



UNIVERSITI PUTRA MALAYSIA

**REMOVAL OF HYDROGEN SULFIDE BY PHYSICO-BIOLOGICAL
FILTRATION USING DRIED ACTIVATED SLUDGE AND RICE HUSK
SILICA**

SEYED MAHMOUD MEHDINIA CHOUBI

FPAS 2011 2



**REMOVAL OF HYDROGEN SULFIDE BY PHYSICO-BIOLOGICAL
FILTRATION USING DRIED ACTIVATED SLUDGE
AND RICE HUSK SILICA**

BY

SEYED MAHMOUD MEHDINIA CHOUBI

**Thesis Submitted to the School of Graduate Studies, Universiti Putra Malaysia,
In Fulfilment of the Requirements for the Degree of Doctor of Philosophy**

April 2011



DEDICATION

This work is dedicated to my wife, son, daughter, father and mother, brother and sisters who are to be patient for praying and waiting for me to complete my study



Abstract of thesis presented to the Senate of Universiti Putra Malaysia
In fulfilment of the requirement for the degree of Doctor of Philosophy

**REMOVAL OF HYDROGEN SULFIDE BY PHYSICO-BIOLOGICAL
FILTRATION USING BIOFILM AND RICE HUSK SILICA.**

By

SEYED MAHMOUD MEHDINIA CHOUBI

April 2011

Chairman: Assoc. Prof. Puziah binti Abdul Latif, Ph.D

Faculty: Environmental Studies

The main goal of this study was to determine the effectiveness of mixed rice husk silica with dried activated sludge as a packing material for physico-biological filter for removal of hydrogen sulphide (H_2S). Removal efficiency (RE), elimination capacity (EC), and pressure drop were used to show the performance of the filters. Three separate filters (physical, biological, and physico-biological filters) were built using PVC cylinder (packed to one litter). Rice husk silica, dried activated sludge and mixture of rice husk silica with dried activated sludge were used in these physical, biological and physico-biological filters, respectively.

In this study, the system was operated under different conditions of two parameters, namely different inlet gas concentration and different inlet gas flow rate. The inlet and outlet concentrations of H_2S were measured using H_2S detector model ppb RAE 3000, USA. In order to determine the characteristics of rice husk silica, the Brunauer-Emmett Teller (BET) specific surface area was performed using a ThermoFinnigan Sorptomatic apparatus and nitrogen adsorption at $-196^\circ C$. Both the shape and pores of rice husk silica were studied using scanning electron microscope (SEM).



The BET method showed that rice husk silica has a very high surface area (226.3 m²/g) with a median pore radius of 2.4 nm and a mesoporous structure. Meanwhile, the chemical composition analysis showed that rice husk silica consisted up to 97.35 % of SiO₂. The physico-biological filter showed more than 99.96 % RE with empty bed residence time (EBRT) of 45 to 90 sec and 300 ppm inlet concentration of H₂S. However, the RE decreased to 96.87 % with the EBRT of 30 s. In the same condition, the biological filter showed 99.37% RE. Nonetheless, the RE was shown to have dropped to 82.09 % with the EBRT of 30 s. The physical filter showed an average RE of 45.83 % with EBRT of 75 to 90 s, and it was saturated after 13 days of operating time with 300 ppm inlet concentration of H₂S. The maximum EC was obtained in the physico-biological filter up to 52.32 gm⁻³h⁻¹, with the RE of 96.87% and H₂S mass loading rate of 54 gm⁻³h⁻¹. The maximum EC in the biological filter was obtained up to 44.33 gm⁻³h⁻¹ with the RE of 82.09% and the H₂S mass loading rate of 54 gm⁻³h⁻¹. In the physical filter, on the contrary, the maximum EC was obtained only up to 11.47 gm⁻³h⁻¹ with the RE of 62.41% and the H₂S mass loading rate of 18.36 gm⁻³h⁻¹. After 53 days of operating time and 54 gm⁻³h⁻¹ of mass loading rates, the maximum pressure drop reached to 3.0 and 8.0 (mm H₂O) for the physico-biological and biological filters, respectively. Nevertheless, the pressure drop did not increase in the physical filter in the same condition. In biological and physico-biological filters, there is a direct and very high relationship between the increase of the H₂S mass loading rate and the amount of pressure drop ($r= 0.98, p < 0.01$) and ($r= 0.96, p < 0.01$), respectively. Based on the findings of this study, mixed rice husk silica and dried activated sludge could be considered as suitable packing material for the physico-biological filter to remove H₂S.

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

**PENYINGKIRAN HIDROGEN SULFIDA OLEH PENURASAN
FIZIKOBIOLOGI DENGAN MENGGUNAKAN BIOFILM
DAN SILIKA SEKAM PADI**

Oleh

SEYED MAHMOUD MEHDINIA CHOUBI

April 2011

Pengerusi: Profesor Madya Puziah binti Abdul Latif, Ph.D

Fakulti : Penganjian Alam Sikitar

Tujuan utama kajian ini adalah untuk menentukan keberkesanan silika sekam padi campur dengan enap cair kering diaktifkan sebagai bahan bungkusan untuk penuras fizikobiologi untuk penyingkiran hidrogen sulfida (H_2S). Kecekapan pembuangan (RE), keupayaan penyingkiran (EC), dan penurunan tekanan digunakan untuk menunjukkan prestasi penuras. Tiga penuras yang berasingan (penuras fizikal, biologi, dan fizikobiologi) yang dibangunkan menggunakan silinder PVC (dipek untuk satu liter). Silika sekam padi, enap cair kering diaktifkan dan campuran silika sekam padi dengan enap cair kering diaktifkan telah digunakan dalam penuras masing-masing; fizikal, biologi dan fizikobiologi.

Dalam kajian ini, sistem ini dikendalikan di bawah keadaan yang berbeza daripada dua parameter, iaitu kepekatan salur masuk gas dan kadar aliran salur masuk gas yang berbeza. Kepekatan salur masuk dan keluar gas H_2S diukur menggunakan pengesan H_2S ppb model RAE 3000, USA. Untuk menentukan ciri-ciri silika sekam padi, Brunauer-Emmett-Teller (BET) luas permukaan khusus dilakukan dengan

menggunakan alat ThermoFinnigan Sorptomatic dan jerapan nitrogen pada -196°C . Kedua-dua bentuk dan leliang silika sekam padi dikaji dengan menggunakan mikroskop elektron imbasan (SEM).

Kaedah BET menunjukkan bahawa silika sekam padi mempunyai luas permukaan yang sangat tinggi ($226.3\text{ m}^2/\text{g}$) dengan radius leliang median 2.4 nm dan satu struktur mesoporus. Sementara itu, analisis komposisi kimia menunjukkan silika sekam padi itu mengandungi sehingga 97.35% SiO_2 . Penuras fizikobiologi menunjukkan lebih daripada 99.96% RE dengan masa mastautin katil kosong (EBRT) dari 45 ke 90 saat dan 300 ppm kepekatan salur masuk gas H_2S . Namun, RE jatuh kepada 96.87% dengan EBRT 30 saat. Dalam keadaan yang sama, penapis biologi menunjukkan 99.37% RE. Walau bagaimanapun, RE ditunjukkan telah menurun ke 82.09% dengan EBRT 30 saat. Penuras fizikal menunjukkan purata RE 45.83% dengan EBRT 75 ke 90 saat, dan ia dipenuhi selepas 13 hari masa operasi dengan 300 ppm kepekatan salur masuk gas H_2S . EC maksimum yang diperolehi pada penuras fizikobiologi mencecah $52.32\text{ gm}^{-3}\text{h}^{-1}$, dengan RE sebanyak 96.87% dan H_2S kadar pemuatan jisim $54\text{ gm}^{-3}\text{h}^{-1}$. EC maksimum yang diperolehi pada penuras biologi mencecah $44.33\text{ gm}^{-3}\text{h}^{-1}$, dengan RE sebanyak 82.09% dan H_2S kadar pemuatan jisim $54\text{ gm}^{-3}\text{h}^{-1}$. Sebaliknya dalam penuras fizikal, EC maksimum diperolehi hanya sampai $11.47\text{ gm}^{-3}\text{h}^{-1}$ dengan RE dari 62.41% dan kadar pemuatan jisim H_2S adalah $18.36\text{ gm}^{-3}\text{h}^{-1}$. Selepas 53 hari masa operasi dan $54\text{ gm}^{-3}\text{h}^{-1}$ kadar pemuatan jisim, maksimum penurunan tekanan mencecah 3.0 dan 8.0 ($\text{mm H}_2\text{O}$) untuk penuras masing-masing; fizikobiologi dan biologi. Namun demikian, penurunan tekanan tidak meningkat dalam penurass fizikal pada keadaan yang sama. Dalam penuras biologi dan fizikobiologi, ada hubungan langsung dan sangat tinggi di

antara peningkatan kadar pemuatan jisim H₂S dan jumlah penurunan tekanan; masing-masing ($r = 0.99, p < 0.01$) dan ($r = 0.95, p < 0.01$). Berdasarkan penemuan kajian ini, silika sekam padi campur dan enap cair kering diaktifkan boleh dianggap sebagai bahan bungkusan sesuai untuk penuras fizikobiologi untuk menyingkirkan H₂S.

ACKNOWLEDGEMENTS

I would like to express my deepest gratitude to:

Allah (S.W.T) who has given me the ability and capacity to complete this dissertation.

Associate Professor Puziah Binti Abdul Latif, my supervisor, for her support and advises who guided and inspired me so much with her comments and advices.

My supervisory committee Associate Professor Ahmad Makmom Bin Abdullah, Associate Professor Latifah Binti Abd Manaf, and Associate Professor Hassan Taghipour who gave me a lot of support, comments and help.

Dean, Deputy Deans and all lectures of Faculty of Environmental Studies that I learnt a lot of things in their classes.

All officials and staffs who helped me throughout my doctoral studies at the Faculty of Environmental Studies.

This research was financially supported by Research University Grant (RUGs) in Universiti Putra Malaysia (UPM) through Project No: 91806.



I certify that an Examination Committee met on 19 April 2011 to conduct the final examination of **Seyed Mahmoud Mehdiinia Choubi** on his **Doctor of Philosophy** thesis entitled **“REMOVAL OF HYDROGEN SULFIDE BY PHYSICO-BIOLOGICAL FILTRATION USING DRIED ACTIVATED SLUDGE AND RICE HUSK SILICA”** 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 student be awarded the Doctor of Philosophy.

Members of the Examination Committee were as follows:

Ahmad Zaharin Aris, PhD

Faculty of Environmental Studies
Universiti Putra Malaysia
(Chairman)

Mohamad Pauzi Zakaria, PhD

Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Internal Examiner)

Shaharin Ibrahim, PhD

Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Internal Examiner)

Peter Brimblecombe

Professor
Faculty of Environmental Sciences
University of East Anglia, Norwich, United Kingdom
(External Examiner)

SHAMSUDDIN SULAIMAN, Ph.D

Professor and Deputy Dean
School of Graduate Studies
Universiti Putra Malaysia



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

Puziah binti Abdul Latif, Ph.D

Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Chairman)

Ahmad Makmom bin Abdullah, Ph.D

Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Member)

Latifah binti Abd Manaf, Ph.D

Associate Professor
Faculty of Environmental Studies
Universiti Putra Malaysia
(Member)

Hassan Taghipour, Ph.D

Associate Professor
Faculty of Health and Nutrition
Tabriz University of Medical Sciences, Tabriz, Iran
(Member)

HASANAHA MOHD GHAZALI, PhD

Professor and Dean
School of Graduate Studies
Universiti Putra Malaysia

Date:



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 institutions.

SEYED MAHMOUD MEHDINIA CHOUBI

Date: 16/May/2011



TABLE OF CONTENTS

	Page
DEDICATION	ii
ABSTRACT	iii
ABSTRAK	v
ACKNOWLEDGEMENTS	viii
APROVAL	ix
DECLARATION	xi
LIST OF TABLES	xvi
LIST OF FIGURES	xviii
LIST OF ABBREVIATIONS	xxiii
 CHAPTER	
1 INTRODUCTION	Page
1.1 General Introduction	1
1.2 Problem Statement	6
1.3 Research Questions	9
1.4 Objectives of the Study	9
1.5 Organization of the Thesis	10
1.6 Significance of the Study	11
1.7 Definition and Description of Terms	12
1.7.1 Empty Bed Residence Time and True Residence Time	12
1.7.2 Removal Efficiency (RE)	14
1.7.3 Elimination Capacity (EC)	14
1.7.4 Mass Loading Rate	16
1.7.5 Volumetric Loading Rate	16
1.8 Expected Results	17
2 LITERATURE REVIEW	
2.1 Introduction	19
2.2 Definition of Air Pollution	19
2.3 Chemical and Physical Properties of Hydrogen Sulphide	21
2.4 Hydrogen Sulphide History	24
2.5 Hydrogen Sulphide Exposure Standards	24
2.6 Hydrogen Sulphide Toxicity	28
2.7 Sources of Hydrogen Sulphide Emission	30
2.7.1 Natural Sources	30
2.7.2 Sources Associated with Human Activities	33
2.8 Exposure to Hydrogen Sulphide	34
2.9 Sulphur and Hydrogen Sulphide Cycles	36
2.10 Biological Process Leading to Biogenic Sulphur Fluxes to the Atmosphere	37
2.11 Effects of Hydrogen Sulphide	40
2.11.1 Effects of Hydrogen Sulphide on Animals and Human	40



2.11.2	Hydrogen Sulphide and Acid Rain	43
2.12	Air Pollution Control Technology	44
2.13	Conventional Methods for Air Pollution Control	46
2.13.1	Condensation	48
2.13.2	Incineration	48
2.13.3	Adsorption	49
2.13.4	Absorption	50
2.13.5	Membrane Systems	51
2.13.6	Biofiltration	51
2.14	Degradation Mechanism of Hydrogen Sulphide	59
2.15	Limitation of Biofiltration System for Treating of Air Pollution	62
2.16	Biological Packing Material (Sewage Treatment Plant Sludge)	63
2.17	Rice Husk	64
2.18	Previous Relevant Research in Literature	69
2.18.1	Previous Relevant Research on Packing Materials	72
2.18.2	Previous Relevant Research on Rice Husk	75
2.18.3	Previous Relevant Research on Microorganisms Adaptation Period	77
2.18.4	Previous Relevant Research on the Performance of Biofilter	78
3	RESEARCH METHODOLOGY	
3.1	Research Approach	79
3.2	Steps of the Thesis	80
3.2.1	Step One (Plan Construction)	81
3.2.2	Step Two (Set-up the System in the Laboratory)	87
3.2.3	Step Three (Data Collection)	87
3.2.4	Step Four (Data Analysis)	89
3.3	Selection of the Methods and Sampling	89
3.3.1	Sampling	93
3.3.2	Measurement Methods	91
3.4	Cultivation and Preparation of the Media for Microorganisms	94
3.4.1	Preparation of Thiosulfate Oxidizing and Sulphur Media	96
3.4.2	Cultivation of Microorganisms	96
3.5	Filtration System	97
3.6	Devices and Materials Used in This Research	101
3.7	The Measured Parameters in This Study	102
3.8	Checking the Accuracy and Precision of the Requirements	102
3.9	Statistical Procedures	103
3.10	The Exploratory Data Analysis (EDA)	106
4	RESULTS AND DISCUSSION	
4.1	The Characterization of Rice Husk Silica	107
4.2	Identification and quantifications of Organisms in the Packing Materials	111
4.3	The Performance of the Filters	113
4.3.1	The Removal Efficiency	114

4.3.2	The Elimination Capacity	124
4.3.3	The Changes in pH	131
4.3.4	The Pressure Drop	138
4.3.5	The Operation of the Physical Filter Using Regenerated Rice Husk Silica (RRHS)	145
4.4	The Relationship between Mass Loading Rate and Pressure Drop	150
4.5	The Relationship between Mass Loading Rate and Elimination Capacity	155
4.6	Limitation of the Research	160
5	SUMMARY, GENERAL CONCLUSION AND RECOMMENDATIONS FOR FUTURE RESEARCH	
5.1	Summary of the Findings	162
5.1.1	The Characterization of Rice Husk Silica	162
5.1.2	Removal Efficiency of the Filters	163
5.1.3	Elimination Capacity of the Filters	163
5.1.4	Pressure Drop in the Filters	164
5.1.5	pH Changes in the Filters	164
5.2	The Relationship between Different Flow Rates and Pressure Drop	165
5.3	The Relationship between Different Flow Rates and Elimination Capacity	165
5.4	The Ability of the Regenerated Rice Husk Silica (RRHS) in Removing H ₂ S	166
5.5	Final Conclusion	167
5.6	Recommendations for Future Studies	168
	REFERENCES	170
	APPENDICES	179
	BIODATA OF STUDENT	191
	LIST OF PUBLICATIONS	192