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Article / Book Information

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Title(English)	Effective Bioprocessing for Production of Polyhydroxyalkanoates with Tunable Properties
著者(和文)	MFADZIL FAKHRUL
Author(English)	Mohd Fadzil Fakhrul Ikhma Bin
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論 文 要 旨

THESIS SUMMARY

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学生氏名： Student's Name	Fakhrul Ikhma Bin Mohd Fadzil	指導教員（主）： Academic Supervisor(main)	柘植　丈治
		指導教員（副）： Academic Supervisor(sub)	

要旨（英文 800 語程度）
Thesis Summary（approx.800 English Words ）

Polyhydroxyalkanoates (PHAs) are biopolyesters synthesized by various bacteria as an energy storage material under stress and nutrient limiting conditions. PHAs are seen as promising new materials for replacing the currently available plastics derived from fossil source due to their ability to degrade naturally in the environment without liberating toxic compounds. There is great interest in developing PHAs as high-performance materials for wide range application. PHAs containing medium-chain-length (MCL) monomers has been extensively studied for improving the current limitations of PHAs, mainly associated with their production strategies and thermo-mechanical properties.

One of MCL-PHA homopolymers, known as poly(3-hydroxydecanoate) [P(3HD)] is highly attractive due to its flexibility and high in transparencies. However, it is still difficult to produce P(3HD) in high amount from fatty acid due to the toxicity of carbon substrate, especially at high concentration. In Chapter 2, methods to solve problems associated with a low yield of MCL-PHA homopolymer production were proposed and its effectiveness was validated. To achieve this aim, carbon feeding by intermittent fed-batch feeding was applied. Maintaining co-carbon source at low concentration is a key for ensuring steady cell growth and hence, an effective bioprocess strategy was elucidated to produce this highly interest PHA in the large amount. By feeding glucose at 1 g/L and sodium decanoate at 0.5 g/L for ten times, 2.91 g/L P(3HD) was acquired. Considering the high conversion yield, further increased in fatty acid feeding concentration at 0.75 g/L was applied. However, P(3HD) production was greatly reduced. Subsequently, optimization of culture medium with yeast extract enrichment had significantly increased the P(3HD) titer to 5.44 g/L, the highest ever reported so far. Yeast extract added to the medium was able to provide extra nutrients and promoted cell growth, thus improved the P(3HD) yield in glucose cultures. Furthermore, the use of other renewable carbon sources illustrated the practicable approach of PHAs production strategies in a sustainable and effective way. However, an up-scale study of P(3HD) production was still incapable to produce high yield due to the implementation of two-stage cultivation.

Chapter 3 demonstrated effective bioprocessing of poly(3-hydroxybutyrate-*co*-3-hydroxyhexanoate) [P(3HB-*co*-3HHx)] by using a by-product of palm kernel oil processing, palm kernel oil fatty acid distillate (PKFAD), as a substrate. Utilization of renewable resources from agricultural industries ensures the renewability and sustainability of the whole PHA life cycle. An antibiotic-free culture was introduced in recombinant *Escherichia coli* strain based upon glycine auxotrophic complementation. PKFAD fed as the sole of carbon source by fed-batch produced 9.2 g/L P(3HB-*co*-3HHx) accounted for 30 wt.%. Maintaining fatty acid feeding at 3 g/L per pulse also enabled to prevent toxicity and avoid cell growth inhibition. The acquired random copolymer contained 28 mol% 3HHx and weight-average

molecular weight (M_w) of 2.1×10^5. The proposed strategy for reliable and economical bioprocessing with high robustness system was proved by the efficient plasmid maintenance, reached to 100%.

Commercially available P(3HB-*co*-3HHx)s are having low in 3HHx content. Considering greater applications, new materials with tunable property can be obtained by blending P(3HB-*co*-3HHx)s with different 3HHx fractions. P(3HB-*co*-3HHx)s behave differently as the 3HHx content varies. P(3HB-*co*-3HHx)s with low 3HHx fractions generally hard, stiff and brittle, while the high in 3HHx fractions are more soft and flexible. In Chapter 4, preparation of tunable PHA materials by blending the soft and hard type P(3HB-*co*-3HHx)s for controlling the thermal property was reported. It was found that the miscibility of P(3HB-*co*-3HHx)s is mainly affected by the 3HHx content up to 49 mol%, while further increase resulted in immiscible blends. Additionally, the molecular weight of the sample used in the blend study had significantly affected their miscibility.