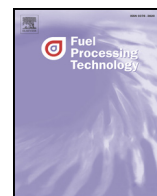




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Research article

## Upgrading of soot sludge from waste to fuel by means of low temperature drying in fixed beds

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## ABSTRACT

Soot sludge is formed in the production of formic acid. Currently, this waste is combusted together with heavy fuel oil in a boiler. The current theoretical lower heating value of the sludge is  $-23.8 \text{ MJ/kg}_{\text{d.b.}}$ . Drying of sludge increases the heating value, and decreases oil use. Drying of pure sludge in a fixed bed is impossible in practice and therefore sawdust is mixed with it before drying. This study aims to fill the gap in knowledge about soot sludge and sawdust drying by studying its drying kinetics experimentally in a fixed bed. The influence of drying on fuel oil consumption is also preliminarily evaluated. Drying curves were determined for bed heights of 100, 200 and 300 mm, inlet air temperatures of 40, 80 and 100 °C, and air velocities of 0.75 and 0.9 m/s. According to the results, the optimal mixture ratio of soot sludge and sawdust is 50%<sub>soot sludge</sub>: 50%<sub>sawdust</sub> (vol.%), with an air velocity of 0.75 m/s and a bed height of 200 mm. Oil savings of 104 MJ/kg<sub>dry, soot sludge</sub> are achieved when the mixture is dried to the final moisture content of 40%.

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## 1. Introduction

Soot sludge is waste which is formed in the production process of formic acid and hydrogen peroxide at the Eastman Chemical Company [1]. This production plant is situated in Oulu, Finland. Soot sludge has extremely high moisture content (95% wet basis). In addition, soot sludge has a very sticky and viscous nature. At present, moist soot sludge is disposed of by combusting it in a combined heat and power (CHP) plant of Laanilan voima Plc [2], which is situated at the production site of Eastman. Fuel oil is mixed with soot sludge in order to combust it. Oil mixing also reduces the viscosity of soot sludge, thus enhancing its pumping properties. This study focuses on the possibility of upgrading soot sludge from waste to fuel by drying it in a direct fixed bed dryer with low temperature air (100 °C or less) before combustion.

Chen et al. [3] and Tunçal and Uslu [4] extensively reviewed different dehydration technologies (removal of water without vaporization and thermal drying) for various municipal and industrial waste sludges, respectively. In [5–9], the authors only studied the dewatering of sludges without vaporization. These studies deal with filtration and belt pressing of conditioned municipal activated sludge and sewage sludge electro-dewatering, for example. Many of the recent studies focus on the thermal drying of various sludges: either municipal sludges [10–17,33], industrial sludges [18–20] or both [21–25]. Both indirect and direct types of thermal dryers are used in sludge drying. Direct

types include rotary-drum, flash, and belt dryers, and indirect types include thin-film, screw, paddle, and disc-type dryers. In some applications, the dryer can also be a combination of both indirect and direct dryers (e.g., in [11]). When indirect dryers are used, the heat source is typically high temperature steam. In this study, low temperature heat sources are used. The other problem regarding indirect dryers is the extremely sticky nature of soot sludge. Due to its sticky nature, soot sludge adheres to the dryer's surfaces, which weakens heat transfer from the heat source to sludge. Due to the previously mentioned reasons, indirect dryers are not necessarily the best alternatives when soot sludge is dried at a temperature of below 100 °C. Scientific studies focusing on the thermal drying of soot sludge discussed in this paper have not been found in the literature. Differences in the origin of various waste sludges mean that they can notably differ from each other, such as in terms of water content, solid structure, chemical composition, ability to concentrate, and drying characteristics [20]. As a result, it is difficult to apply the drying kinetics of a certain sludge to sludge with a different origin.

The present study partly aims to fill the gap in knowledge about soot sludge drying and its drying kinetics by experimentally investigating drying kinetics with the help of drying curves and pressure drops over the bed. However, the drying of pure soot sludge in a fixed bed is in practice impossible and therefore sawdust is mixed with sludge before drying. Li et al. also studied the drying properties and kinetics of sludge and sawdust mixtures in a fixed bed [10,12]. In these studies, the sludge was municipal sewage sludge, the properties of which differ from those of soot sludge. The moisture content of sewage sludge is 85.5% (wet basis) after mechanical dewatering [12], which is clearly lower than

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the moisture content of soot sludge after the centrifugation process (95% wet basis). Li et al. also mention that the stiffness of sewage sludge is poor [10], which indicates that sewage sludge is not as viscous and sticky as soot sludge. In this study, sawdust was chosen as a mixing material because it is available at the CHP plant of Laanilan voima, but other materials could also be used as mixing materials (e.g., wood chips).

The other goal of the study is to determine the fuel oil reduction achieved by drying the soot sludge and sawdust mixture in a continuous belt dryer before combustion in the CHP plant. Experimentally measured drying curves in a fixed bed can be used directly to calculate the cross-sectional area of a continuous belt dryer. The optimal drying parameters of soot sludge and sawdust drying are tentatively evaluated in the study by determining the cross-sectional area needed for a belt dryer.

When suitable waste heat streams are available for the heat source of the dryer, drying is a potential option for converting the negative lower heating value of the soot sludge to a positive one (i.e., from waste to fuel). This reduces the use of fuel oil at the CHP plant. In general, it is usually not meaningful to dry moist fuels before combustion if waste heat is not available, because all dryers consume more heat than the lower heating value of the fuel increases. According to Laanilan voima, the final moisture content of the soot sludge and sawdust mixture should be approximately 40% w.b. ( $\sim 0.7 \text{ kg}_{\text{H}_2\text{O}}/\text{kg}_{\text{d.b.}}$ ) to be able to combust it properly in the boiler and to avoid the dusting of material. Suitable waste heat streams for the dryer are available either from the plant of Laanilan voima Plc or the production plant of Eastman. The most potential waste heat sources are hot water streams, the temperatures of which may range from over 40 °C to slightly over 100 °C, depending on the available water stream. The earnings gained from reduced oil use are presented in this study.

## 2. Materials and methods

### 2.1. Formation of soot sludge

Fig. 1 illustrates the formation process of soot sludge. Among other products, the Eastman Chemical Company [1] produces formic acid and hydrogen peroxide using heavy fuel oil as the main raw material. The production chain starts by gasifying the heavy fuel oil with oxygen (partial oxidation) and steam at high pressure to produce syngas. The syngas is mainly composed of hydrogen and carbon monoxide as well as carbon soot (1–2% of the amount of used heavy oil) after gasification.

After cooling in a waste heat boiler, the syngas is scrubbed with water, and gas (CO, H<sub>2</sub>, CO<sub>2</sub> and H<sub>2</sub>S) and soot water are separated. The gas is led to an amine-wash for the recovery of sulphur and CO<sub>2</sub> and then further to the separation of CO and H<sub>2</sub>, which are used for the production of formic acid and hydrogen peroxide. Soot sludge from the centrifugation process is mixed with oil before combustion to improve the flowability of sludge. Fuel oil is also used as a supporting fuel at a CHP plant. [26] In Eastman's process, moist soot sludge (c. 95% w.b.) is formed approximately 20 t<sub>w,b./d.</sub>

### 2.2. Test rig

The test rig used in the experimental drying tests is illustrated in Fig. 2. The dryer used is a fixed-bed batch-type. Heat transfer to the bed material is convective in the dryer. Laboratory air is used as the drying gas. Pressurized air is heated in an electric heater to a higher temperature (43–113 °C) than the desired temperature of the drying air (40–100 °C). This pressurized air is then mixed with the indoor hall air (20–22 °C) in order to obtain the desired temperature for the drying air before the bed. By modifying the rotation speed of the fan, the air velocity before the bed is regulated to the desired level. The drying air flows through the mixture bed from top to bottom. The drying chamber lies on a scale, which saves the mass of the mixture sample on the computer after a given time interval (10 s used in the tests). When the mixture sample dries, its mass decreases. The drying curve is defined on the basis of the mass change. [27]

### 2.3. Initial values

Soot sludge with extremely high initial moisture content (95% w.b.) was dried in the experimental tests together with sawdust. Fig. 3a and b shows examples of soot sludge and sawdust samples, respectively. Ultimate analyses for soot sludge and sawdust are shown in Table 1. Soot sludge also includes small amounts of polycyclic aromatic hydrocarbons (PAH) [28].

Gasification circumstances and the quality of heavy oil have an influence on properties of soot formed in a gasification process under pressure. Soot is formed so quickly that it does not crystallize completely. Soot crystals form mainly spherical particles (known as primary particles) which have a diameter of 0.1–1 μm. The primary particles form bigger agglomerations with a diameter of 8–100 μm. [28]

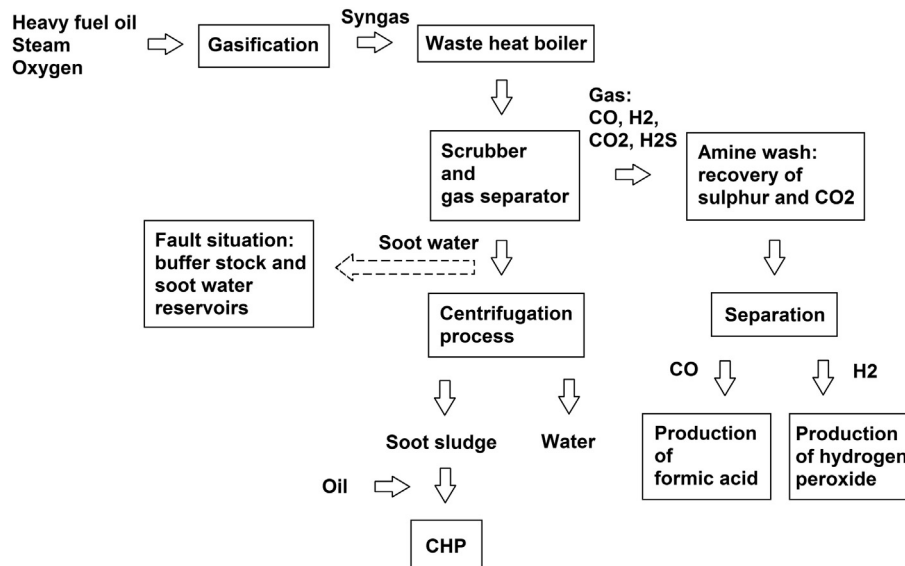


Fig. 1. Formation of soot sludge. Reproduced from the reference [26].

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