UNIVERSITI TEKNOLOGI MARA

CHARACTERIZATION AND DISINTEGRATION PERFORMANCE OF THERMOPLASTIC STARCH-BASED FILM INCORPORATED WITH ALOE VERA GEL AND POLYETHYLENE

SITI FATMA ABD KARIM

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

College of Engineering

November 2023

ABSTRACT

Thirty years ago, researchers blended thermoplastic starch (TPS) and polyethylene (PE) to enhance the tensile strength of TPS and improve PE's disintegration ability to reduce plastic waste accumulation. Despite toxic compatibilizer/crosslinker addition to improve TPS/PE compatibility, TPS/PE compatibility was uncertain due to inconsistent tensile strength. Plus, the PE ratio in the TPS/PE blend was high, contributing to insignificant disintegration performance. Therefore, the incorporation of aloe vera (AV) to replace toxic compatibilizer while acting as a crosslinker into TPS to reduce the amount of PE used in TPS/PE film was investigated. The effects of AV and PE on TPS characteristics and disintegration performance were evaluated. Melt-blending and hot-press techniques were applied to produce TPS + AV + PE film, while the ISO 20200 technique was implemented to assess the disintegration performance. The formulation yielding the best tensile strength was TPS + 30% AV + 10% PE, with an average of 9.64 MPa due to the crosslinking effect that occurred between TPS and 30% AV gel and grafting formation between TPS + 30 % AV with 10% PE. The increment in melting and decomposition temperatures of TPS + 30% AV film verified the crosslinking performance between TPS and 30% AV. At the same time, the grafting of 10% PE on TPS + 30% AV film is supported by a decrement in water solubility percentage. The film's thickness, visual appearance, functional group changes, and crystallinity were also assessed. The TPS + 30% AV + 10% PE was considered biodegradable since 99% of the film disintegrated in the synthetic solid waste based on ISO 20200. The Hill model was chosen as the most suitable kinetics model to represent the disintegration behavior of TPS + 30% AV + 10% PE with the highest R^2, lowest RMSE, and accuracy factor (AF) greater than one and closer to 1. Three phases of the disintegration process were proposed: fragmentation, hydrolysis, bioassimilation, and mineralization. The increment in weight loss percentage, void formation, and TPS functional compound disappearance suggested the proposed disintegration process. The increment in water contact angle percentage validated the disintegration mechanism. In conclusion, this study revealed the benefits of 30% AV gel and 10% PE in enhancing the TPS film. The disintegration ability of TPS + 30% AV + 10% PE within 90 days based on ISO 20200 suggests its high potential to be used as a commercialized biodegradable film packaging with a suitable application.

ACKNOWLEDGEMENT

Alhamdulillah, Thank you, Allah!

An outstanding and supportive family is the main factor contributing to this beautiful thesis. My husband, Mohd Husaini bin Saidi, and his endless support cannot be imagined with his wonderful smile and words. I am so sorry for the kids, Siti Sumayyah and Ahmad Yasir, because most of my time was spent on this thesis instead of playing and spending time with you two. You both are the fabulous and most excellent gifts from Allah. I also want to thank my entire family, especially my parents and my parents-in-law, who have supported me spiritually.

I sincerely thank my supervisor, Honorary Prof Dr Ku Halim Ku Hamid, and co-supervisor, Assc. Prof Dr Junaidah Jai, for the continuous support of my PhD study and research. Their trust, patience, motivation, enthusiasm, and immense knowledge supported me. The guidance helped me during the research and writing of this thesis. Not to mention Dr Haji Khairol Nizam Zainan Nazri, my PhD mentor who offered invaluable advice and encouragement as I worked to maintain a healthy work–life balance and write my dissertation in its final year. Splendid appreciation is given to Assc. Prof Dr Juferi Idris for his knowledge and close guidance in helping me with high-impact paper publication.

I am immensely thankful to Skim Latihan Akademik Bumiputera / Skim Latihan Akademik IPTA, Ministry of Higher Education, UiTM Shah Alam, and Internal grant UiTM Shah Alam, *Geran Penyelidikan Khas*, (RMI file no: 600-RMC/GPK 5/3 (146/2020)) for their support in completing this study. I sincerely thank all the volunteer experts, colleagues, and assistant engineers for contributing to this study, Dr Sufian So'aib, Dr Sharmeela Mathali, Dr Sakinah Mohd. Alauddin, Dr Nur Azrini Ramli, Dr Muhammad Syafiq Hazwan Ruslan, Dr Nurul Fadhilah binti Kamalul Aripin, Assc. Prof Dr Kamariah Noor Ismail, Assc. Prof Dr Siti Shawalliah, Ms Sulastri Manap, Mrs Nurul Iman Ahmad Bukhari, Mr Mohd Idris Desah and many more who were involved directly or indirectly. Their presence was not just a coincidence but a gift from Allah SWT throughout my PhD journey. Without their precious input, this study would not have been possible. Many thanks also to the professional proofreader team.

Former students, I am deeply grateful for all your support and assistance from 2019 through 2022 as I conducted experiments. It was with them that I did most of my laboratory work. It was a frustrating cycle of wrong formulation, challenging sample process, unlucky equipment breakdown, weekend lab time, knowledge sharing, and constant momentum and support from them. They always believed in me. Abdul Wafi Bin Abd Jalil, Fatini Norsyahida Binti Norhisam, Fatin Nor Amira Binti Abd Rahaman, Wan Nur Afina Binti Wan Fauzi, and Siti Khadijah Binti Mohd Radzi are amongst them.

Finally, the constant silent support that I embrace each semester is given by the WIP members, Puan Nadia Kamaruddin, Puan Rabiatul Adawiyah, Dr Jefri Jaafar and Mr Mohibah Musa.

TABLE OF CONTENTS

CHAPTER ONE
INTRODUCTION

1.1 Research background

Malaysia had a severe challenge when it was rated seventh in 2010 in plastic trash production, and the value kept increasing over time (H. L. Chen et al., 2021; Jambeck et al., 2015). Worsening the condition, since 2017, Malaysia has been the world's largest importer of plastic waste. Yet, in 2018, Malaysia produced more than 0.94 million tonnes of mismanaged plastic waste per year (*Malaysia's Roadmap towards Zero Single-Use Plastics 2018 - 2030*, 2018). Food producers are among the most notable users of plastic as a principal packaging material. The increased use of plastic results from human habits, as purchasing packaged meals is preferable (Jaafar et al., 2018; Majid et al., 2018). Due to this worldwide challenge, the demand for green goods and ecologically sustainable techniques has surged. Industries seek biodegradable plastics to replace conventional plastics (Ministry of Environment and Water (KASA), 2021). Plus, the depletion of fossil fuels, which are non-renewable resources, and the difficulty of decomposing have added to this concern (Majid et al., 2018; Yun et al., 2018). The demand for bio-based food packaging is primarily due to worries about food safety and increased knowledge about environmental issues, most notably the inability of plastic to disintegrate even after a long period (Domínguez et al., 2018). Numerous studies have been conducted to develop biodegradable, renewable, and edible food packaging (Piñeros-Hernandez et al., 2017). Unfortunately, synthetic-based polymer packaging accounts for more than one-third of worldwide sales. Degradable plastics are still far behind the production of synthetic-based polymer packaging (Datta & Halder, 2018; Layla & Rothenberg, 2021).

Conventional plastics for food packaging applications are made from polyethylene (PE). PE is derived from petroleum and is known as a synthetic polymer used in most packaging material applications because of its excellent commercial success (Makhtar et al., 2013). High-density PE (HDPE), linear low-density PE (LLDPE), and low-density PE (LDPE) are the three common PE types with different chemical structures. PE is used to develop films to make plastic bags, wraps, and mulch films (Khanoonkon et al., 2016). PE is very stable and works well for packaging films