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Research article

Advanced treatment of gamma irradiation induced livestock manure using bioelectrochemical ion-exchange reactor

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ABSTRACT

Organic matter and nitrogen in livestock manure was pre-treated by gamma irradiation. The optimal dose ranged 30–50 kGy for solubilization of organic matter and nitrogen. Carbohydrates and proteins increased with the applied dose. Lipids did not show a regular increase pattern. A large amount of organic nitrogen in livestock manure was solubilized after gamma irradiation. The pre-treated livestock manure was treated using a bioelectrochemical ion-exchange reactor. High removal of organic matter and nitrogen was achieved with the applied dose of 50 kGy. The maximum 88.5% of chemical oxygen demand removal was obtained in the bioelectrochemical ion-exchange reactor due to readily biodegradable chemical oxygen demand fraction. Nitrogen removal was significantly affected by ammonia flux of ion-exchange membrane between anaerobic and aerobic chamber. With a high ammonia flux of 4.7513 mg/m^2/sec, the maximum ammonia removal was 79.1%.

1. Introduction

Livestock contribute approximately one half of agricultural output and support the livelihoods and food security of almost 1.3 billion people. Husbandry, one of the fastest growing sectors of the agricultural economy, is growing rapidly along with the human population. Much of this growth was connected to several emerging issues and challenges such as resource scarcity, environmental pollution and pathogen (FAO, 2013; Thornton, 2010). Several reports warned about environmental challenges resulting from livestock manure. Contaminants and pathogens can leach soils and rivers through the poorly designed wastewater treatment. Pollutants derived from manure result in significant contamination of water and soil resources. High strength of manure easily leads to oxygen depletion and eutrification in water bodies. Soil and aquifer can be severely contaminated by untreated manure. Livestock manure can be a significant threat to human health when manure left the farm as surface runoff or groundwater infiltration or reused for irrigation. Occurrence of several antimicrobial resistant bacteria originated from livestock manure and emergence of human disease results from animal production were reported (Burkholder et al., 2007; Otte et al., 2007).

Anaerobic digestion of livestock manure has been thoroughly adopted in the world. Concentrated solids in manure were mostly suitable for slurry mixing type. However, low removal efficiency and long hydraulic retention time lead researchers to find advanced treatment processes. This technology also takes times to be compliant to stringent guidelines without additional treatment. Several investigators have paid attention to develop more effective process for removing non-biodegradable organic matter in livestock manure. Unlike waste sludge, macromolecular substances such as proteins and carbohydrates in manure, which are represented as slowly biodegradable chemical oxygen demand (SBCOD) including some readily biodegradable chemical oxygen demand (RBCOD), are mainly composed of the breakdown fraction of insoluble food. Most SBCOD in livestock sludge are caused by fermented fraction in manure and animal debris. Advanced oxidation processes such as Fenton, Photo-Fenton and photocatalytic system was applied for treating organic matter and pathogens in livestock wastewater (Asha et al., 2015; Park et al., 2006; Lee and Shoda, 2008). Fenton process using hydroxyl radicals is effective to remove non-biodegradable organic matter, but generating large quantity of chemical sludge. Radiation process was considered as a promising technology for treating livestock manure. Ionizing irradiation can destroy macromolecular structures into RBCOD by a lot of radical species produced during water radiolysis. Radiolytic

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ionization processes such as gamma irradiation and electron beam carry out excellent organic matter removal without any by-products. Biodegradability of SBCOD such as textile wastewater, landfill leachate, waste sludge and petroleum production was enhanced after ionizing irradiation (Bae et al., 1999; Chu et al., 2011; Duarte et al., 2004; Kim et al., 2004; Wu et al., 2014; Yin and Wang, 2015).

In this study, to obtain higher fraction of RBCOD in livestock manure, the advanced oxidation by gamma irradiation was carried out. Removal of the organic matter and nitrogen in solubilized livestock manure using a bioelectrochemical ion-exchange reactor was investigated. The solubilized organic matter contributed to enhanced organic matter and denitrification in the reactor.

2. Materials and methods

2.1. Livestock manure

Livestock manure generated in Korea was generally transported into a livestock wastewater treatment plant. In order to obtain representative sample, livestock manure was collected from a livestock wastewater treatment plant in Jeongeup, Korea. The plant adopts an anoxic-aerobic process with a chemical phosphorus precipitation. Sample was collected every month and stored at 4 °C until use. The physicochemical characteristics of livestock manure were measured including pH, oxygen reduction potential (ORP), organic matter and nitrogen (Table 1).

2.2. Pre-treatment of livestock manure

After collecting livestock manure, it was physically oxidized by gamma irradiation. Gamma irradiation was achieved at different doses of 10, 30, 50 and 70 kGy using ^{60}Co source (Nordion, Canada) at Korea Atomic Energy Research Institute. The dose rate was 10 kGy/hr, and the radioactivity of source was 1.47x10^{17}Bq. The absorbed dose was measured using the alanine-EPR dosimetry system (ISO/ASTM 51607:2003).

2.3. Bioelectrochemical ion-exchange reactor

The reactor was configured with three separated chambers; anaerobic-aerobic-anoxic. Aerobic chamber was hydraulically separated, but the others connected. Cations in the influent were ion exchanged via a cation exchange membrane (CEM, ASTOM, Tokyo, Japan) between anaerobic and aerobic chamber. To accelerate cations movement, 3 V of DC power (OTM-501T, ODA, Korea) was supplied to anode and cathode. Each electrode (Stainless steel; 2 mmx150 mmx100 mm) installed at anaerobic and aerobic chamber was immersed to a 5 cm depth. To maintain aerobic conditions, 5L/min of air was continuously provided at aerobic chamber. Anions at aerobic chamber were ion exchanged via an anion exchange membrane (AEM, ASTOM, Tokyo, Japan) between aerobic and anoxic chamber. Anaerobic and anoxic chamber were thoroughly mixed. The fermented organic matter at anaerobic chamber was provided for denitrifiers at anoxic chamber. The solubilized livestock manure by gamma irradiation was treated during 85 days at room temperature. Operating conditions of the bioelectrochemical ion-exchange reactor during the study period were shown in Table 2.

Table 1
Physicochemical characteristics of livestock manure.

Parameter	Value (Ave.±Std.)[a]
pH	7.8–8.6 (8.2 ± 0.4)
ORP (mV)	−415.6 - -100.6 (−370.0 ± 254.5)
DO (mg/L)	0.2–0.8 (0.3 ± 0.5)
Alkalinity (mg $CaCO_3$/L)	9358.4–16520.6 (12840.0 ± 505.4)
BOD (mg/L)	2100.8–6800.3 (4200.5 ± 944.2)
TCOD (mg/L)	8475.2–13186.8 (10503.7 ± 766.5)
SCOD (mg/L)	5414.0–11050.0 (7818.6 ± 868.6)
Carbohydrates (mg/L)	145.5–354.7 (233.3 ± 45.2)
Proteins (mg/L)	976.3–2134.2 (1472.4 ± 153.5)
Lipids (mg/L)	3550.5–6834.5 (4595.0 ± 154.8)
T-N (mg/L)	1056.7–2688.8 (1742.6 ± 102.4)
NH_3-N (mg/L)	968.6–1969.8 (1497.6 ± 135.5)
NO_2-N (mg/L)	0.0–5.6 (0.3 ± 0.5)
NO_3-N (mg/L)	0.9–53.8 (13.6 ± 14.7)

[a] Minimum-maximum (average ± standard deviation).

2.4. Analytical methods

Samples were collected every two each weeks. The characteristics of livestock manure were measured according to Standard Methods for the Examination of Water and Wastewater (APHA, 1998). Carbohydrates were measured by the Dubois method (1956), proteins by the Layne method (1957) and lipids by the Bligh method (1959). Cations and anions were measured with an ion chromatography (ICS-2000, Dionex, Sunnyvale, USA).

3. Results and discussion

3.1. Characteristic of livestock manure after gamma irradiation

Livestock manure is one of representative refractory waste containing over 40% of total organic matter is non-biodegradable. Most refractory organic matter in livestock manure is originated from animal feedings and some animal debris. Since digestive molecules are mostly composed of humic substances, it is necessary to oxidize them to small molecules (Andreadakis, 1992). Radiation-induced degradation of pollutants can be a very effective tool for cleavage of high molecular weight humic substances. Water radiolysis produces several radicals such as hydroxyl radical (·OH), hydrogen radical (·H), and solvated electron (e^-_{eq}). These radicals play a role in degrading livestock manure. The concentration change of organic matter in livestock manure after gamma irradiation is shown in Fig. 1. The biodegradability enhancement of refractory wastes is one of main aims at advanced oxidation by gamma irradiation. Solubilization of organic matter in livestock manure was carried out by gamma radiolytic ionization. Carbohydrates concentration in manure was 233.3 mg/L before gamma irradiation. It sharply increased up to a dose of 30 kGy. However, over 50 kGy of gamma irradiation little affects carbohydrates concentration. The change of carbohydrates concentration was maintained between 340.7 and 352.5 mg/L. The maximum 51.1% increase of carbohydrates was shown at a dose of 70 kGy. Proteins concentration continuously increased with gamma irradiation, but slightly decreased at a dose of 70 kGy. While 2543.5 mg/L (72.7% of proteins) was solubilized after gamma dose of 50 kGy, 409.3 mg/L of proteins decreased at 70 kGy. It was shown that 19.2% decrease of proteins as compared of that at 50 kGy. Proteins can be oxidized into smaller nitrogen-containing molecules by hydroxyl radicals. It was inferred that proteins in livestock manure were transferred into more simple fraction such as amino acids with 70 kGy of gamma irradiation. For lipids, the obvious change pattern by gamma irradiation was not shown. Lipid degradation is achieved by direct radical oxidation or indirect chain reaction mechanism of free radicals. Radicals produce a fatty acid radicals, which is initiated by ·OH and ·OOH combined with a hydrogen atom. However, 17.3% of lipids decreased after gamma irradiation at a dose of 30 kGy. When the concentration of radicals is enough, radical recombination is usually observed in radiolytic reaction. The