Controlled release NPK fertiliser encapsulated by a biodegradable polyester

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Abstract

The commercial NPK granular fertilizer was coated using a biodegradable polyester namely polylactic acid (PLA) in order to prepare controlled release NPK fertilizers (PLA-coated NPK fertilizers). The coatings were prepared from the polymer solution of PLA by immersion method. The effects of PLA concentration (5, 15 and 25% w/v) and number of coating (single and double coatings) on nutrients release behaviour in water (in terms of conductivity) of PLA-coated NPK fertilizers were investigated at room temperature for a period of 28 days. The physical properties of the coatings affect the nutrient release rate. The results also revealed that the release of NPK nutrients from double PLA-coated NPK fertilizers were slower than that of single PLA-coated NPK fertilizers. The release of nutrients through the coating made up of 25% w/v of PLA was the slowest. Among the single and double PLA-coated fertilizers, only 25% w/v double PLA-coated fertilizer (D25) meets the controlled release criteria by the European Committee for Standardization (CEN) . Therefore, this study demonstrated that D25 could acts as controlled release fertilizers (CRFs) which are suitable for sustainable agricultural and horticultural applications.

Keywords: Controlled release fertilizer, polylactic acid, coating, NPK

1. Introduction

Human population has now reached approximately 7.0 billion and forecasted to increase to 9.5 billion by 2050 (Azeem *et al.*, 2014). Global food demand has also increased, with per capita food demand anticipated to double by 2050 (Brown *et al.*, 2009). However, soil fertility and land degradation are among the factors affecting low crop yields, thus threaten global food security (Liliane & Charles, 2020). High usage of fertilizers to increase crop production will certainly cause health problems and environmental impacts (Chandini *et al.*, 2019). Hence, it is vital to develop an approach to enhance crop production and reduce environmental issues.

Global consumption of fertilizer increased drastically in recent years causing serious environmental issues (Savci, 2012). Nowadays, modern agriculture mainly focuses on environmental impacts of crops, quality of crop products, reducing production costs as well as the balanced use of fertilization (Yousaf *et al.*, 2016). Recently, controlled release fertilizers (CRFs) have been used as the most convenient method to increase the efficiency fertilizers application, reduce nutrient loss and environmental effects (Kusumastuti *et al.*, 2019). CRFs are the fertilizer granules encapsulated with materials that control the exposure of soluble fertilizer core to water and/or nutrients release by diffusion (Morgan *et al.*, 2009) to supply active fertilizer nutrient in synchrony with essential need of plant, thus enhanced the nutrient efficiency (Azeem *et al.*, 2014).

A number of natural and synthetic polymers have been introduced for the production of coated fertilizers (Yang *et al.*, 2017; Treinyte *et al.*, 2018; Dai *et al.*, 2018; Wang *et al.*, 2020). However, there are few drawbacks of these polymers which limit their applications. Some of the natural polymers are costly, complicated extraction process (Muhamad *et al.*, 2015) and uncontrollable rate of degradation (Reddy *et al.*, 2021). Meanwhile, synthetic polymers are toxic, non-biodegradable (Muhamad *et al.*, 2015), microbial and heavy metal contaminations (Maghsoudi *et al.*, 2020). Thus, the low-cost biodegradable polymer has been used as an alternative to the traditional polymers and may become a magnificent solution to the environmental pollution (Ojeda, 2013; Naz and Sulaiman, 2017). Different polymeric materials such as polypropylene/poly(acrylic acid) (Yang *et al.*, 2013), polyvinyl alcohol/biochar/bentonite (Liu *et al.*, 2019), polyurethanes/zeolite 4A (Li *et al.*, 2017) and starch/chitosan (Savitri *et al.*, 2019) have been used to develop more eco-friendly controlled release fertilizers in order to gain commercial attention (Olad *et al.*, 2018). In addition, multiple layered CRFs using different polymeric materials have also been reported in several previous studies to overcome the drawbacks of poor release characteristics. For example, a double-layer polymer coated urea (DPCU) with an inner layer of corn stover-based polyurethane and an outer layer of superabsorbent from chicken feather meal was developed by Yang *et al.* (2013). They reported that the release rate was controlled by the coating thickness of the inner layer. Later, Wang *et al.* (2016) formulated a new double-coated CRF by using food-grade microcrystalline wax (MW) and marine polysaccharide derivatives (calcium alginate and chitosan-glutaraldehyde copolymer) which resulted in a controllable and efficient release of nitrogen. In addition, the nitrogen release after 28 days was <75% which meets the European standard for CRFs. Furthermore, Djamaan *et al.* (2018a) applied single and double polystyrene/polycapro lactone coating materials and found that the release rate of active substance release was slower in the double-coated slow-release NPK granules as compared to that of uncoated and single-coated slow-released NPK granules. In comparison to other polymers, polylatic acid (PLA) is an attractive candidate of coating material used in CRFs due to its biocompatibility, better thermal processibility, energy savings, remarkable barrier properties and suitable for packaging items (Farah *et al.*, 2016). Many researchers have utilised CRFs produced from various PLA-based composite encapsulating the fertilizers granules. For instance, Xia *et al.* (2020) have prepared PLA-polycaprolactone composite for preparing slow-release urea. Recently, a study on using polyvinyl alcohol and polylactic acid (PVA-PLA) composite as core/shell fibers of CRFs was explored by Nooeaid *et al.* (2021) for encapsulating nitrogen-phosphorus-potassium (NPK) nutrients. Another investigation by Cen (2021) has integrated the PLA with biochar to develop an innovative biochar-based controlled-release nitrogen fertilizer.

However, to the best of our knowledge, applications of single polymeric material in preparing CRF are still lacking. Therefore, the main objective of this study was to prepare CRFs using a biodegradable polymer namely PLA. Thus, in this study we investigated the effects of percentage w/v (% w/v) of PLA and number of coatings on the NPK release in water in terms of conductivity of CRFs. The optimum conditions of the parameters for NPK release in water were also determined in this study. The optimum PLA coating, uncoated NPK and optimum double PLA-coated NPK fertilizers were characterized by Fourier-transform infrared (FT-IR) spectroscopy and thermal gravimetric analyzer (TGA).

2. Experimental

2.1 Materials

Polylactic acid (PLA) pellets were obtained from NatureWorks (United States) and chloroform ($CHCl_3$) (>99 %) was purchased from Merck, Germany. The commercial NPK fertilizer in granular form was acquired from D Syira Enterprise (Malaysia). The materials were used without further purification. Physical and chemical properties of the uncoated NPK fertilizer are shown in Table 1 and Figure 1, respectively.

Table 1. Physical and chemical properties of the uncoated NPK fertilizer.

Property	
Diameter (mm)	3-5
Color	brown
Nitrogen, N (%)	15
Phosphorous, P (%)	15
Potassium, K (%)	15
Sulphur, S (%)	2
Trace element (%)	0.01

Figure 1. Image of the uncoated NPK fertilizer.

2.2 Methods

2.2.1 Preparation of controlled release NPK fertilizers

2.2.1.1 Preparation of PLA coating solutions

Firstly, the PLA pellets were dried in an oven at 40 ºC for 2 h. Then, the dried PLA pellets (5, 15 and 25 g) were added into 100 mL of chloroform in the conical flasks to prepare different concentrations of PLA solutions (5, 15, and 25 % w/v). Subsequently, the conical flasks were covered with aluminium foil and the mixtures were stirred at 200 rpm for 3 hours at 60 ºC until the homogeneous PLA solutions were obtained. Figure 2 shows the schematic diagram of the preparation of PLA solution.

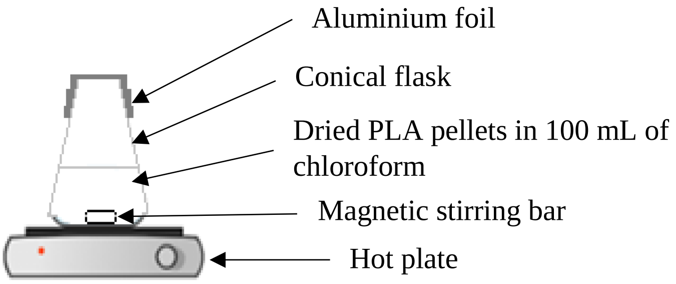


Figure 2. Schematic diagram of the preparation of PLA solution.

2.2.1.2 Preparation of PLA-coated NPK fertilizers

PLA-coated NPK fertilizers were prepared by the immersion method as previously described by Costa *et al.* (2013) using different number of coatings (single and double coatings) and different concentrations of PLA solution (5, 15, and 25% w/v).

Single PLA-coated NPK fertilizer was prepared by immersing 1 g of NPK fertilizer in 5 mL of PLA solutions (5, 15, and 25 % w/v) in different beakers and heated on a hot plate to 50 ºC. After 10 seconds of immersion time, the PLA-coated fertilizers were removed from the beakers, followed by air dried for 3 hours on a clean silicone mat in a fume hood to vaporize the excess PLA coating solution. Then, the samples were kept in screw-cap polyethylene bottles. These single PLA-coated NPK fertilizers were labelled as S5 (5% w/v single PLA-coated NPK fertilizer), S15 (15% w/v single PLA-coated NPK fertilizer), and S25 (25% w/v single PLA-coated NPK fertilizer).

For the preparation of double PLA-coated NPK fertilizers, single PLA-coated NPK fertilizers were re-immersed in their respective PLA coating solutions (5, 15, and 25% w/v) and heated on a hot plate at 50 ºC in a fume hood for 3 seconds. The coated fertilizers were air-dried for 4 hours to obtain the double PLA-coated NPK fertilizers. These double PLA-coated fertilizers were kept in screw-cap polyethylene bottles and labelled as D5 (5% w/v double PLA-coated NPK fertilizer), D15 (15% w/v double PLA-coated NPK fertilizer), and D25 (25% w/v double PLA-coated NPK fertilizer).

2.2.2 Controlled release behaviour of CRFs in water

To monitor the nutrient release rates by the uncoated NPK and the PLA-coated NPK fertilizers in water, 1 g of each fertilizers (3-5 mm in diameter) were added into 200 mL of deionized water in a 500 mL plastic beaker covered with aluminium foil. The nutrients released from the fertilizers were measured at room temperature for 1, 3, 7, 10, 14, 21, 27~~,~~ and 28 days using a conductivity meter (Ohaus Starter 3100M) with a sensitivity of 0.01 µS/cm. Each sample was analyzed in duplicates, and the average value was measured. The cumulative nutrient release (%) of the fertilizers was calculated using equation 1:

$$Cumulative\ nutrient\ release = \frac{Nutrient}{Nutrient_{\ uncoated,28}} \times 100\ \% \tag{1}$$

where *Nutrient* is NPK nutrients release from uncoated NPK or PLA-coated NPK fertilizers (µS/cm) at each time of sampling and $Nutrient_{uncoated,28}$ is NPK nutrients release from NPK fertilizer at day 28 (µS/cm).

The average results of cumulative nutrients release were reported and the European Standard Committee (CEN) standard requirements were used to identify the optimum CRFs.

2.2.3 Preparation of PLA25 film

PLA film was prepared by pipetting 5 mL of 25% w/v PLA solution onto a petri dish. Then, the petri dish was covered with a lid and left overnight at room temperature in a fume hood. On the next day, the PLA film was peeled from the petri dish and abreviated as PLA25.

2.2.4 Characterization of samples

Characterization of PLA25 film, NPK fertilizers and optimum CRFs were conducted using Fourier-transform infrared (FT-IR) spectrometer (Spectrum 100, Perkin Elmer) and thermal gravimetric analyzer (TGA) (STA 8000, Perkin Elmer).

2.3.4.1 Fourier Transform Infrared spectroscopy (FTIR)

The FTIR spectra of the samples were recorded at the wavenumber in the range from 650 to 4000 cm^{-1} using the attenuated total reflectance (ATR) method.

2.3.4.2 Thermal Analysis

Thermogravimetric analysis was performed by heated an approximately 5 mg of each sample from $30^{o}C$ to $700^{o}C$ under nitrogen atmosphere at a constant flow rate of 20 mL/min and a heating rate of $10^{o}C$/min. The temperature, weight and derivative of the sample were determined after heating.

3. Results and discussion

3.1 NPK nutrient release analysis

The most important characteristics for CRFs are nutrient release behavior and rate (Fertahi *et al.*, 2021). Distilled water is widely used as the environment for release testing by most researchers due to its validity and readily available release environment for comparison results (Fertahi *et al.*, 2021). For instance, Djamaan *et al*. (2018b) applied water immersion tests at different time intervals to evaluate the NPK release in order to examine the performance of the uncoated and coated fertilizers by measuring the amount of phosphate release. Therefore, in this study, the nutrient release behaviours of uncoated NPK fertilizers, single and double PLA-coated NPK fertilizers in water were evaluated.

NPK nutrient release study in water (in terms of conductivity) were conducted using uncoated NPK fertilizers, single and double PLA-coated NPK fertilizers at 1, 3, 7, 10, 14, 21, 27 and 28 days to determine the optimum PLA-coated NPK fertilizer based on CEN's standard. In this study, effect of the concentration of PLA coating (5, 15, and 25% w/v) and coating number (single and double) on cumulative nutrient release rates of NPK in water from the uncoated NPK and PLA-coated NPK fertilizers were investigated, as shown in Figure 3.

Figure 3 illustrates the cumulative NPK release rates of uncoated NPK, single and double-coated NPK fertilizers. As shown from Figure 3, 76% NPK nutrients released immediately from uncoated NPK during the first 24 h and achieved constant release rate (95%) at day 7. The nutrients release rates of single PLA-coated NPK fertilizers (S5, S15 and S25) were slower than that of uncoated NPK fertilizers. Within 24 h, the S5 released 9 % of the NPK nutrients and the rate accelerated at day 3 (78%), followed by a steady released after day 7. The S5 achieved 95% NPK nutrients release at day 14, which is 2 times slower than that of uncoated NPK. For S15, small amount (5 %) of nutrients was released at day 1 and almost complete nutrients release at day 27 which corresponding to 3.9 times slower than uncoated NPK. The release rate of NPK from S25 was initially slow (less than 10 %) and then increased drastically (47 %) at day 3, then became slower until day 28 (83 %). The results demonstrated that a longer period is required for S25 to achieve complete release of nutrients compared to that of S5, S10 and uncoated NPK fertilizers. Hence, these results can prove that single PLA-coated NPK fertilizers were able to control the NPK nutrients released into water at slow rates.

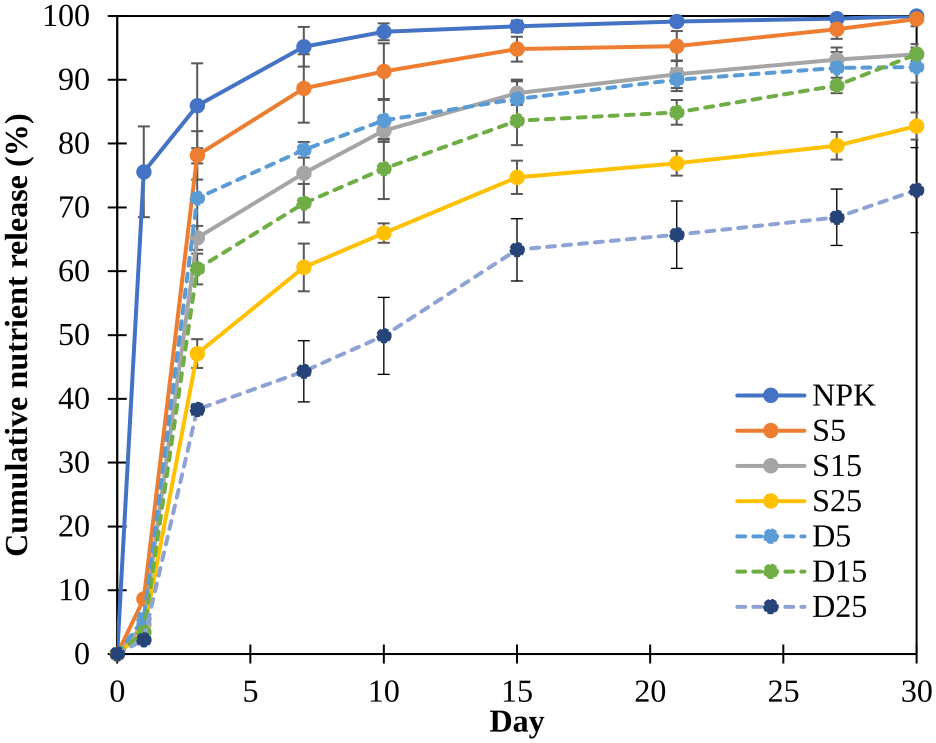


Figure 3. Cumulative nutrient release rates of NPK and PLA-coated NPK fertilizers in water.

As expected, increasing the thickness of the coating delayed the release of nutrient compared to uncoated and single PLA-coated NPK fertilizers (Figure 3). The results revealed that the nutrients release rates from double PLA-coated NPK fertilizers (D5, D15, and D25) differ significantly from the uncoated NPK fertilizers (achieved more than 90 % release within 7 days). At the beginning of the release period, D5, D15 and D25 released less than 6 %, followed by higher release in the range of 36% to 71 % at day 3. Then, the release rates became slower and only D5 and D15 achieved almost complete release at day 27 and 28, respectively. As shown in the curve, approximately 6% and 71% of nutrients were released from D5G at day 1 and 3, respectively. At day 27, the nutrient released from D5G was 92%, which is 3.9 times slower compared with the uncoated NPK fertilizer. The D15 also required similar period to achieve 92% of nutrient release but slightly slower nutrients released at day 1 and day 3 as compared to D5. However, D25 required longer period than both D5 and D15 in order to attain complete release of the nutrients.

Slightly slower NPK released by S5 and D5 as compared to uncoated NPK, probably attributed by the thin PLA coatings owing to small amount (5% w/v) of PLA used. Based on Figure 4, the results also demonstrated that the NPK nutrients release decreased with increasing PLA concentrations (5 to 25% w/v) for all PLA-coated NPK fertilizers. Similar findings were also reported by Da Cruz *et al*. (2017), Niu and Li (2012), and Fern´andez-P´erez *et al*. (2008). Lower amount of coating material and consequently a thinner layer (Da Cruz *et al*., 2017) will cause swelling and easier water transport to the fertilizers (Mulder *et al*., 2011) which resulted in higher nutrient release rate. In contrast, higher amount of coating material, for example NPK with 25% w/v of PLA coating exhibited significant delays in NPK nutrient release of 83% and 69% in S25 and D25, respectively, at 28 days. As expected, a high concentration of polymer increased the thickness of the coating film and slowed the release of nutrients owing to a greater thickness and lower porosity in the coating layers of fertilizer granules (Fertahi *et al.*, 2021).

One (Beig *et al.*, 2020; Fertahi *et al.*, 2021) or several layers (Kusumastuti *et al.*, 2019; Cui *et al.*, 2020; Fertahi *et al.*, 2021) of coating material can be coated on the fertilizer granules using the same coating solution, but single and double layers are the most common number of coating thickness used to coated the fertilizers (Fertahi *et al.*, 2021; Beig *et al.*, 2020; Kusumastuti *et al.*, 2019; Cui *et al.*, 2020). Multiple layers increase the coating thickness and decrease the release nutrient rate due to a more compact structure of the coating and a lower porosity compared to a single coating (Fertahi *et al.*, 2021; Jarosiewicz and Tomaszewska, 2003). As presented in Figure 3, the release profile of double PLA-coated fertilizers was slower than single PLA-coated fertilizers. These results also agree with the previous findings from Liang and Liu (2006) which reported that the release rate of the double polystyrene-coated urea granules was slower than that of single-coated. Hence, it can be concluded that PLA concentration and number of coating thickness highly affected the rate of NPK nutrients released.

The standard of CEN was applied to identify the optimum CRFs from single and double PLA-coated NPK. Figure 4 presents the cumulative nutrients released from uncoated NPK, single and double PLA-coated NPK fertilizers at day 1 and day 28. The results verified that the cumulative nutrients released from all PLA-coated NPK fertilizers were lower than 15% on the first day. However, only D25 released nutrients less than 75% on the 28 days. These findings denoted that the controlled release behaviour of the D25 formulation prepared in this study fulfilled the CEN's criteria for CRFs.

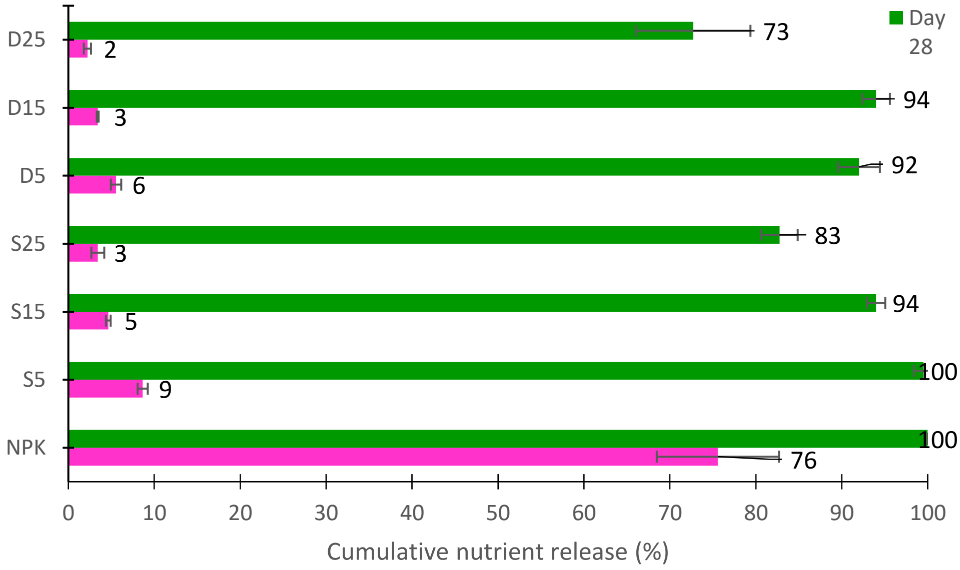


Figure 4. Comparison of NPK release from uncoated, single and double PLA-coated NPK fertilizers at day 1 and day 28.

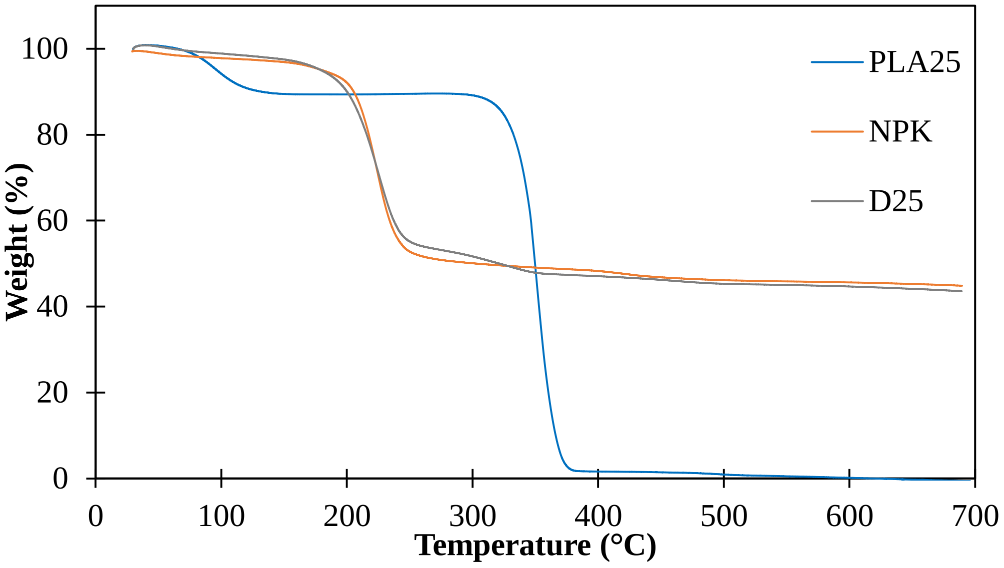
3.2 Thermal analysis

Thermal analysis (thermal gravimetric analysis (TGA) and derivative thermal gravimetric (DTG) were conducted to investigate the thermal decompositions of PLA25, uncoated NPK and D25. As shown in Figure 5(a), the TGA thermograms of PLA25 shows two thermal degradation stages in which the initial decomposition of PLA25 occurred from degradation range of 47ºC to 154ºC with 10% weight loss due to the elimination of chloroform solvent (Fredi *et al.*, 2021) and moisture (Rahaman *et al.*, 2020) through dehydration process. Final decomposition occurred at 385°C with 88% weight loss which corresponding to the decomposition of main and side chain groups of polymers (Król-Morkisz and Pielichowska, 2019) (Figure 5(a)). The maximum degradation temperature of PLA25 was 355 °C (Figure 5(b)). Hoidy *et al.* (2010) also found that PLA decomposed from 265°C to 381°C (Wisam *et al.* 2010), while Alhadadi *et al.* (2019) found the maximum degradation temperature for PLA at 350 ^{o}C. As shown in Figure 5(a), complete decomposition of PLA25 was observed at 551 ^{o}C.

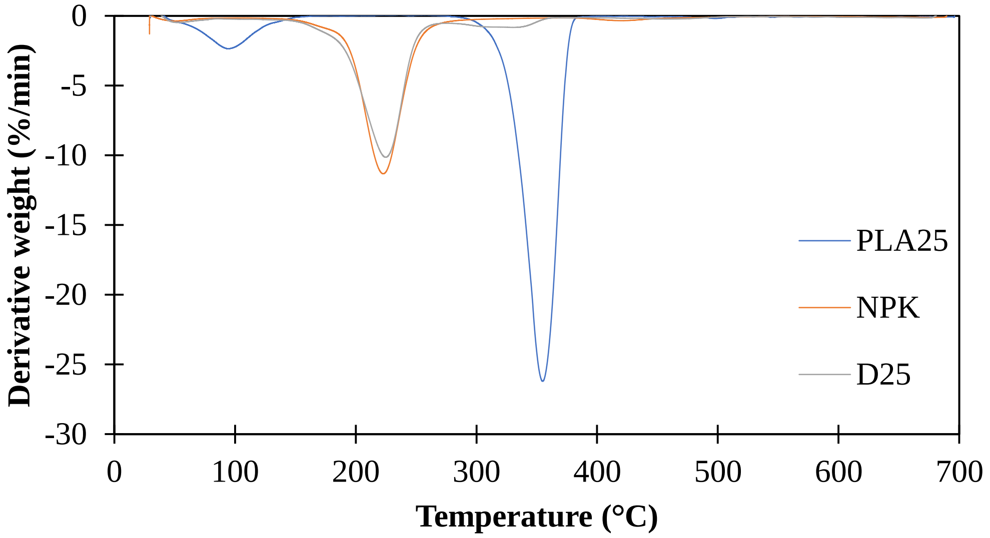
Based on the TGA and DTG thermograms (Figure 5), uncoated NPK have three thermal degradation stages. The multi degradation of NPK fertilizers also found by Harmaen *et al.* (2014) after characterized the NPK fertilizer and poly(lactic acid)/fertilizer/oil palm fibers blends biocomposites. The initial weight loss of uncoated NPK was 1% from 31ºC to 83ºC which corresponded to the moisture loss (Noppakundilograt *et al.*, 2014). The second weight loss of 47% was recorded between 145ºC to 294ºC which could be due to decomposition nitrogen nutrient in the form ammonium sulfate (Noppakundilograt *et al.*, 2014). The final stage of thermal degradation that might be associated with the decomposition of phosphorus nutrient in the form of phosphorus pentoxide (P_2O_5) (Noppakundilograt *et al.*, 2014) was observed between 395°C to 457°C (weight loss 2%). Mohan *et al.* (2007) also mentioned that P_2O_5 melts at 340^{o}C and sublimes at 360^{o}C. It is observed that the maximum degradation temperature of the uncoated NPK was at 223^{o}C (Figure 5(b)) and the residual content of the uncoated NPK was 45% at the temperature close to 700^{o}C (Figure 5(a)). The complete decomposition of uncoated NPK fertilizer might take place when the temperature of the system is higher than 700^{o}C. This phenomenon was also supported by Harmaen *et al.* (2014), stated that the decomposition of the fertilizer takes place only when the temperature of the system is higher than certain value for particular conditions.

TGA curve of D25 shows four thermal degradation stages (Figure 5(a)). The first degradation stage started from 34ºC to 86ºC with a weight loss of 1% (Figure 5(a)) due to the elimination of chloroform solvent (Fredi *et al.*, 2021). Then, D25 further degraded from 148ºC to 271ºC, 324ºC to 363ºC, and 415ºC to 501ºC with weight losses of 44%, 2%, and 2%, respectively. Other than decomposition of nitrogen and phosphorus nutrients from NPK fertilizer at second and fourth stages, respectively, the weight losses occurred at third stage could be attributed to the decomposition of PLA coating. The third stage decomposition (324ºC to 363ºC) of D25 was supported by TGA thermogram of PLA25 from 281^{o}C to 385^{o}C (Figure 5(a)). The residual content found in D25 (44 %) at 689^{o}C owing to the incomplete decomposition fertilizers such as potassium compound (Noppakundilograt *et al.*, 2014).

It can be concluded that the uncoated NPK and D25 were more thermally stable than PLA25 as indicated by their high residual contents. Thermal stabilities of D25 were lower than uncoated NPK fertilizers. Saffian *et al.* (2016) also reported similar results that the degradation temperature of NPK combined with PLA was lower than the neat PLA degradation temperature.



(a)



(b)

Figure 5 (a) TGA and (b) DTG thermograms of PLA25, uncoated NPK and D25 samples.

3.3 Fourier Transform Infrared (FT-IR) analysis

To examine the existence of polymer, FT-IR analysis were performed and the spectrum of PLA25 was compared with the spectra of uncoated NPK fertilizer and PLA-coated NPK fertilizer (D25) (Figure 6).

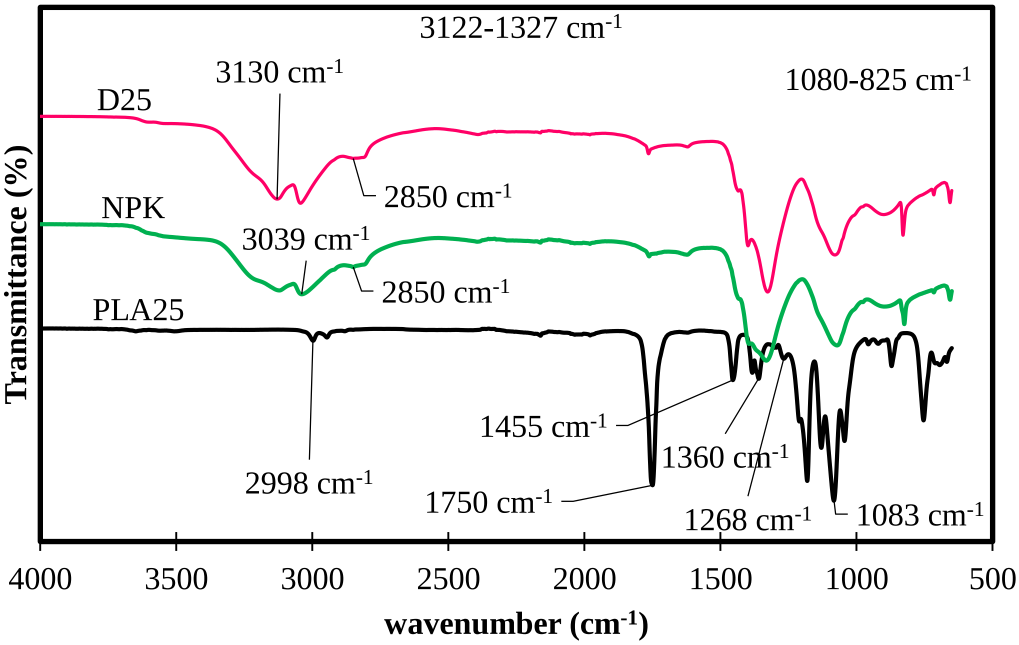


Figure 6 FT-IR spectra of PLA25, D25 and uncoated NPK fertilizers.

The infrared spectrum of PLA25 with characteristic peaks of PLA at 2998, 1750, 1455, 1360, 1268, 1083 cm^{-1} are shown in Figure 6. The presence of peaks at 2998 cm^{-1} corresponded to -CH stretching of alkane and methylene group (Birnin-Yauri *et al.*, 2017; Alhadadi *et al.*, 2019), 1750 cm^{-1} corresponded to -C=O stretching of the carbonyl group (Birnin-Yauri *et al.*, 2017; Alhadadi *et al.*, 2019), 1455 and 1360 cm^{-1} corresponded to C-H bending (Birnin-Yauri *et al.*, 2017; Alhadadi *et al.*, 2019), and 1083 cm^{-1} corresponded to C-O stretching in the ester group (El Assimi *et al.*, 2020) in PLA.

The FTIR spectrum of the uncoated NPK showed similarities with the D25 spectrum as shown in Figure 6. The FTIR spectra of the uncoated NPK and D25 were observed in the regions of 3122–1332 cm^{-1} and 3130–1327 cm^{-1}, respectively, which were attributed to the stretching and deformation vibrations of NH_4^+ ion (Liang *et al.*, 2017). The peaks in the regions of 1072–825 cm^{-1} and 1080–829 cm^{-1} of the uncoated NPK and D25, respectively, were probably indicated the presence of symmetric and asymmetric stretching vibrations of P-O-P groups (Yang *et al.*, 2014; Liang *et al.*, 2017). In other report, the existence of phosphate P-O stretching vibration was observed at peak 879 cm^{-1} (Dhlamini *et al.*, 2020). In addition, the peaks of uncoated NPK at 3039 cm^{-1} and D25 at 3130 cm^{-1} were corresponded to the stretching vibration of broad hydroxyl (O-H) group (Udvardi *et al.*, 2019), while the peaks at 2850 cm^{-1} of uncoated NPK and 2850 cm^{-1} of D25 might assigned as stretching vibration of -C-H group (Razak *et al.*, 2020).

The similarity between FTIR spectra of uncoated NPK and D25 proved there is no significant interactions between NPK granules and PLA coating in D25. Therefore, it can be proved that NPK fertilizers and PLA polymer coating are compatible (Suardi *et al.*, 2020). Similar explanations also documented by Djamaan *et al.* (2018a), Suharti *et al.* (2016), Watson (2005) and Ben *et al.* (2020).

4. Conclusion

In this work, single and double-coated PLA CRFs were successfully prepared by coating the NPK fertilizers with PLA coating using immersion method. The effects of the PLA concentrations and coating number on nutrients release behaviour were investigated. In general, the concentration of the PLA coating and the number of coatings applied onto NPK granules affected the nutrient release rate in water. The NPK nutrient release from all PLA-coated NPK fertilizers were slower than that of uncoated NPK fertilizer, and the slowest release rate was observed using 25% w/v double PLA-coated NPK fertilizer. The results also demonstrated that the NPK nutrients release decreased with increasing PLA concentrations (5 to 25 % w/v) for all PLA-coated NPK fertilizers. Based on the study of the coating number on the nutrients released profile, the results showed that the release of NPK nutrients from double PLA-coated NPK fertilizers were slower than their respective single PLA-coated NPK fertilizers. Moreover, D25 released nutrients less than 15 and 75 % at day 1 and 28, respectively, which proved that D25 satisfies the controlled release criteria of the European Standards for CRFs. Hence, it was concluded that D25 coated formulation was the optimum CRF and has good potential in agricultural and horticultural applications.

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