



**UNIVERSITI PUTRA MALAYSIA**

***EFFECT OF VANADIUM SOURCE AND DIFFERENT DRYING TECHNIQUES ON  
THE PROPERTIES OF MoVTenb OXIDE CATALYSTS PREPARED BY REFLUX  
METHOD***

**CHE KU NOR LIANA BINTI CHE KU HITAM**

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**MASTER OF SCIENCE  
UNIVERSITI PUTRA MALAYSIA**

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REFLUX METHOD**

By

**CHE KU NOR LIANA BINTI CHE KU HITAM**

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia, in  
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**October 2012**

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**October 2012**

**Chairman: Associate Professor Irmawati Ramli, PhD**

**Faculty: Science**

MoVTeNb oxide has been reported as an active and selective catalyst for the direct oxidation of propane to acrylic acid. However, its catalytic performance is closely related to the preparation of the catalysts. Therefore, the investigation of the preparation parameters for a catalyst is important for the understanding of the effects on the catalyst structure and the performance. In this study, MoVTeNb oxide samples were prepared via reflux technique. This technique is economical since the apparatus are very simple and easy to assemble. The ratio of Mo : V : Te : Nb was 1 : 0.3 : 0.16 : 0.12. The aims of this project were to investigate the effect of different vanadium sources and drying techniques on the physico-chemical properties of MoVTeNb oxide catalysts. The x-ray diffraction (XRD) analysis indicated that the precursors were of Anderson-type which is commonly found in amorphous synthesized species. Phase evolutions were observed; with all the MoVTeNb oxide samples are highly crystalline when they were calcined at

553 K in air and 873 K in N<sub>2</sub> flow. In studying the effect of vanadium sources, the sample synthesized using vanadium pentoxide (V<sub>2</sub>O<sub>5</sub>), designated as MoVTeNb<sub>V<sub>2</sub>O<sub>5</sub>cal</sub> showed a better catalytic performance compared to that of vanadium oxide sulphate (VOSO<sub>4</sub>) sample (MoVTeNb<sub>VOSO<sub>4</sub>cal</sub>). This is due to the former contains orthorhombic M1 and hexagonal M2 phases which are the active and selective phase of the MoVTeNbO catalyst. The preparation of the samples was also varied by employing different drying methods which are spray drying (MoVTeNb<sub>spr</sub>), rotary evaporation (MoVTeNb<sub>rot</sub>) and oven drying (MoVTeNb<sub>oven</sub>). Significant XRD peaks corresponded to the desired phases, which are orthorhombic M1 and hexagonal M2, were observed from the spray dried sample, but none was observed from the rotary evaporated and oven dried samples. Furthermore, the spray drying method prevented the compositional segregation of the sample, hence proved to be the best drying method for the preparation of this catalyst. The elemental composition was similar to the nominal composition of the raw materials used (Mo<sub>1.0</sub>V<sub>0.30</sub>Te<sub>0.16</sub>Nb<sub>0.12</sub>) as shown in the inductively coupled plasma-atomic emission spectroscopy (ICP-AES) analysis. The catalytic test proved that the catalyst dried by this technique is active and selective for the oxidation of propane to acrylic acid.

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

**KESAN SUMBER VANADIUM DAN TEKNIK PENERINGAN BERBEZA TERHADAP SIFAT MANGKIN MoVTenNb OKSIDA YANG DISEDIAKAN MELALUI KAEDAH REFLUKS**

Oleh

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MoVTenNb oksida telah dilaporkan sebagai mangkin yang aktif dan selektif untuk pengoksidaan terus propana kepada asid akrilik. Walau bagaimanapun, pencapaian pemangkinannya berkait rapat dengan penyediaan mangkin. Oleh itu, kajian terhadap parameter penyediaan mangkin adalah penting untuk pemahaman tentang kesannya terhadap struktur dan pencapaian mangkin. Dalam kajian ini, sampel MoVTenNb oksida telah disediakan melalui teknik refluks. Teknik ini menjimatkan kerana peralatannya sangat mudah dan senang dipasang. Nisbah Mo : V : Te : Nb ialah 1 : 0.3 : 0.16 : 0.12. Matlamat projek ini adalah untuk mengkaji kesan sumber vanadium dan kaedah pengeringan yang berbeza terhadap ciri-ciri kimia-fizik bagi mangkin MoVTenNb oksida. Analisis pembelauan sinar-x (XRD) menunjukkan bahawa prekursor-prekursor adalah daripada jenis Anderson yang biasanya dijumpai pada spesies amorfos yang dihasilkan. Evolusi fasa telah diperhatikan, dengan kesemua sampel MoVTenNb oksida berada dalam keadaan hablur yang baik apabila dikalsin pada suhu 553 K dalam udara dan suhu

873 K dalam aliran  $N_2$ . Dalam mengkaji kesan sumber vanadium, sampel yang disediakan menggunakan vanadium pentoksida ( $V_2O_5$ ), dinamakan sebagai  $MoVTeNb_{V_2O_5cal}$  menunjukkan pencapaian pemangkinan yang lebih baik berbanding sampel vanadium oksida sulfat ( $VOSO_4$ ) ( $MoVTeNb_{VOSO_4cal}$ ). Ini kerana sampel sebelumnya mengandungi fasa-fasa ortorombus M1 dan heksagonal M2 iaitu fasa yang aktif dan selektif untuk mangkin  $MoVTeNbO$ . Penyediaan sampel juga dibezakan dengan kaedah pengeringan yang berbeza iaitu penyemburan kering ( $MoVTeNbspr$ ), penyejatan berputar ( $MoVTeNbrot$ ) dan pengeringan oven ( $MoVTeNboven$ ). Puncak-puncak penting XRD yang mewakili fasa-fasa yang dikehendaki, iaitu ortorombus M1 dan heksagonal M2, dapat diperhatikan daripada sampel penyemburan kering, tapi tiada satu pun dapat diperhatikan daripada sampel penyejatan berputar dan pengeringan oven. Tambahan pula, kaedah penyemburan kering menghalang pengasingan komposisi sampel, maka membuktikan bahawa ia adalah kaedah pengeringan terbaik dalam penyediaan mangkin ini. Komposisi unsur sama dengan komposisi nominal bahan-bahan awal yang digunakan ( $Mo_{1.0}V_{0.30}Te_{0.16}Nb_{0.12}$ ) seperti yang ditunjukkan dalam analisis spektroskopi penggandingan plasma teraruh-pancaran atom (ICP-AES). Ujian pemangkinan membuktikan bahawa mangkin yang dikeringkan dengan kaedah ini adalah aktif dan selektif terhadap pengoksidaan propana kepada asid akrilik.

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I certify that a Thesis Examination Committee has met on 8 October 2012 to conduct the final examination of Che Ku Nor Liana binti Che Ku Hitam on her thesis entitled “Effect of Vanadium Source and Different Drying Techniques on the Properties of MoVTeNb Oxide Catalysts Prepared by Reflux Method” 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 Master of Science.

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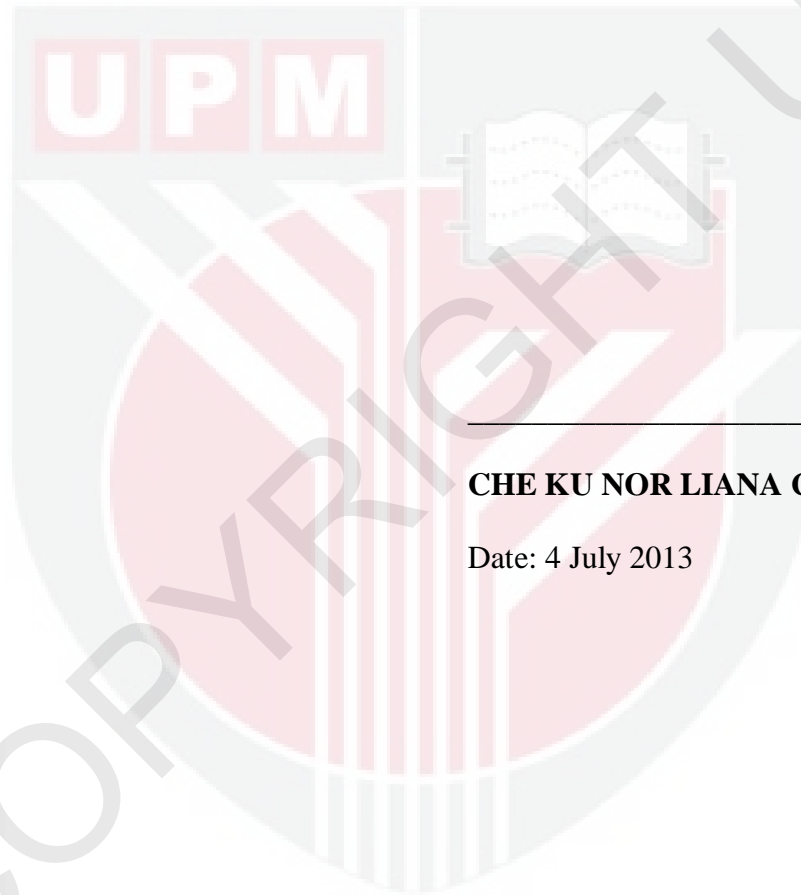
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## DECLARATION

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 at any other institution.



**CHE KU NOR LIANA CHE KU HITAM**

Date: 4 July 2013

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