-BUTANA KEPADA MALEIK ANHIDRIDA

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Pengenalan Bi-Fe sebagai dopan dan rawatan secara mekanokimia telah digunakan dalam penyediaan prekursor mangkin, $\text{VOHPO}_4 \cdot 0.5\text{H}_2\text{O}$ yang telah disintesiskan melalui penurunan $\text{VOPO}_4 \cdot 2\text{H}_2\text{O}$ menggunakan isobutanol dan diikuti dengan pengkalsinan dalam aliran campuran *n*-butana/udara (0.75% *n*-butana dalam udara) untuk 75 j pada 673 K. Beberapa kaedah seperti XRD, SEM, ICP-AES, pengukuran luas permukaan, titratan redox, H_2 -TPR dan O_2 -TPD telah digunakan untuk mencirikan mangkin. Ciri-ciri kemangkinan untuk pengoksidaan terpilih *n*-butana juga telah dibuat. Keputusan eksperimen telah menunjukkan tambahan dopan Bi-Fe ke dalam kekisi mangkin vanadium fosfat telah mengurangkan luas permukaan mangkin, kecuali mangkin yang didop dengan kepekatan Bi yang lebih tinggi. Selain itu, mangkin dop Bi-Fe telah mengalakkan pembentukan fasa α_{II} - VOPO_4 dan sebagai akibatnya mengurangkan struktur $(\text{VO})_2\text{P}_2\text{O}_7$. Ia didapati

bahawa morfologi mangkin dop Bi-Fe adalah dipengaruhi oleh nisbah dopan/V. Mangkin dop Bi-Fe adalah didapati memberi kesan buruk kepada penukaran *n*-butana. Walau demikian, selektiviti maleik anhidrida telah meningkat disebabkan oleh kewujudan lebih fasa V^{5+} . Saling-kait yang baik telah ddapati antara penukaran *n*-butana dengan jumlah spesies oksigen yang berkaitan dengan fasa V^{4+} . Rawatan secara mekanokimia menggunakan tenaga tinggi kisaran bebola pada prekursor mangkin, $VOHPO_4 \cdot 0.5H_2O$ telah meningkatkan FWHM dan mengurangkan saiz hablur dan sebagai akibatnya meningkatkan luas permukaan mereka. Walau demikian, kisaran lebih lama kepada 120 min telah menyebabkan penurunan dalam luas permukaan mangkin yang disebabkan oleh penggumpalan semula bahan. Morfologi mangkin yang berbentuk ros telah hilang selepas proses pengisaran. Kisaran bebola untuk 60 min menunjukkan peningkatan reaktiviti dan mobiliti kekisi oksigen dengan merendahkan suhu penurunan dan menggalakkan lebih banyak amaun spesies oksigen telah dilepaskan. Penambahan spesies oksigen berkait dengan fasa V^{4+} yang saling-kait kepada aktiviti mangkin dan spesies oksigen berkait dengan V^{5+} yang lebih sedikit juga menyumbang kepada aktiviti. Dalam kes yang menggunakan media kisaran berlainan, mangkin kisaran dalam medium kering mengurangkan luas permukaan yang disebabkan oleh timbunan pepejal yang pecah, di mana ia mungkin merupakan sumbangan utama sifat kemungkinan yang lemah. Penggunaan larutan seperti sikloheksana dan etanol memperbaiki luas permukaan dan morfologi mangkin. Mangkin yang 30 min kisaran dalam etanol menunjukkan selektiviti maleik anhidrida paling tinggi dibandingkan mangkin yang tidak dikisar atau dikisar dalam medium kering dan

sikloheksana di bawah keadaan yang sama. Walau demikian, sikloheksana adalah didapati larutan yang paling baik untuk penukaran *n*-butana untuk bahan yang 30 min kisaran.

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I certify that an Examination Committee has met on 12th June 2006 to conduct the final examination of Goh Chee Keong on his Doctor of Philosophy thesis entitled “Bi-Fe Doping and Mechanochemical Treatment of Vanadium Phosphate Catalysts for Selective Oxidation of N-butane to Maleic Anhydride” 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 are as follows:

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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.

GOH CHEE KEONG

Date: 22 October 2006

TABLE OF CONTENTS

	Page
DEDICATION	ii
ABSTRACT	iii
ABSTRAK	v
ACKNOWLEDGEMENTS	vii
APPROVAL	viii
DECLARATION	x
LIST OF TABLES	xiv
LIST OF FIGURES	xvi
LIST OF ABBREVIATIONS	xix
CHAPTER	
1 INTRODUCTION	1
1.1 Heterogeneous Catalysis	1
1.2 Selective Oxidation Reactions	3
1.3 Selectivity and Active Site in Catalytic Oxidation	6
1.4 Selective Oxidation of Light Alkanes	8
1.5 Summary and Objectives	10
2 LITERATURE REVIEW	14
2.1 Introduction	14
2.2 Production of Maleic Anhydride	14
2.3 Vanadium Phosphorus Oxide (VPO) Catalysts	19
2.4 Role of Preparation Method of Vanadium Phosphorus Oxide Catalysts	20
2.4.1 Method of Preparation of Active Phase Precursor	21
2.4.2 Influence of P/V Ratio	25
2.4.3 Activation	27
2.5 Role of Redox Properties of Vanadium Phosphorus Oxide Catalysts	29

2.6 Nature of Active Site of Vanadyl Pyrophosphate Catalysts	32
2.7 Role of Promoters	34
2.8 Mechanochemically Assisted Synthesis of Vanadium Phosphate Catalysts	45
3 MATERIALS AND METHODS	51
3.1 Materials and Gases	51
3.2 Preparation of VOPO ₄ ·2H ₂ O	52
3.3 Preparation of Bi-Fe Oxide (Bi/Fe = 2) Complex	52
3.4 Preparation of Undoped Precursors	52
3.5 Preparation of Bi-Fe Doped Precursors	53
3.6 Preparation of Precursors Doped with Bi-Fe Oxide (Bi/Fe = 2)	53
3.7 Mechanochemical Treatment of VOHPO ₄ ·0.5H ₂ O with Bi-Fe Oxide (Bi/Fe = 2)	54
3.8 Mechanochemical Treatment of VOHPO ₄ ·0.5H ₂ O	54
3.9 Catalysts Activation	55
3.10 Catalysts Characterisation	55
3.10.1 Bulk Composition Analysis	55
3.10.2 Volumetric Titration Method	56
3.10.3 Surface Area Measurements	57
3.10.4 X-ray Diffraction (XRD) Analysis	58
3.10.5 Laser Raman Spectroscopy (LRS)	58
3.10.6 Scanning Electron Microscopy (SEM) Analysis	58
3.10.7 Temperature-Programmed Analyses	59
3.11 Catalytic Test	60
3.11.1 Microreactor Design	61
3.11.2 Microreactor Operation	62
4 PHYSICOCHEMICAL STUDY AND CATALYTIC PROPERTIES OF Bi-Fe DOPED VANADIUM PHOSPHATE (VPO) CATALYSTS	63
4.1 Introduction	63
4.2 Vanadium Phosphate Catalysts Doped with Bi-Fe Nitrate	65
4.2.1 BET Surface Area and Chemical Analysis	65
4.2.2 X-Ray Diffraction (XRD)	67
4.2.3 Scanning Electron Microscopy (SEM)	71
4.2.4 Oxygen Temperature-programmed Desorption (O ₂ -TPD)	74
4.2.5 Temperature-programmed Reduction (TPR) in H ₂ /Ar	76
4.2.6 Catalytic Test	81
4.3 Vanadium Phosphate Catalysts Doped with Bi-Fe Oxide	86
4.3.1 BET Surface Area and Chemical Analysis	86
4.3.2 X-Ray Diffraction (XRD)	88
4.3.3 Laser Raman Spectroscopy (LRS)	92
4.3.4 Scanning Electron Microscopy (SEM)	93
4.3.5 Oxygen Temperature-programmed Desorption (O ₂ -TPD)	94

	4.3.6 Temperature-programmed Reduction (TPR) in H ₂ /Ar	96
	4.3.7 Catalytic Test	99
	4.4 Conclusions	102
5	EFFECT OF MECHANOCHEMICAL TREATMENT ON THE PROPERTIES OF VANADIUM PHOSPHATE (VPO) CATALYSTS	103
	5.1 Introduction	103
	5.2 Mechanochemical Treatment of VPO Catalysts in Dependent on	
	Time	104
	5.2.1 BET Surface Area and Chemical Analysis	104
	5.2.2 X-Ray Diffraction (XRD)	106
	5.2.3 Scanning Electron Microscopy (SEM)	111
	5.2.4 Temperature-programmed Reduction (TPR) in H ₂ /Ar	114
	5.2.5 Oxygen Temperature-programmed Desorption (O ₂ -TPD)	116
	5.2.6 Catalytic Test	119
	5.3 Influence of Milling Media	128
	5.3.1 BET Surface Area and Chemical Analysis	128
	5.3.2 X-Ray Diffraction (XRD)	130
	5.3.3 Scanning Electron Microscopy (SEM)	134
	5.3.4 Temperature-programmed Reduction (TPR) in H ₂ /Ar	136
	5.3.5 Oxygen Temperature-programmed Desorption (O ₂ -TPD)	138
	5.3.6 Catalytic Test	141
	5.4 Conclusions	145
6	CONCLUSIONS	147
	REFERENCES	150
	APPENDIX	162
	BIODATA OF THE AUTHOR	166