Waste Management xxx (2017) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Low temperature circulating fluidized bed gasification and co-gasification of municipal sewage sludge. Part 2: Evaluation of ash materials as phosphorus fertilizer

Tobias Pape Thomsen [a,*], Henrik Hauggaard-Nielsen [b], Benny Gøbel [c], Peder Stoholm [d], Jesper Ahrenfeldt [a], Ulrik B. Henriksen [a], Dorette Sophie Müller-Stöver [e]

[a] Technical University of Denmark, Department of Chemical Engineering, DTU Risø Campus, Technical University of Denmark Building 313, Frederiksborgvej 399, 4000 Roskilde, Denmark
[b] Roskilde University, Department of Environmental, Social and Spatial Change, 4000 Roskilde, Denmark
[c] DONG Energy Thermal Power A/S, Nesa Allé 1, 2820 Gentofte, Denmark
[d] Danish Fluid Bed Technology ApS, Industrivej 38, 4000 Roskilde, Denmark
[e] University of Copenhagen, Department of Plant and Environmental Sciences, Plant and Soil Science, Thorvaldsensvej 40, Frederiksberg, Denmark

ARTICLE INFO

Article history:
Received 11 January 2017
Revised 23 April 2017
Accepted 26 April 2017
Available online xxxx

Keywords:
Municipal sewage sludge
Cereal straw
Thermal gasification
Phosphorus fertilizer ash
Heavy metals

ABSTRACT

The study is part 2 of 2 in an investigation of gasification and co-gasification of municipal sewage sludge in low temperature gasifiers. In this work, solid residuals from thermal gasification and co-gasification of municipal sewage sludge were investigated for their potential use as fertilizer. Ashes from five different low temperature circulating fluidized bed (LT-CFB) gasification campaigns including two mono-sludge campaigns, two sludge/straw mixed fuels campaigns and a straw reference campaign were compared. Experiments were conducted on two different LT-CFBs with thermal capacities of 100 kW and 6 MW, respectively. The assessment included: (i) Elemental composition and recovery of key elements and heavy metals; (ii) content of total carbon (C) and total nitrogen (N); (iii) pH; (iv) water extractability of phosphorus after incubation in soil; and (v) plant phosphorus response measured in a pot experiment with the most promising ash material. Co-gasification of straw and sludge in LT-CFB gasifiers produced ashes with a high content of recalcitrant C, phosphorus (P) and potassium (K), a low content of heavy metals (especially cadmium) and an improved plant P availability compared to the mono-sludge ashes, thereby showing the best fertilizer qualities among all assessed materials. It was also found that bottom ashes from the char reactor contained even less heavy metals than cyclone ashes. It is concluded that LT-CFB gasification and co-gasification is a highly effective way to purify and sanitize sewage sludge for subsequent use in agricultural systems.

1. Introduction

Phosphorus (P) is an essential macro nutrient and the availability of P in agricultural systems is often a limiting factor especially in older soils as the ones found in mid and lower Africa, Asia and Australia (Cordell and White, 2014; Van Vuuren et al., 2010). The main source for P fertilizer, mined phosphate rock, is a critical non-renewable globally demanded resource and there is an increasing concern about the commercial availability of this resource in the near future (Cordell and White, 2014). Determinations of the P depletion rate and especially quantifications of the remaining P resource have been the subject of a recent scientific debate (Edixhoven et al., 2013; Scholz and Wellmer, 2016, 2013), but it is generally agreed on that the geopolitical importance of the P resource is increasing as is the urgency of developing more efficient P management strategies (Chowdhury et al., 2016; Ott and Rechberger, 2012).

A substantial proportion of P used in agriculture ends up in municipal sewage sludge (MSS) (Kahiluoto et al., 2015), and recycling this fraction via direct application of MSS to agricultural soil has been considered a cheap and efficient way to enhance and fertilize soils (Chowdhury et al., 2016; Fytili and Zabaniotou, 2008; Linderholm et al., 2012). However, the extent of direct MSS soil application varies greatly among countries and regions, and the variation is caused by many different factors including practical alternatives, differences in sludge quality as well as political and cultural restrictions (Fytili and Zabaniotou, 2008; Hukari et al.,

* Corresponding author.
E-mail address: ttho@kt.dtu.dk (T.P. Thomsen).

http://dx.doi.org/10.1016/j.wasman.2017.04.043
0956-053X/

2016; Kelessidis and Stasinakis, 2012). In recent years, there has been a growing concern in many countries of the potential risks associated with the content of emerging organic pollutants and xenobiotics in MSS, including antibiotics, fragrances, UV-filters, antiseptics, micro plastics, phthalates, hormones and much more (Choban and Winkler, 2008; Igos et al., 2012; Krüger et al., 2014; Michael et al., 2013). As a consequence of this growing concern, there is an increasingly restrictive political attitude towards direct application of sewage sludge in many countries (Krüger and Adam, 2015), and alternative management options are continuously developed and implemented. Thermal gasification of sludge is one of these alternatives (Qian and Jiang, 2014).

All types of thermal conversion of municipal sewage sludge leads to production of one or more ash- and/or char fractions. The quality and quantity of these products can vary substantially with the quality of the parent sludge and the design of the thermal process (Fericelli, 2011; Jakobsen and Willett, 1986; Li et al., 2015; Qian and Jiang, 2014). Many scientific studies have been conducted to examine the potential application of incineration ashes and pyrolysis chars in agricultural systems as fertilizers and/or soil enhancers. These studies usually involve investigation of one or several of the following characteristics: Content and type of toxins; fate and mobility of heavy metals; determination of eco-toxicity levels; content and availability of macro- and micro nutrients; technical routes for down-stream upgrading and potential long-term carbon sequestration (Fraser and Lum, 1983; Furr et al., 1980, 1979; Hossain et al., 2015; Jakobsen and Willett, 1986; Liu et al., 2014; Lu et al., 2013; Mellbye et al., 1982; Méndez et al., 2012; Song et al., 2014; Sousa and Figueiredo, 2015). However, to this date, no published studies on the fertilizer quality of ashes and chars from thermal gasification of municipal sewage sludge have been identified. Most studies on thermal gasification of sewage sludge are focused on the potential energy recovery, the technical feasibility of the process and the gas quality (Manara and Zabaniotou, 2012; Seggiani et al., 2012; Zhu et al., 2015).

The aim of this study was to test the use of two Low-Temperature Circulating Fluidized Bed Gasifiers (LT-CFBs) to convert MSS focusing on the quality of the solid process residuals and the potential use of these solid products as fertilizers and/or soil enhancers. Gasification ashes were collected from two MSS campaigns, two MSS/straw co-gasification campaigns and a straw reference campaign. Ash analysis included (i) Composition and elemental balances of macro nutrients and heavy metals; (ii) pH measurements and (iii) incubation studies and pot experiments to determine phosphorus fertilizer quality.

This work is part of a larger assessment of the suitability of the LT-CFB gasification technology as a platform to convert municipal sewage sludge to electricity, heat and ash fertilizer. The complete assessment is composed of the following two parts:

- Part 1: Assessment of process feasibility and stability, process performance and energy efficiency, product distribution and gas product characteristics (Thomsen et al., 2017).
- Part 2: Characterization of LT-CFB ashes as P fertilizer and determination of key elemental balances (this study).

2. Materials and methods

2.1. The LT-CFB gasifier

The LT-CFB gasifier has several characteristic features which make it a very suitable platform for MSS gasification. The technology is very fuel flexible and can operate on many different types of fuels (e.g. straw, biogas- and manure fibers and different organic residues from industry) as long as the primary fuel requirements related to particle size (<0.5 cm) and moisture content (<30 wt%, wet basis) are complied with (Ahrenfeldt et al., 2013). The ability to operate on high alkaline fuels makes the LT-CFB an interesting platform in Denmark and other countries with similar large resources of herbaceous biomass, and it also expands on the possibilities to co-gasify MSS and biomass.

Two LT-CFB units currently exist; a 100 kWth pilot scale plant at the Technical University of Denmark located at Risø near Roskilde, Denmark, and a 6 MWth demonstration unit at DONG Energy's Asnaes Powerplant in Kalundborg, Denmark. Both plants are involved in the present study. The LT-CFB technology is commercially registered as the Pyroneer Gasifier by DONG Energy and has been described previously (Ahrenfeldt et al., 2013; Narayan et al., 2016; Nguyen et al., 2013; Nielsen, 2007; Thomsen et al., 2017, 2015). A generalized process flow chart of the LT-CFB gasifier is provided in Fig. 1.

Operation is usually conducted with maximum temperatures below 750 °C and total system air equivalence ratios around 0.3. In all campaigns, quartz sand with 50 wt% of particles below 0.13 mm and 95 wt% of particles below 0.25 mm in diameter is applied as bed material (DanSand.dk, 2013). No additives were added.

2.2. The sewage sludge fuels and LT-CFB campaigns

The applied MSS samples originated from three different Danish wastewater treatment plants (WWTPs) and the samples were collected at different seasons. The three WWTPs that supplied the sludge samples for the study are characterized as follows:

Stegholt WWTP (Aabenraa, Denmark): Constructed as a Mechanical-Biological (active sludge loop)-Nitrification-Denitrification-Chemical cleaning facility (MBNDC). However, the use of precipitation chemicals for P capture has been phased out through constant process optimization. Iron chloride is still applied in the clarifiers to optimize the sedimentation of floating sludge. The MSS is treated by thermophilic anaerobic digestion for up to 18 days at the WWTP before it is dewatered mechanically and exported.

Randers WWTP (Randers, Denmark): A representative Danish MBNDC where P is captured approximately 50% biologically and 50% chemically using a mix of iron chloride and aluminum. This estimation is based on the annual use of precipitation chemicals at the time of the LT-CFB experiment and an assumption of a 1:1.8 capturing rate for aluminum and of 1:3 for iron. All sludge goes through the active sludge cycle before it is digested anaerobically in a mesophilic process with a retention time close to one month. The sample for the LT-CFB campaign was dried at the WWTP in a Krüger BioCon dryer with temperatures ranging from 100 to 175 °C.

Bjergmarken WWTP (Roskilde, Denmark): Another representative Danish MBNDC facility where precipitation chemicals are added before the active sludge cycle and phosphorous is captured approximately 70% biologically and 30% chemically. Iron chloride sulfate and aluminum chloride is used to capture P and precipitate sludge. All sludge goes through the active sludge cycle before it is digested anaerobically for almost three weeks in a thermophilic digestion process. After digestion, the sludge is mechanically dewatered and dried in a Krüger BioCon dryer (100–175 °C). MSS products can be delivered as de-watered sludge, as dry granules or as dry pellets.

The LT-CFB campaigns include a reference campaign on straw (REF campaign), two campaigns on mono sludge fuels (SLU campaigns) and two campaigns with co-gasification of sludge and straw (MIX campaigns). A short description of the campaigns is provided in Table 1. More details and data on fuels and LT-CFB campaigns can be found in part 1 of the study (Thomsen et al., 2017). The fuels applied in the MIX-ST and MIX-BJ experiments