UNIVERSITI TEKNOLOGI MARA

SUSTAINABLE TWO-STAGE THERMOCHEMICAL CONVERSION OF SEWAGE SLUDGE FOR THE CO-PRODUCTION OF BIOCHAR AND CARBON NANOTUBES

MOHD SYAZWAN BIN MOHD GHAZALI

Thesis submitted in fulfilment
of the requirements for the degree of
Doctor of Philosophy
(Chemical Engineering)

Faculty of Chemical Engineering

October 2025

ABSTRACT

The increasing volume of sewage sludge, along with its significant environmental burden and disposal challenges, necessitate sustainable waste management solutions. This study investigates the potential of co-producing biochar and carbon nanotubes (CNTs) from sewage sludge through a two-stage process integrating pyrolysis and catalytic chemical vapor deposition (CCVD), presenting an innovative sustainable approach to waste valorization. In the first stage, sewage sludge underwent slow pyrolysis at 550°C, yielding 66 wt.% biochar. The produced biochar exhibited a BET surface area of 24.40 m^2/g and elemental composition of C (20.9%), H (0.4%), N (2.4%), S (1.7%) and O (3.3%). The organic vapor generated from sewage sludge pyrolysis was then introduced into a secondary reactor for CCVD, where a cobalt catalyst facilitated the formation of CNTs. Process parameters, including temperature (650 to 950°C) and catalyst loading (0.2 to 0.8 g), were optimized using response surface methodology (RSM) coupled with a central composite design (CCD) model. The optimal conditions for CNTs production were determined to be a temperature of 800°C and a catalyst load of 0.5 g. Characterization techniques such as Raman spectroscopy, X-ray diffraction (XRD), field emission scanning electron microscopy (FESEM), and transmission electron microscopy (TEM) were employed to analyze the CNTs. The findings from this novel two-stage thermal-catalytic process demonstrated a sustainable waste valorization approach to converting sewage sludge into materials with enhanced properties. The biochar exhibited desirable physicochemical characteristics suitable for various applications, while the CNTs displayed multiwall structures with inner diameters of approximately 3.2 nm and outer diameters ranging from 20 to 40 nm. The obtained CNTs exhibited a high degree of disorder and low crystallinity, as evidenced by Raman I_D/I_G ratio of 0.992. The maximum yield of CNTs production reached 30 wt. %. The study revealed that synthesis temperature significantly influenced both the yield and structural properties of the CNTs. Under optimal conditions (800°C, and 0.5 g catalyst loading), the predicted yield of 30.53% almost matches the observed experimental maximum yield. This research highlights sewage sludge as a sustainable feedstock for biochar and CNTs, with biochar applicable in soil conditioning and remediation, and CNTs in nanocomposites, energy storage, and pollutant removal. The outcomes indicate a strong foundation for future studies and applications, promoting circular economy principles and contributing to environmental sustainability.

ACKNOWLEDGEMENT

First and foremost, I express my deepest gratitude to the Almighty ALLAH s.w.t for granting me the opportunity to pursue and successfully complete my PhD journey.

I extend my heartfelt appreciation to my esteemed supervisor, Professor Ir Ts Dr Syed Shatir Asghrar Syed Hassan for his exceptional guidance, unwavering support, and profound insights that have shaped my research and academic growth. I am indebted to my late co-supervisor, Associate Professor Ir Dr Ahmad Rafizan Mohamad Daud, for his invaluable guidance and support. May Allah grant him eternal peace and a place in Jannah. Also, to my other co-supervisor Professor Ts Dr Norazah Abd Rahman, for her support and mentorship throughout this academic endeavor.

I would like to extend my sincere thanks to my dedicated labmate, Ir. Mohd Saufi Md Zaini, for his collaboration and assistance in the laboratory, which significantly contributed to the success of this research project. I would also like to thank Encik Mohibah Musa, Encik Mustafa Mokhtar, Puan Siti Zaharah Roslan and Puan Nur Faradilla Anuar for their support and camaraderie during this academic journey.

I dedicate this thesis to the loving memory of my late father, Mohd Ghazali Setapa, and mother, , for their vision, dedication, and unwavering belief in the importance of education. I also dedicate this thesis to my siblings, my wife (Nik Yuslina Nik Yusoff), and my three lovely sons (Muhammad Syafi Rifqi, Muhammad Syabil Nazih, and Muhammad Syatir Asghrar Yusoff—named in honor of my supervisor). Their sacrifices, support, and encouragement throughout this work have been invaluable, and for that, I am eternally grateful.

TABLE OF CONTENTS

CHAPTER 1
INTRODUCTION

1.1 Introduction

Sewage sludge is an inevitable by-product of municipal wastewater treatment plants. Its generation has surged rapidly due to urbanization, population growth, and economic development, becoming a significant socio-environmental issue that demands immediate attention. According to Jellali *et al.* (2021), approximately 45 million dry tonnes of sewage sludge are generated annually [1]. This raises significant concerns, including the potential for nutrient leaching, adverse impacts on soil biodiversity, and the release of greenhouse gases such as CH_4 and N_2O [2]. Exposure to sewage sludge can lead to various health risks, including the spread of pathogens, toxic chemical contaminations, and the potential for accumulating heavy metals [4], [5].

Conventional methods of sewage sludge management, such as incineration, landfilling, and land application, are increasingly unviable due to stringent regulations, land scarcity, and other issues associated with environmental impact, resource utilization, and energy efficiency [5]. The improper management and disposal of sewage sludge can degrade soil quality, pollute water sources, and harm overall ecosystem health [6], [7]. Addressing this issue requires implementing effective management strategies that align with the principles of sustainable development [8]. Therefore, the motivation for this PhD study lies in the promising potential of adopting waste-to-value approaches to tackle this challenge, transforming sewage sludge from an environmental burden into a valuable resource, thus adhering to circular economy principles and the United Nation's Sustainable Development Goals (SDGs).

1.2 Background of Study

Policymakers and scientists worldwide agree that sewage sludge management practices should shift from merely treating waste to recovering embedded energy and valuable chemical assets. According to Peccia and Westerhoff (2015), this shift *"must be guided by the application of green engineering principles to ensure economic, social, and environmental sustainability"*. Researchers are actively exploring innovative