INTERNATIONAL RESEARCH JOURNAL OF PHARMACY
www.irjponline.com
ISSN 2230 – 8407

Research Article

# USE OF BIOBLEND POLYSTYRENE-POLY (3-HYDROXYBUTYRATE) AS THE COATING MATERIAL OF THE NPK SLOW RELEASE FERTILIZER

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Article Received on: 08/09/18 Approved for publication: 08/10/18

**DOI: 10.7897/2230-8407.0910229**

## ABSTRACT

Conventional Nitrogen-Phosphorus-Potassium (NPK) fertilizer granules is absorbed by plants inefficiently. A certain proportion of the fertilizer is released into the environment. The aim of this research was to formulate NPK slow release fertilizer granules using polystyrene - poly 3-hydroxybutyrate polymers. NPK fertilizer granules were coated by spray coating method using polystyrene-polyhydroxybutyrate bioblend. NPK slow release granules obtained were evaluated in term of SEM and FTIR analysis; percentage of coating and released of $P_2O_5$; dissolution efficiency and rate of release of $P_2O_5$; and the order of release kinetics. FTIR analysis of slow release NPK fertilizer showed no chemical interaction between NPK fertilizer granules and coating polymer materials. The percentage of release of $P_2O_5$ from uncoated and coated NPK granules after 48 hours were 94.17 ± 7.88, and 46.45 ± 0.77%, respectively ($p < 0.05$). The rate of release of $P_2O_5$ from uncoated and coated NPK fertilizer granules, was 1.90 ± 0.06, and 0.98 ± 0.01 $\%.h^{-1}$ respectively, ($p < 0.05$). Kinetics release of slow release NPK fertilizer followed the Langenbucher kinetics (R = 0.993). The release efficiency of $P_2O_5$ from uncoated and coated NPK fertilizer granules were 65.67 and 33.66% ($p < 0.05$), respectively. It can be concluded that coating of NPK fertilizer granule using poly-hydroxy butyrate could influence the rate and release efficiency of $P_2O_5$ ($p < 0.05$).

**Keywords:** polystyrene, poly(3-hydroxybutyrate), granules, NPK, slow release

## INTRODUCTION

Fertilizer is an essential material to support crop production. Ben *et al.* (2018) reported that the climatic conditions and conventional using method cause 90% of the fertilizer used does not reach the target[1]. Rain and irrigation water rinse fertilizer significantly and can cause inefficient in using it by the plant[2,3]. Economic losses and environmental pollution occur because of inefficient used of conventional fertilizers. It can be anticipated by using slow release fertilizer (SRF). SRF is designed to obtained gradually released of fertilizer into the soil[4]. Addition of the coating polymer can prolong the release of the fertilizer. SRF maintains an effective concentration of fertilizer in the soil and available for plants. Using the SRF will reduce the lots of fertilizer due to water-washing of fertilizers, and will decrease fertilizer needed[5,6,7]. It will also minimize environmental pollution[4].

The use of conventional coating polymers will result in high material costs and high synthetic polymer residues on the ground. Use of biodegradable polymers can reduce polymer residues in the soil[8]. Utilization of SRF with conventional type will be able to improve the fertilizer efficiency, reduce the rinsing of fertilizer by rain or irrigation water, provide longer fertilizer release for longer time[9].

Polystyrene is a synthetic low-cost polymer usually used as a protective material in the wrapping of electronic equipments. It is unbiodegradable itself. It could be degraded by being changed into polystyrene bioblend. Bioblend is a mixture of a biodegradable polymer with a nonbiodegradable polymer[10]. In this study, bioblend polymers was prepared by mixing polystyrene, PS, with poly(3-hydroxybutyrate), P(3HB). P(3HB) is a biopolymer produced by bacteria eg. *Ralstoniaeuthopha* and *Erwina sp* USMI-20[11,12,13] . This biopolymer has biodegradable and non toxic characteristics[14]. Over the past few years, several studies have been performed using combination of fertilizer with polymers to obtain efficient fertilizer products. The mechanism of process is based on the slow-release pharmaceutical drug delivery technology to be applied in agriculture[15,16]. The aim of this research was to obtain a slow release NPK fertilizer using polystyrene-P(3HB)bioblend as the coating material.

## MATERIALS AND METHOD

### Material and Tools

Materials used in this research was NPK fertilizer granules, polystyrene, poly (3-hydroxy- butyrate), chloroform, paraffinum liquidum, Span 80, sulfuric acid, ammonium molybdate, potassium antimonyl tartrate, ascorbic acid, distilled water.Tools used in this research wereScanning Electron Microscope, FTIR Spectrophotometer, UV-Vis spectrophotometer, compressor, spray gun, hair dryer, rotary drum coater, measuring pipette, analytical balance, hot plate, and other glasses apparatus usual used in laboratory.

### Coating Material Formulations

1.5 g of polystyrene was mixed with 0.5 g P(3HB) and, dissolved in the mixture of 50 mL chloroform and 1 mL paraffin liquidum. The solution was stirred using a magnetic stirrer at 380 rpm for 10 minutes. The coating process was performed by spray-coating method. Uncoated NPK granules was sprayed with the coating solution. 25 g of NPK granules were placed into the container of the spray gun coating pan machine. The NPK granules was sprayed into the coating pan with coating solution at an adjusted rotating speed, and temperature at 70 °C. The coated NPK granules was dried using oven at 70 ºC for 1 hour to ensure the solvent vaporized and completely dried. The dried coated granules obtained was weighed.

### Surface morphology characterization of coated granules by SEM

The surface of NPK granule and also cross section of SEM micrograph of NPK with PS-P(3HB) coating polymers at magnification of 300x was observe using JEOL-JSM-6360 SEM Accel at voltage of 15 kV.

### Interaction of Coating Materials

Interaction between components of coating materials and/or NPK granules was observed using Universal ATR Perkin Elmer FTIR Spectroscopy.

### Preparation of Standard Curve of Phosphorus

0.143 g of $KH_2PO_4$ was dissolved in distilled water for total volume of 100 mL in a 100 mL measuring flask to obtain at concentration equal to 1 mg/mL of $P_2O_5$ main standard solution. A series of standard solution of phosphorus was prepared by dilution of the main standard solution to obtain concentrations of 1, 2, 3, 4, 5, 6, and 7 µg/mL. The absorbance of solutions were measured using a UV-Vis spectrophotometer at maximum wave length of 716.5 nm .

### Evaluation of the release of phosphorus from coated PS-P(3-HB) NPK granules

Evaluation of the release of NPK from the coated granules was performed at room temperature using our own developed method. The release test apparatus contains of a glass cup with specificacy shown in Figure 1 below. Diameter and height of cup were 7, and 10 cm, respectively. The cup was covered with a sponge with diameter and thickness of 6.8, and 2.5 cm, respectively. The cup was filled with 100 mL distilled water. A certain amount of coated NPK granules equal to 1 gram of NPK granules was placed into the cup glass. The apparatus was kept without any treatment. Fertilizers were allowed to release into medium solution. Five mL of sample solutions were withdrawn at 0.083; 0.17; 0.25; 0.5; 1; 4; 12; 24; 48 hours. Then the medium in the container was replaced with the fresh distilled water with the same volume and temperature up to 100 mL. The concentration of phosphorus in sample solutions were analyzed using visible spectrophotometer at wavelength of 716.5 nm.

### Percentage of coating

The percentage of coating was calculated by the following equation:

$$\% \text{ Coating} = [\text{M polymer residue (g)} / (\text{total M (g)}] \times 100$$

where:
Percentage of coating is percentage of polymer coated on NPK fertilizer granules. M polymer is mass of polymer residue. M total is total mass (NPK + polymer)[17].

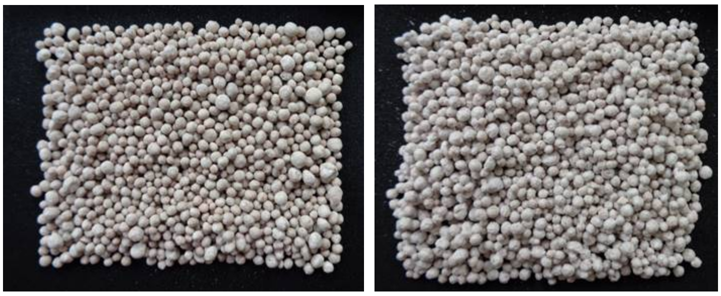
A B

**Figure 1: The profile ofuncoated NPK granules(A), and PS-P(3HB) bioblend coated NPK granules (B)**

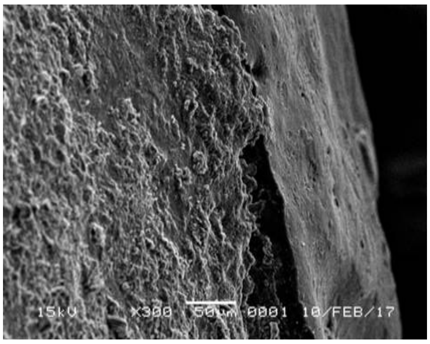

**Figure 2: SEM micrograph image of cross-section NPK PS-P(3HB) bioblend coated granules at magnification 300 times**

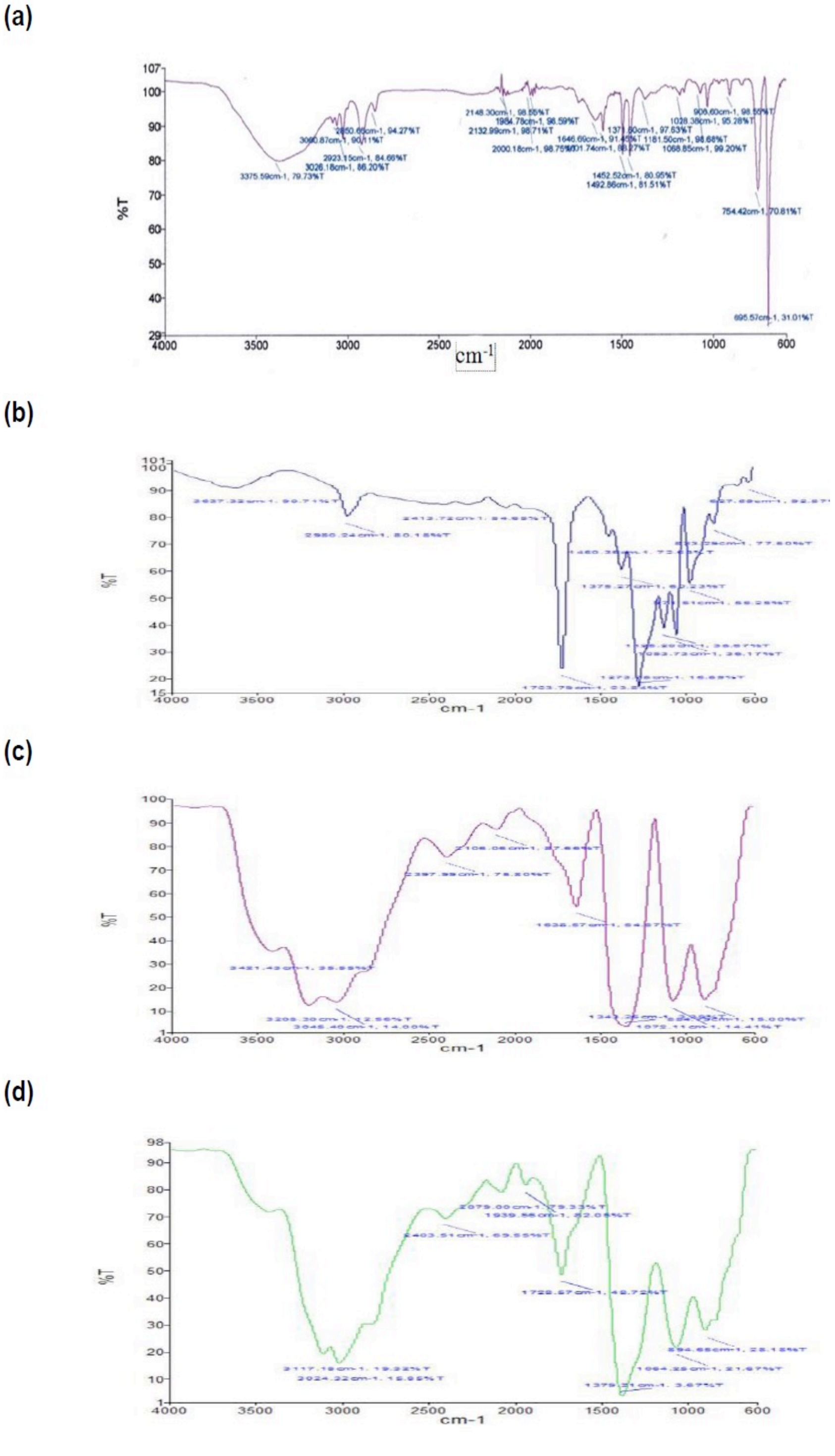

**Figure 3: FTIR spectrum of (a) PS, (b) P(3HB), (c) uncoated NPK granules, (d) PS-P(3HB) coated NPK granules**

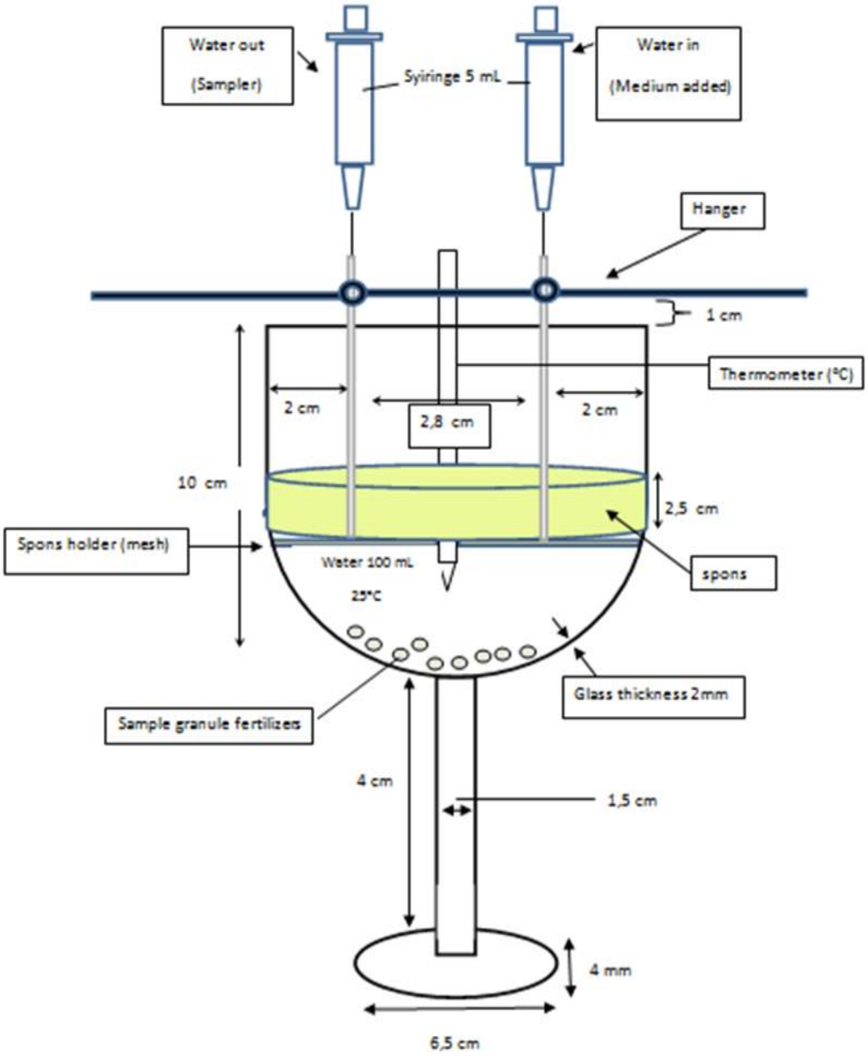

**Figure 4: Profile of the release test apparatus used in this study**

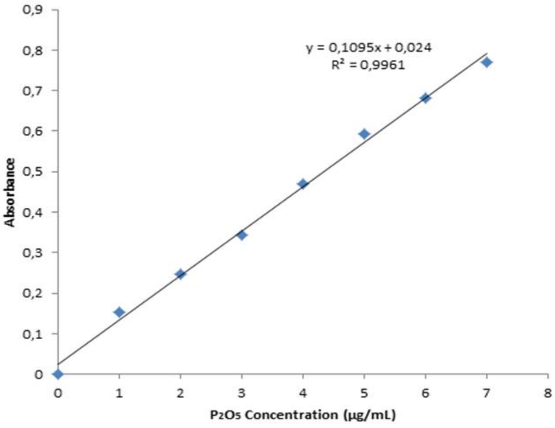

**Figure 5: Standard curve of $P_2O_5$**

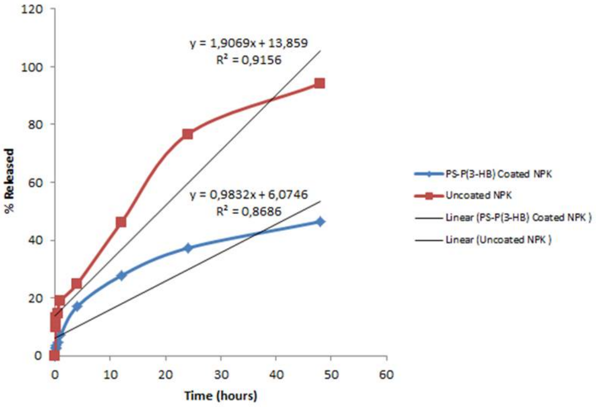

**Figure 6: The profile of $P_2O_5$ released from coated and uncoated NPK granules**

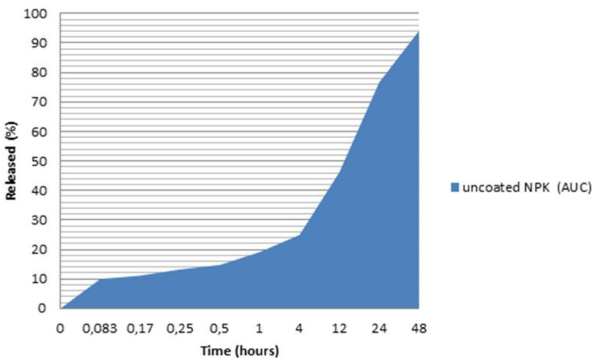

**Figure 7: The profile of release efficiency of uncoated NPK granules**

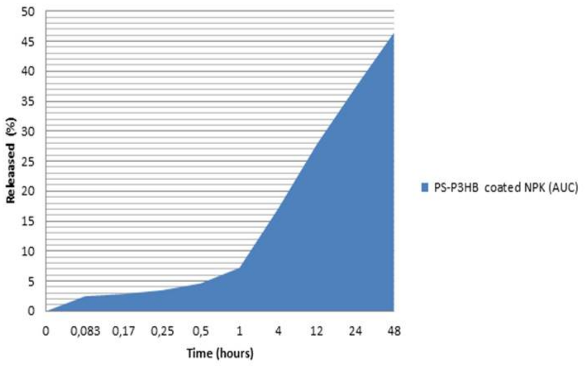

**Figure 8: The profile of $P_2O_5$ released from coated and uncoated NPK granules**

## RESULTS AND DISCUSSION

Figure 1 shows the appearance of uncoated (A) and coated (B) NPK granules using the mixture of PS and P(3HB). Physically uncoated NPK fertilizer is round and yellowish brown in color and quite smooth surface, while for coated granules were rounded, rough, porous and light brown in color. Physically it also appears that the coated fertilizer has a larger granule size than the uncoated NPK granules.

Figure 2 is cross section of SEM micrograph of NPK with PS-P(3-HB) coating polymers at magnification of 300x using JEOL-JSM-6360 SEM Accel at voltage of 15 kV.

SEM micrograph shown that the coating layer and NPK granules was could be noticed clearly. Thickness of the coating layer at 300x magnification was about 47.0 µm. The thickness on each side of the NPK granules surface was slightly different. It was due to the rough coated NPK granules surface structure so that the thickness of the coating layer generated on each side is varied not significantly.

The identification of a substance can be performed using FTIR spectrometer. The spectra of the uncoated and coated NPK granules using PS and P(3HB) were compared with those of PS, P(3HB), and NPK granules. FTIR spectra of NPK granules showed that it has several wave numbers. Wave numbers of 3241, 3208, and 3045 $cm^{-1}$ showed the presence of N-H, OH, and CH groups, respectively. PS FTIR spectrum of PS in Figure 3(a), showed the presence of =C-H, C-H cyclic aromatic, and C-H alkyl groups at wave number of 3375, 3060-3020, and 2923-2850 $cm^{-1}$, respectively. It also has C=C aromatic cyclic, and monosubstitution benzene at wave number of 1646, and 754-695 $cm^{-1}$, respectively. While, P(3HB) FTIR spectrum in Figure 3(b), showed the presence of C=O, C-H, C-N, C-O, C-O, and C-H groups at wave number of 1723, 1357, 1273, 1126, 1053, and 971 $cm^{-1}$, respectively.

FTIR spectra of PS-P(3HB) NPK coated granules has an almost similar profile. When its were compared to the IR spectra of raw materials, the formula has some of the same spectrum with spectrum on PS and P(3-HB). FTIR spectra of PS-P(3HB) coated NPK granules is in wave numbers between 3117, 3024, 2403, 1728, 1379, 1064, and 894 $cm^{-1}$ indicated the group O-H, C-H,O-H, C=O, C-H, C-O, and C-H respectively.

Figure 3(c) shows FTIR spectrum of coated NPK similar to the spectra in each coating (PS and P(3HB)) and NPK without coating. In figure 4(d) it was clear that no chemical reaction in NPK coated granules, so it can be concluded that no new substance formed in NPK coating process.

Figure 3(d) also indicated that the FTIR spectrum of original NPK granules, PS and P(3HB) similar with PS-P(3HB) coated NPK granules. It means no chemical interaction occurs in NPK coating process. The addition of the PS-P(3HB) coating polymer does not produce a new substance[18,19].

### Evaluation of release of NPK granules

The release of fertilizer from NPK granules were observed by determination of phosphorus as $P_2O_5$ released in aqueous media using equipment as shown in Figure 4.

Determination of phosphorus levels by spectrophotometric method is based on the formation of complex compounds of phosphomolibdate to produce a stable blue color within 15 minutes. The maximum absorbance was at visible wave length of 716.5 nm. The linear regression of standard curve obtained was $y = 0.1095x + 0.024$ ($R = 0.996$) (Figure 5). In an acidic environment the orthophosphate compound will react with ammonium molybdate to form the ammonium phosphomolybdate complex.

The percentage of $P_2O_5$ released from PS-P(3HB) bioblend coated NPK granules compared to uncoated NPK granules within 48 were 94.17 ± 7.88, and 46.45 ± 0.77% (Figure 6),respectively ($p < 0.05$). Its mean that coated fertilizers could postponed the release of $P_2O_5$.

The release efficiency of $P_2O_5$ from uncoated (Figure 7) and coated (Figure 8) NPK fertilizergranules were 65.67 and 33.66%, respectively. It was indicated that fewer NPK released from the PS-P(3HB) coated NPK granules compared to uncoated NPK granules ($p < 0.05$).

Kinetic profile of uncoated NPK 99.9% of fertilizer released from uncoated NPK granules occured after 58,13 hours. While, the kinetic profile of the PS-P(3HB) coated NPK granules showed the Langenbucher kinetics. Coated NPK granules released by 99.9% in 3647.5 hours, or about 5.06 month.
The release rate of $P_2O_5$ from uncoated and coated NPK granules were 1.90 ± 0.06 and 0.98 ± 0.01 $\%.h^{-1}$, respectively($p < 0.05$). The slow release fertilizer is an alternative to minimize the loss of fertilizer. It can be obtained by using a coating.

To observe the layer of polymer in coated granules, the cross section of the coated NPK granules were observed using a SEM.

There were two layers, the core and outer layer. The thickness of the coating layer was 47.0 µm approximately (at magnification of 300x). The coating layer of the polymer on the outside of the NPK granules NPK will retard the release of fertilizers. The polymer coating serves as a physical barrier for transferring of fertilizers, slowering the diffusion rate of water into the core and diffusion of fertilizers out of the core[17,20,21].
The release of phosphorus from PS-P(3HB) original and coated NPK granules in the distilled water after 48 hours were 94.17 ± 7.88, and 46.45 ± 0.77%, respectively ($p<0.05$ ). While, the release efficiency of phosphorus from original and coated NPK granules were 65.67, and 33.66%, respectively ($p <0.05$). The release rate of phosphorus from original and coated NPK granules were 1,90 ± 0.06, and 0.98 ± 0.010 mg/h, respectively ($p <0.05$).

The T test results showed the coating on the NPK granules with a significant PS-P(3HB) layer ($p <0.05$) at the release rate of $P_2O_5$. From the efficiency and release rate data, it can be assummed that the release of $P_2O_5$ from the PS-P(3HB) coated NPK granules slower than original NPK granules. The lower the release efficiency of $P_2O_5$, the smaller the NPK amount released. So that the smaller amounts of fertilizer can be washed and disposed into the environment. So the frequency of fertilizer administration can be minimized[18].

For the release of 99.9% of uncoated NPK fertilizer granules was achieved within 58.13 hours. The kinetics profile of NPK fertilizer granule fertilizer which has been coated with PS-P(3HB) polymer resembles kinetics langenbucher. This slow-release NPK fertilizer granulator is released 99.9% in 3647.5 hours or about 5.06 months. The kinetic model is used to predict how profile releases a dosage. In the pharmaceutical field the langenbucher model is usually used to compare drug release profiles using matrices[22,16].

## CONCLUSION

The addition of PS-P(3HB) coating polymer retarded the release of NPK fertilizer granules longer until about 5 months. FTIR analysis of slow release NPK obtained showed no new substances formed or no chemical interaction between NPK granules and coating polymers. The percentage of release of $P_2O_5$ from uncoated and coated NPK granules after 48 hours were 94.17 ± 7.88, and 46.45 ± 0.77% ($p <0.05$), respectively. The release rate of $P_2O_5$ from conventional and coated NPK fertilizer granules, was 1.90 ± 0.06, and 0.98 ± 0.01 %. $h^{-1}$, respectively ($p <0.05$).

The efficiency of release of $P_2O_5$ from uncoated and coated NPK fertilizer granules were 65.67 and 33.66% ($p <0.05$), respectively. Kinetics release of slow release NPK fertilizer follows Langenbucher kinetics ($R = 0.993$). NPK granules coating using PS-P(3HB) had a significant effect on the rate and release efficiency of $P_2O_5$ ($p <0.05$).

## ACKNOWLEDGMENT

The authors would like to express special thanks to the Rector of University of Andalas, Ministry of Research and Higher Education, Goverment of Republic of Indonesia for supporting of the research grant through The Accelerated Research Cluster Scheme to Professor, University of Andalas, Fiscal Year 2018, Contract Number 60/UN.16.17/PP/LPPM/2018.

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**Cite this article as:**

Akmal Djamaan *et al*. Use of bioblend polystyrene-poly (3-hydroxybutyrate) as the coating material of the NPK slow release fertilizer. Int. Res. J. Pharm. 2018;9(10):73-79 http://dx.doi.org/10.7897/2230-8407.0910229

Source of support: Ministry of Research and Higher Education, Goverment of Republic of Indonesia, Conflict of interest: None Declared