Water Research 91 (2016) 11–18

Contents lists available at ScienceDirect

Water Research

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

Predicting the drying properties of sludge based on hydrothermal treatment under subcritical conditions

Mikko Mäkelä [a, *], Laurent Fraikin [b], Angélique Léonard [b], Verónica Benavente [c], Andrés Fullana [c]

[a] Swedish University of Agricultural Sciences, Department of Forest Biomaterials and Technology, Skogsmarksgränd, 90183 Umeå, Sweden
[b] University of Liège, Department of Chemical Engineering, Agora, B6c, Allée du 6 Août, 4000 Liège, Belgium
[c] University of Alicante, Department of Chemical Engineering, P.O. Box 99, 03080 Alicante, Spain

ARTICLE INFO

Article history:
Received 2 September 2015
Received in revised form
15 December 2015
Accepted 24 December 2015
Available online 29 December 2015

Keywords:
Biosolids
Moisture diffusivity
Hydrochar
Hydrothermal carbonization
Shrinkage
X-ray microtomography

ABSTRACT

The effects of hydrothermal treatment on the drying properties of sludge were determined. Sludge was hydrothermally treated at 180–260 °C for 0.5–5 h using NaOH and HCl as additives to influence reaction conditions. Untreated sludge and attained hydrochar samples were then dried under identical conditions with a laboratory microdryer and an X-ray microtomograph was used to follow changes in sample dimensions. The effective moisture diffusivities of sludge and hydrochar samples were determined and the effect of process conditions on respective mean diffusivities evaluated using multiple linear regression. Based on the results the drying time of untreated sludge decreased from approximately 80 min to 37–59 min for sludge hydrochar. Drying of untreated sludge was governed by the falling rate period where drying flux decreased continuously as a function of sludge moisture content due to heat and mass transfer limitations and sample shrinkage. Hydrothermal treatment increased the drying flux of sludge hydrochar and decreased the effect of internal heat and mass transfer limitations and sample shrinkage especially at higher treatment temperatures. The determined effective moisture diffusivities of sludge and hydrochar increased as a function of decreasing moisture content and the mean diffusivity of untreated sludge ($8.56 \cdot 10^{-9}$ m^2 s^{-1}) and sludge hydrochar ($12.7–27.5 \cdot 10^{-9}$ m^2 s^{-1}) were found statistically different. The attained regression model indicated that treatment temperature governed the mean diffusivity of hydrochar, as the effects of NaOH and HCl were statistically insignificant. The attained results enabled prediction of sludge drying properties through mean moisture diffusivity based on hydrothermal treatment conditions.

1. Introduction

Handling of sludge residues generated by biological and chemical wastewater treatment is becoming ever more challenging due to rapid urbanization and increasing efficiency requirements for municipal and industrial wastewater treatment plants. Sludge residues are widely complex materials due to a variety of structural components such as extracellular polymeric structures, filamentous bacteria, cationic salts and the potential presence of pollutant precursors, e.g., proteins and fats, pathogens, parasites, trace metals, polychlorinated biphenyls, dioxins and other slowly decomposable compounds (Vaxelaire and Cézac, 2004; Yoshikawa and Prawisudha, 2014a). Currently the most common sludge handling methods include incineration, composting, use in agriculture or disposal in landfills (Mahmood and Elliott, 2006; Mowla et al., 2013), although current trends in European regulation are increasingly hindering landfill deposition of organic substances. As sludge is an inherently wet material decreasing associated handling, storage and transportation costs generally requires active drying especially for smaller-scale treatment plants. However current means of mechanical dewatering suffer from difficulties in removing intracellular and chemically bound water from the polymeric matrix of sludge suspensions (Mowla et al., 2013; Stoica et al., 2009).

* Corresponding author. Current address: Tokyo Institute of Technology, Department of Environmental Science and Technology, G5-8, 4259 Nagatsuta-cho, Midori-ku, Yokohama 226-8502, Japan.
E-mail addresses: makela.m.aa@m.titech.ac.jp, mikko.makela@slu.se (M. Mäkelä), laurent.fraikin@ulg.ac.be (L. Fraikin), a.leonard@ulg.ac.be (A. Léonard), veronica.benavente@ua.es (V. Benavente), andres.fullana@ua.es (A. Fullana).

http://dx.doi.org/10.1016/j.watres.2015.12.043
0043-1354/

Hydrothermal treatment performed with water acting as a solvent, a reactant and even a catalyst or catalyst precursor does not require prior drying of a sludge feedstock and allows simultaneous elimination of biologically active organisms or compounds (Peterson et al., 2008). Hydrothermal treatment under subcritical conditions can be used for producing carbonaceous hydrochar with relatively high yields (Libra et al., 2011) and enables the conversion of non-traditional feedstock such as municipal and industrial sludge residues. The increasing ion product of water under hydrothermal conditions typically favours reactions which are catalysed by acids or bases and is generally understood to proceed via a network of hydrolysis, dehydration, decarboxylation, polymerization and aromatization of biomass components (Kruse et al., 2013; Sevilla and Fuertes, 2009). As hydrothermal treatment is ideally operated under saturated steam pressure the latent heat requirement of evaporation can also be avoided although post-separation of solid and liquid phases is still required. Irrespective of potential hydrochar applications such as direct solid fuel replacement, soil amelioration or metal/metalloid adsorption (Alatalo et al., 2013; Libra et al., 2011; Titirici and Antonietti, 2010), elimination of hydroxyl and carboxyl groups followed by subsequent aromatization can enhance the drying properties of sludge due to increased hydrophobicity and sludge cell breakage (Wang et al., 2014; Yoshikawa and Prawisudha, 2014b; Zhao et al., 2014b).

The drying properties of a material are conveniently characterized through the drying curve where drying rate is described as a function of drying time under specific drying conditions. The attained drying curve allows the identification of controlling mechanisms such as moisture evaporation from saturated or unsaturated surfaces or diffusion within a material (Mujumdar, 2007). In addition, effective moisture diffusivity can be used for compiling empiric drying data to a single parameter describing moisture transfer independent of the actual mechanisms involved (Gómez-de la Cruz et al., 2015). Although diffusivity coefficients are often determined through a simplification of Fick's second law of diffusion, reliable estimations for non-rigid materials can only be attained if material shrinkage is taken into account (Bennamoun et al., 2013). Non-extrusive imaging techniques such as X-ray microtomography have been shown to provide detailed quantitative information on material shrinkage and thus enable a better understanding of the relationships between drying properties and the evolution of size and shape (Léonard et al., 2004, 2008).

For reliably determining the effect of hydrothermal treatment on the drying properties of sludge, we illustrate the use of X-ray microtomography for monitoring sample shrinkage during drying of sludge and hydrochar samples. Respective effective moisture diffusivities affected by shrinkage were then determined by applying recent developments on the interpretation of experimental drying data. Finally, the effect and statistical significance of treatment conditions such temperature, retention time and additive quality on the mean moisture diffusivity of hydrochar were determined using multiple linear regression. The attained results help in understanding the effect of process conditions on the drying properties of hydrothermally treated sludge and controlling these properties based on treatment conditions.

2. Material and methods

2.1. Sampling and sample preparation

Sludge samples were attained from a Swedish pulp and paper mill using virgin sulphate and recycled fibre pulp for the production of unbleached kraft/euroliner for corrugated cardboard. Mill effluents were treated by primary gravitational settling followed by biological activated sludge treatment. Approximately 300 kg of mixed sludge (containing 60% of primary sludge and 40% of biosludge) was sampled after primary sludge and surplus biosludge from secondary sedimentation had been mixed and dewatered to approximately 27% dry solids (2.7 kg H_2O kg^{-1} db, dry basis) using a belt filter and a centrifuge at the mill. The 300 kg sample was coned and quartered (Gerlach et al., 2002) to a representative 10 kg subsample which was stored in +4 °C during the experiments.

2.2. Hydrothermal treatment

Hydrothermal experiments were performed with a 1 L non-stirred stainless steel reactor (Amar Equipments PVT Ltd., Mumbai, India) illustrated Fig. 1a. A constant 300 g mass of sampled sludge was thoroughly mixed with 75 mL of additive and loaded into the reactor. The reactor was heated to reaction temperature using a 1.5 kW electric heating resistance and an additional heating plate placed under the reactor. The 1.5 kW heating resistance was PID controlled as the additional heating plate was set to reaction temperature. Reactor pressure was indicated by a pressure gauge and was approximately equivalent to saturated vapour pressure of water under respective reaction temperatures (i.e., 1–5 MPa). As the isothermal retention time was complete, the reactor was cooled with pressurized air and the gases released into a fume hood. The solid and liquid phases were subsequently separated by vacuum filtration through a grade 413 VWR® filter paper (VWR International LLC, Radnor, PA, USA).

The experiments were conducted according to an experimental design including reaction temperature (180–260 °C) and log10 transformed retention time (0.5–5 h) as continuous controlled variables. In addition, additive type was included as a discrete variable to influence reaction conditions by mixing 75 mL NaOH (0.01 N, pH 12.1) or HCl (0.01 N, pH 2.5) with the feed material prior to loading into the reactor. Deionized H_2O was used as a control (conductivity <6 mS cm^{-1}) and both NaOH and HCl used for making the solutions were reagent grade. NaOH was chosen as it is commonly used in the chemical recovery cycles of pulp mills and the inclusion of HCl enabled evaluating the effect of a wider pH range. Additive concentrations were chosen based on Lu et al. (2014) and the final solid load adjusted to remain in the range applicable to dewatered sludge within the pulp and paper industry. As the design included a discrete variable it was constructed to allow the use of dummy variables in linear regression and response surface methodology (Myers et al., 2009). The final design (see Table 1) was composed of 15 individual experiments.

2.3. Drying experiments and X-ray microtomography

Prior to drying the individual sludge and hydrochar samples were shaped to ensure comparability of initial stress states. The samples were compressed for 1 min under 50 N in a cylindrical compression cell (⌀ 20 mm) consisting of a movable piston and a closed grid that allowed water to be removed. The obtained cylindrical samples were then cut to lengths equivalent to 4.10 ± 0.041 g in sample mass. The corresponding sample volumes were hence dependent on the specific properties of sludge or hydrochar.

Drying experiments were performed with a laboratory microdryer by following sample mass loss under constant and reproducible drying conditions, Fig. 1b (Léonard et al., 2002). The prepared samples were inserted to a drying chamber (cross-section 41 × 46 mm) on a grid suspended under a precision scale to allow convective drying on the entire external surface. The mass of the samples were then recorded every 5 s under an air velocity of 1.5 m s^{-1}, a temperature of 105 °C and an absolute humidity of