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Research article

Effects of process parameters on the distribution characteristics of inorganic nutrients from hydrothermal carbonization of cattle manure

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ABSTRACT

In this study, dairy manure was converted into solid and liquid products via hydrothermal carbonization (HTC) to determine optimal reaction conditions to retain the greatest amounts of inorganic nutrients. The influence of three parameters, reaction temperature (150–270 °C), residence time (0.5–6 h) and manure/water ratio (5/200–25/200 g/mL), on the total nutrient content (TNC) in the solid and liquid products were investigated using a 5-level, 3-factor orthogonal test and principal components analysis (PCA). Also, the distribution characteristics of inorganic nutrients were determined using single factor tests. The maximum TNC of the solid product was 22.92 mg/g, obtained at manure/water ratio of 20/200 g/mL and performed at 240 °C for 6 h. For obtaining the maximum TNC of liquid product, the most optimal conditions were 150 °C, 1 h and 20/200 g/mL which produced TNC of 1619.55 mg/L. The proportion of ammonium (NH_4^+-N) and orthophosphate (PO_4^{3-}) were also analyzed in the liquid product. The amount of NH_4^+-N increased with reaction temperature increasing and manure/water ratio decreasing, the main form of phosphorus (P) was PO_4^{3-} in the liquid product, and most of the potassium (K) were also dissolved in the liquid product. These results indicate that the HTC could be a promising approach for the utilization of dairy manure as inorganic fertilizer in the future.

1. Introduction

Rapid development of the livestock and poultry industry promotes economic growth, but it also leads to increasingly serious environmental pollution. In addition to the multiple nutritive elements and heavy metals, livestock manure contains many pathogenic bacteria, parasite ova and other microorganisms, which easily releases greenhouse gas and toxic gas (Zhang and Wu, 2010). Zhu et al. (2008) predicted that by the year 2020, the total emission of livestock manure in China will reach 4.22 billion tons annually, which would cause enormous amounts of environmental pollution and may hinder the sustainable development of the livestock and poultry industry. Therefore, it seems to be urgent to realize the harmless treatment and resource utilization of livestock manure.

To date, the primary applications of livestock manure are as fertilizer, feed stuff and an energy resource. Livestock manure and compost are two kinds of high quality organic fertilizers, as evidenced by their common usage in direct application to agricultural fields. However, commercial composting requires greater production costs and longer processing times than collecting and processing livestock manure for fertilizer. As a good feed resource, due to different properties of different kinds of livestock manure, chicken manure has attracted more attention to be converted into renewable feed (Zhang and Wu, 2010). Additionally, livestock manure is a good energy resource as the energy of one ton of dry livestock manure was equal to the energy of 0.375 ton of standard coal (Tu et al., 2007). Energy utilization of livestock manure not only makes a high added-value product, but also produces economic profits. Nevertheless, as the main proposal method of energy utilization, anaerobic treatments has found wide application, nevertheless, which accounts for only 5–10% in the usage of livestock manure (Huang et al., 2008). Therefore, it has been the key issue of finding a new technology to realize the conversion of livestock manure to increase the energy utilization.

Thermochemical conversion technology can be used to convert livestock manure to solid, liquid and gaseous fuels that have high energy density and commercial value and are more convenient for storage and transportation (Huang et al., 2008). Hydrothermal carbonization (HTC) is one of the thermochemical technologies

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which converts biomass in water at high temperatures and pressures to hydrocarbon fuels (Mumme et al., 2011). Much attention has been attracted to the usage of hydrochar to fabricate functional carbonaceous materials, which has been applied to carbonaceous adsorbent for pollutant adsorption and removal (Regmi et al., 2012), carbon nanomaterials (Titirici and Antonietti., 2010), carbon-based catalyst (Demir-Cakan et al., 2010), carbon-based fuel cell (Titirici and Antonietti., 2010) and soil amelioration (Reza et al., 2014). Recently, it has been a research focus that how to recycle the liquid product from HTC process, which is rich in inorganic nutrients, minerals and soluble organic matter. A number of studies have investigated the production of soluble organic matter in the aqueous product by the hydrothermal treatment. Cheng et al. (2008) demonstrated that during the hydrolysis of fish waste, chicken waste, hair and feather, 18 kinds of amino acids were obtained, and most amino acids reached maximum yield at a reaction temperature range of 200–290 °C and a reaction time range of 5–20 min. Quitain et al. (2002) investigated the production of low-molecular-weight carboxylic acids from the hydrothermal treatment of organic wastes. Their results showed that organic acids such as acetic, formic, propionic, succinic and lactic acids were obtained in significant amounts. Moreover, the presence of oxidants favored formation of organic acids with acetic acid being the major product. Kang and Chun (2004) elucidated the behavior of protein decomposition to amino acids in near-critical water, and observed that serine, aspartic acid and other complex amino acids obtained initially in significant amount, gradually decreased as reaction time and temperature increased.

Furthermore, extensive research has focused on the effect of hydrothermal treatment conditions on the distribution characteristics of inorganic nutrients and minerals. Wang et al. (2016) studied the effects of reaction time on the transformation of N, P, K and heavy metals in sewage sludge. The results indicated that the transfer rate of N in the sludge transferring to aqueous product increased gradually, almost all of the P remained in the solid phase and most of the K (57–62%) was still in the solid phase. The transferring behavior of heavy metals was different with the increase of reaction time. Ekpo et al. (2016) performed thermal hydrolysis and hydrothermal processing of microalgae, manure, and digestate. They observed that most of the N and K were present in the aqueous phase and the proportion of organic-N is higher at a lower temperature. Furthermore, extraction of P was linked to the presence of metal elements such as Ca, Mg and Fe in the feedstock.

Dry dairy manure is a high-quality raw material, consisting of crude protein (13.74%), crude fat (1.65%), coarse fibre (43.61%) and nitrogen free extract (22.94%). The content of available organic matter can measure up to 81.93% (Moore and Gamroth, 1991). However, none of the studies have systematically tested how the three processing parameters during HTC of dairy manure, including reaction temperature, residence time and manure/water ratio, influence the amounts of inorganic nutrients from livestock manure and what could be the most optimal conversion conditions for liquid and solid products respectively. This study aimed to investigate the effect of three HTC conditions on the distribution of N, P, K and other metals in the manure-derived solid and liquid products.

2. Material and method

2.1. Material

Fresh dairy manure was obtained from Dong Zheng Livestock Breed Co., Ltd. in Hubei Province, China. Prior to weighing and being used in HTC experiments, the dairy manure samples were dried for 24 h at 105 °C and milled to the desired particle size of 40 mesh. These prepared samples were stored in a sealed plastic bag for the HTC treatment.

2.2. Experimental design

This study identified the distribution characteristics of N, P, K and other metals in HTC of dairy manure at temperatures ranging from 150 °C to 270 °C, residence times ranging from 0.5 h to 6 h and manure/water ratios ranging from 5/200 g/mL to 25/200 g/mL. These experimental conditions are summarized in Table 1. The factors that may affect the total content of NPK were determined primarily through a 3-factor, 5-level orthogonal test, coupled with principal components analysis (PCA). Then, the optimal combination of factors and levels tested that influenced the distribution characteristics of N, P, K and other metals of the solid and liquid products were selected on the basis of the same HTC experiment to conduct single factor tests.

2.3. Experimental procedure

The HTC of dairy manure was conducted in a 1 L Hastelloy reactor (GSH-L). For each experiment, dry dairy manure was weighed and transferred into the reactor and mixed with deionized water. The mixture was continuously stirred at 600 revolutions per minute (rpm) during the HTC process. The reactor was heated to and maintained at the preset temperature for the duration of a programmed time. After residence time at the required temperature, the reactor was cooled down by an internal cooling tube with cooling water and an external fan. It took approximately 60–90 min to cool from inner temperature to 65 °C. The pressure release valve opened and the gaseous products were allowed to vent in the fume hood, and the pressure was reduced to the atmospheric level. The solid and liquid products were separated by vacuum filtration through a Buchner funnel and preweighed filter paper. The solid product was dried overnight at 105 °C and stored in a dryer for further analysis. The liquid product was frozen and stored in a refrigerator for further analysis. Each experiment was carried out three times, and the solid and liquid products were examined individually.

2.4. Analysis of products

The solid product yield by weight was calculated as the ratio of the dry weight of solids product to the dry weight of the dairy manure sample, according to Eq. (1):

$$W = (m_1/m_0) \times 100 \quad (1)$$

where W is the solid product yield, %; m_0 is the dry weight of dairy manure, g; m_1 is the dry weight of solid product, g.

The solid and liquid products and dry dairy manure were digested in sulfuric acid and hydrogen peroxide (V/V = 5/1) using a MARS 6 microwave digestion system, and the digestive solution was diluted with deionized water to 500 mL. Then, the total nitrogen (TN) and total phosphorus (TP) concentrations of the diluted

Table 1
Factor levels and values for orthogonal experiment.

Factors	Code	Code Level				
		1	2	3	4	5
Temperature (°C)	A	150	180	210	240	270
Residence time (h)	B	0.5	1	2	4	6
Manure/water ratio (g/mL)	C	5/200	10/200	15/200	20/200	25/200