UNIVERSITI TEKNOLOGI MARA

DEVELOPMENT AND CHARACTERIZATION OF A NOVEL, LIGHTWEIGHT AND POROUS MATERIAL FORMED VIA STRONG COVALENT ORGANIC FRAMEWORKS FOR HYDROGEN STORAGE

MUHAMMAD AFIQ AIZUDDIN B MUSA

Thesis submitted in fulfilment of the requirements for the degree of Master of Science

Faculty of Chemical Engineering

JUNE 2011

CANDIDATE'S DECLARATION

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Name of Candidate : MUHAMMAD AFIQ AIZUDDIN B MUSA

Candidate's ID No. : 2008294588

Programme : MASTER SCIENCE IN CHEMICAL ENGINEERING (RESEARCH)

Faculty : FACULTY OF CHEMICAL ENGINEERING

Thesis Title : DEVELOPMENT AND CHARACTERIZATION OF A NOVEL, LIGHTWEIGHT AND POROUS MATERIAL FORMED VIA STRONG COVALENT ORGANIC FRAMEWORK FOR HYDROGEN STORAGE

Signature of Candidate : .....

Date : .....

ABSTRACT

The depletion of fossil fuels and the ever present issue of global warming has spawned significant research in cleaner technology, which aims to achieve zero emissions. Whilst considering alternatives to fossil fuels, researcher came upon hydrogen; a safe, abundant, high energy content, renewable and more economical than current fuels. The crux of the problem in replacing fossil fuels with hydrogen is that there is lack of lightweight, high capacity storage systems, which are necessary in order to ensure wider application, especially in the transportation industry. The implementation of nanoporous materials is becoming increasingly more prevalent due to their unique characteristics, which make them highly applicable in a multitude of different functionalities. Covalent Organic Frameworks (COFs) are one of the new types of nanoporous material constructed from covalently bonded non-metals. COFs exhibit several advantages such as low density, high thermal stability and the framework can be tailored for specific applications. Modifying the well-established COF-1 synthesis route, three new COFs have been synthesized; namely COF-Sb_2O_3, Sb_2O_3/mesitylene/dioxane through the addition of antimony trioxide and MeCOF-1 through the addition of silicon oxide. Analysis of the new frameworks with respect to published COF-1 analyses it is evident that the syntheses have been successful and that there are inherent similarities due to the use of common precursor building blocks (BDBA, mesitylene and dioxane). The COF-1 synthesized in this research exhibits the highest BET surface of all the synthesized compounds; 681 m^2/g, followed by MeCOF-1; 557.30 m^2/g, COF-Sb_2O_3; 130.34 m^2/g and Sb_2O_3/mesitylene/dioxane; 80.44 m^2/g. The newly synthesized MeCOF-1 out performs COF-1 with respect to pore volume (0.96 cm^3/g) and exhibits a highly mesoporous structure with an average pore diameter of 29.0 Å compared to that for COF-1 (0.39 cm^3/g, 20.0 Å), COF-Sb_2O_3 (0.16 cm^3/g, 13.6 Å) and Sb_2O_3/mesitylene/dioxane (0.10 cm^3/g and 25.8 Å); consequently MeCOF-1 exhibits a higher N_2 uptake than COF-1, COF-Sb_2O_3 and Sb_2O_3/mesitylene/dioxane. MeCOF-1 exhibits a classic IUPAC Type IV isotherm during nitrogen adsorption-desorption analysis thereby classifying it as a highly mesoporous compound, whereas COF-1, COF-Sb_2O_3 and Sb_2O_3/mesitylene/dioxane exhibit typical IUPAC Type I isotherms and are classified as microporous compounds. COF-1 exhibits a relatively high hydrogen uptake of 40 m^3/g compared to COF-Sb_2O_3 and Sb_2O_3/mesitylene/dioxane, which exhibit uptakes less than half that of COF-1. Hydrogen gas adsorption-desorption analysis was not performed on MeCOF-1 due to time constraints and inconsistencies in the experimental equipment, however based upon the other determined physical characteristics MeCOF-1 has the potential to outperform COF-1 due to its higher pore volume and pore diameter that present of both micro- and meso-pore in the synthesize structure. TGA results indicate that MeCOF-1 is thermally stable up to 1000°C, which is at least as good as COF-1 and better than the other two synthesized frameworks. All of the findings indicate that the synthesis of this new mesoporous framework, MeCOF-1, has the potential for storing large volumes of hydrogen gas and may provide a suitable stepping stone in the development of better tailored frameworks with even more extensive storage capacities thereby opening the possibility for generic hydrogen-energy applications with zero emissions.

ACKNOWLEDGEMENTS

Firstly of all, my deepest gratitude and thanks to ALLAH S.W.T for blessing and strengthening me to endure all the challenges that I need to go through to complete my research. I would also like to express my sincere and enormous gratitude to my supervisor, Dr. Robert Michael Savory, and my co-supervisor, Dr. Yin Chun Yang, for their knowledge, enthusiasm, valuable discussion, suggestions, support and encouragement throughout this study. Their help and guidance are greatly appreciated.

Special thanks to the Dean of the Faculty of Chemical Engineering, Prof. Dr. Sharifah Aishah Syed Abd Kadir, the Head of Postgraduate Studies, Dr. Kamariah Noor Ismail for their support during this work. I would also like to extend my gratitude to all the lecturers, technicians and administrative staff in the faculty for helping me in completing my research. Not forgetting of course, all my postgraduate friends who have helped me with my research, given me encouragement and suggestions that have culminated in the successful conclusion of my studies and more importantly making my life as a postgraduate student enjoyable.

I would like to thank the Ministry of Science, Technology and Innovation (MOSTI), Malaysia for providing the E-Science grant with which the chemicals and apparatus necessary for my research were bought. Thanks also go to MOSTI and IPSIS for awarding me with a PGD scholarship for the duration of my Master's programme.

Last but not least, my deepest appreciation goes to my father, Hj Musa B Muidu, my mother, Hjh Mariam Bt Lebam, my brothers ,my sisters and my friends, for all their support, which has given me the strength and spirit to complete this thesis.

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