



Preparation and characterization of slow-release fertilizer encapsulated by biochar-based waterborne copolymers



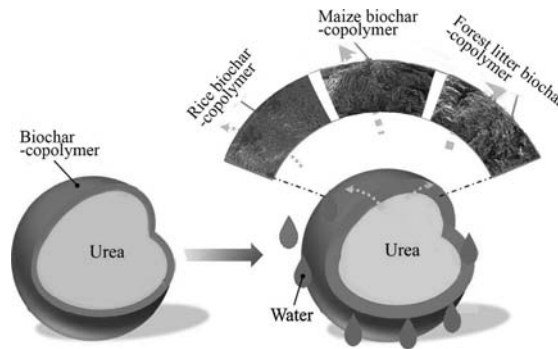
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HIGHLIGHTS

- Biochar improve water resistance and degradability of copolymer films.
- Botanical origins and doses of biochar affect structures and properties of the blend films.
- Rice biochar-based copolymer coated urea exhibit an excellent release behavior of 65.28% nutrient leaching on the 22th day.

GRAPHICAL ABSTRACT



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ABSTRACT

To enhance the effectiveness of polyvinyl alcohol (PVA) coated fertilizers, a novel slow-release fertilizer was developed using biochar and waterborne copolymer of PVA and polyvinylpyrrolidone (PVP) as coating materials. The effects of botanical origins and doses of biochar as well as copolymer concentrations on biochar-copolymer structures and properties were investigated. Our results indicated that the water absorbency of blend films differing in biochar origins showed different responses to these three factors. Generally, biochar decreased water absorbency of copolymer with an increased degradability, and contributed to improving the slow-release property of coated urea. In particular, rice biochar-based copolymer (S5 film) had less hydrophilic —OH bonds and encapsulated urea granules more compactly and densely. The urea particles coated with rice biochar-based copolymer (S5 film) exhibited an excellent release behavior of 65.28% nutrient leaching on the 22th day. Therefore, this work has demonstrated the potential of biochar-based copolymer from different botanical biochar for improving the effectiveness of fertilizers.

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1. Introduction

Synthetic fertilization plays crucial role in guaranteeing food security around the world, which contributes almost 50% of the increase in crop yield (Roberts, 2009; Sutton, 2011; Yang et al., 2016). However, sub-optimal or over-fertilization results in a decrease of fertilizer use efficiency and subsequently leads to severely environmental and ecological

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problems (Azeem et al., 2014; Fishedick et al., 2014; González et al., 2015; Pereira et al., 2015; Vries et al., 2013).

Controlled release fertilizers (CRFs) could release nutrients gradually, which try to coincide with the nutrient demand during crop growth. Thus, the CRFs products show great potential in improving fertilizer use efficiency and alleviating environmental problems caused by fertilizer management (Qiao et al., 2016; Li et al., 2008; Wu et al., 2008). Generally, CRFs products are prepared by physically encapsulating fertilizer granules with hydrophobic inorganic and/or organic materials that serves as a diffusion barrier (González et al., 2015; Han et al., 2009).

Among those CRFs, polymer coated fertilizer is one of the most widely used types of the slow-release fertilizers. Currently, several polymer materials have been proposed for CRFs encapsulation. These polymer materials are mainly divided into two categories according to the solvent types used in CRFs production. Compared with organic-solvent-dissolvable polymers, waterborne polymers are relatively cheap and environmentally non-toxic during production (Yang et al., 2012). Consequently, increasing interests are focused on waterborne polymer coatings which exhibit excellent coating properties (Zhou et al., 2015).

Polyvinyl alcohol (PVA) is a hydrophilic, biodegradable and non-toxic polymer which is mainly composed of C—C bonds (Tudorachi et al., 2000; Zhao et al., 2006). However, the effects of PVA coated fertilizer on retarding nutrients release are limited especially at field scale. This efficiency could be improved though cross-linking PVA with oxalic acid or glutaraldehyde, which forms a high density structure with low permeation flux (Das et al., 2011). In our previous study, we find that masking hydroxyl groups in PVA improves water resistance and slow-release properties of PVA (Zou et al., 2015). Polyvinylpyrrolidone (PVP) has advantages of water-soluble, low toxicity, good film-forming, chemical stability and well compatible with PVA. Meanwhile, —H of C=O in PVP molecules could bond —OH groups of PVA, which improves the physical properties of PVA films (Kamada et al., 2003).

Biochar could increase soil porosity and water-holding capacity, whereas reduce soil bulk density for its porous structure. Meantime, biochar could be used as a support material for delivering plant nutrients (González et al., 2013; González et al., 2015; Lehmann et al., 2011). Previous studies indicate that biochar addition increases the degradability of polymer film, which is attributed to the fact that biochar could adsorb soil microorganisms (Kallenbach and Grandy, 2011; Lehmann et al., 2011; Novak et al., 2009). Moreover, biochar-modified waterborne polyacrylate materials enhance the mechanical strength and prolong the nutrient release period, hence demonstrate great potential for improving waterborne polymer-coated CRFs (Zhou et al., 2013). However, the slow-release behaviors of biochar-based waterborne copolymer of PVA and PVP are still unknown.

The present work attempted to develop a novel slow-release nitrogen fertilizer through coating urea granules with biochar-based waterborne copolymers. We prepared blend films of biochar and copolymers of PVA and PVP through cross-linking. The effects of botanical origins and doses of biochar as well as copolymer concentrations on biochar-copolymer structures and properties were investigated. Then, the slow-release fertilizers coated with biochar-based copolymers were prepared, and their nutrient release behaviors in soil were evaluated.

2. Materials and methods

2.1. Materials

All chemicals used in the present work were analytically pure and commercially available. PVA, PVP and ammonium sulfate were purchased from Sinopharm Chemical Reagent Co., Ltd. (China). Allochroic silicagel was from Shanghai Xinhua silicone factory. Large granular urea was supplied from Shanxi Tianze coal chemical group Co., Ltd. (Total N \geq 46.4%). Xinlianxin slow release urea was from Henan Xinlianxin Chemical Group Company (Total N \geq 43.4%).

2.2. Experimental design

The feedstocks of biochar were maize straw, rice straw and forest litter, respectively. For each biochar source, an orthogonal experiment including 3 factors and 3 levels was designed, and L9 (3^4) orthogonal table was adopted. The experiment for each biochar source included 9 treatments and 3 controls without biochar addition (Table A1), with 3 replications.

2.3. Preparation of water-based copolymer and biochar coated urea

2.3.1. Biochar

Maize straw (Y), rice straw (S) and forest litter (K) samples were collected from the experimental station of Shenyang Agricultural University. The samples were cut into small pieces of 2–3 cm, and dried at 70 °C for 24 h. The dried samples were crushed and ground to pass a 40-mesh sieve. Then, the crushed samples (20 g for each sample) placed into porcelain crucibles were heated to 500 °C at a rate of 15 °C min⁻¹ in the muffle furnace. The temperature of 500 °C was maintained for 2 h. Afterwards the samples were cooled to ambient temperature, and grounded to pass 60, 100 and 200 mesh sieves, respectively. Ultimately, biochar samples were put into sealed bags for subsequent films synthesis.

2.3.2. Biochar-based waterborne copolymer films

The synthesis of films was started by placing distilled water and PVA into the three-necked flask. The temperature was slowly raised to 90 °C until the mixture was completely dissolved through continuously stirring. When the temperature was decreased to 60 °C, PVP and 0.5 g butanol were added and continuously stirred for 2 h. The purpose of butanol addition was to avoid frothing. Then, biochar was added and thoroughly mixed for another 1 h at 60 °C. Finally, the mixtures were cast onto leveled glass plates and naturally air dried.

2.3.3. Biochar-based waterborne copolymers coated urea

Before coating, urea granules were sieved to obtain fertilizer particles with a diameter of 2–3 mm, and those with surface defects were removed. Urea granules (1 kg) were preheated in a sugar-coating machine (TN-IIA, Dandong Medicinal Machine Co., Ltd.) for 0.5 h. The ratio of biochar-based copolymer film:urea was 1:20 (w/w). Using a side-spray nozzle (PQ-2) under 0.7 MPa pressure, the urea granules were coated with the mixture solution of biochar and copolymer (50 ml) for 0.5 h. The angle and speed of drum were 45° and 60 r min⁻¹. The nitrogen content of the coated urea prepared in our experiment was determined using the Kjeldahl Method.

2.4. Water absorbency (WA) of biochar-based copolymer films

The films were cut into 3 cm \times 3 cm pieces, and immersed in a certain amount of distilled water for 3 h. The swollen films were weighed after removing excess water (M_1). The weight of dried films (M_2) was measured through the following procedure: The swollen films were first dried in a vacuum oven at 60 °C for 3 h and weighted, afterwards dried 1 h and weighted. This drying duration of 1 h was repeated until the weight of films did not change. WA was calculated as follows:

$$WA = M_1/M_2 \times 100\% \quad (1)$$

where WA is the water absorbency, M_1 is the weight of saturated films, and M_2 is the weight of dried film (Kima et al., 2002).

2.5. Biodegradability of biochar-based copolymer films

Soil burial degradation was performed as described by Zhao et al. (2006) with a slight modification. The air-dried film samples (0.1–0.3 mm thickness) were cut into 3 \times 3 cm pieces and buried in

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