Drying characteristics of biosludge from pulp and paper mills

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ABSTRACT: Handling and disposal of biosludge has been a persistent problem for many pulp and paper mills primarily because of the material's high moisture content and poor dewatering and drying characteristics. Most biosludge is landfilled or incinerated, and some is used as a soil amendment. To incinerate biosludge, the mills mix it with primary sludge, mechanically dewater the mixture to 15%–30% solids, and burn it with hog fuel in biomass boilers. For biosludge to burn effectively, however, it must be dewatered or dried to at least 30%–35% solids. A systematic study was conducted to examine the drying characteristics of biosludge collected from pulp and paper mills at various temperatures using a thermogravimetric dryer. The drying behavior of the tested biosludges was similar, adding fillers such as wood fines and biomass boiler fly ash to biosludge up to 30 wt% did not significantly affect the drying rate, higher organic content might make biosludge more difficult to dry, and increasing drying temperature significantly increases the drying rate as expected.

Application: Mills can optimize their biosludge handling and disposal practices by understanding the material's drying characteristics.

Two types of sludge are produced from the wastewater treatment process of pulp mills: primary and secondary. Primary sludge is generated in the primary clarifier and is composed mainly of wood fibers. Secondary sludge (biosludge) is a colloidal suspension of small solids particles produced in the secondary clarifier. Biosludge is typically composed of microbes, undigested wood fibers, and organics, and it has a low solids content of about 2 wt% [1]. A portion of this sludge is recycled back into the wastewater treatment process to maintain the microbial community, and the remainder is disposed. The amount and composition of the sludge varies among mills and even among days at the same mill. Approximately 10–100 m^3 water is required per ton of paper produced [2], and with it 0.2–0.6 wet sludge per ton of pulp is produced [3]. Furthermore, the production of paper mill sludge is projected to increase between 48% and 86% from the amount produced in 2000 [4].

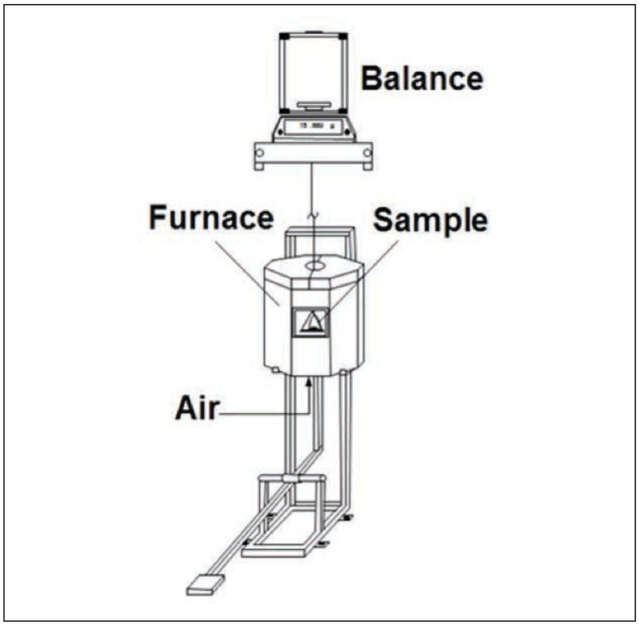
Most sludge is either landfilled or burned in biomass boilers, and some is used as a soil conditioner. Over the past decade, it has become more common for mills to burn the sludge as landfill space declines. To successfully burn wasted biological sludge, the solids content must be increased to at least 30%–35%. The challenge is to remove sufficient water so that the sludge can sustain combustion with no net thermal loss incurred by the boiler because of the high moisture content.

Drying is considered as a key step in the disposal of biosludge where waste heat from mill processes can be harnessed. Drier sludge is less costly to handle and easier to burn. Although much research has been done on the drying behavior of municipal sludge [5,6], not much work has been done on the drying behavior of pulp and paper mill biosludges. It is not known how biosludges from different mills compare or how biosludge contrasts with municipal sludge. It is also not clear whether drying can be facilitated with the addition of solids. The objective of this research was to investigate the fundamental drying characteristics of pulp and paper mill biosludge and methods to facilitate drying.

EXPERIMENTAL

The biosludge samples in this study were collected from a sulfite mill in Ontario and a kraft mill in Alberta, Canada. The samples were different in appearance and odor. The raw sulfite mill biosludge was dark brown (almost black) and contained 1.7% solids, whereas the kraft mill biosludge contained 3.3% solids, was brown and much thicker, and had a stronger odor. The samples were mixed with a 0.5 wt% solution of polymer (Zetag 8165, BASF; Ludwigshafen, Germany) to flocculate the solids, filtered by gravity, and pressed using a crown press to 13% solids. Assorted wood fines and biomass boiler fly ash, which were dried in an oven overnight at 105°C, were added to the dewatered sludge cakes in amounts of 10 wt%, 20 wt%, and 30 wt% to determine the effect of additives on the drying rate. The samples were molded into cylinders approximately 3.2 cm in diameter with heights varying from 1.2–3.0 cm depending of the amount of solids added. All tests were done using mechanically dewatered biosludge.

A thermogravimetric furnace (**Fig. 1**) was used to isothermally dry the samples. The apparatus can continuously measure and record the sample mass during drying, using a high-precision balance capable of measuring up to 200 g with a readability of 1 mg. Details of this apparatus have been previ-



1. Laboratory-scale thermogravimetric furnace.

ously described [7]. In this study, a weighing dish containing a sludge cake sample was hung from the balance directly above the opening of the furnace. The sample was then inserted into the furnace. By stepping on the pedal to lift the entire furnace assembly, the weighing dish and sample were situated in the center of the furnace chamber. Air was passed through the furnace from the bottom at a flow rate of 1 L/min to remove water vapor. The drying temperature varied from 110°C to 220°C at 1 atm. The changes in sample appearance and physical conditions were monitored using a high speed video camera focused through a viewing glass window at the side of the furnace.

	Sulfite Mill	Kraft Mill
Proximate (wt%), Dry Basis		
Volatiles	71.8	56.7
Fixed carbon	17.8	18.7
Ash	14.0	25.0
Higher heating value, MJ/kg	20.5	17.5
Ultimate (wt%)		
Carbon	44.7	39.8
Hydrogen	5.5	4.6
Nitrogen	6.0	3.2
Sulfur	2.1	2.4
Oxygen	31.3	27.0

I. Composition of biosludge from a sulfite mill and a kraft mill.

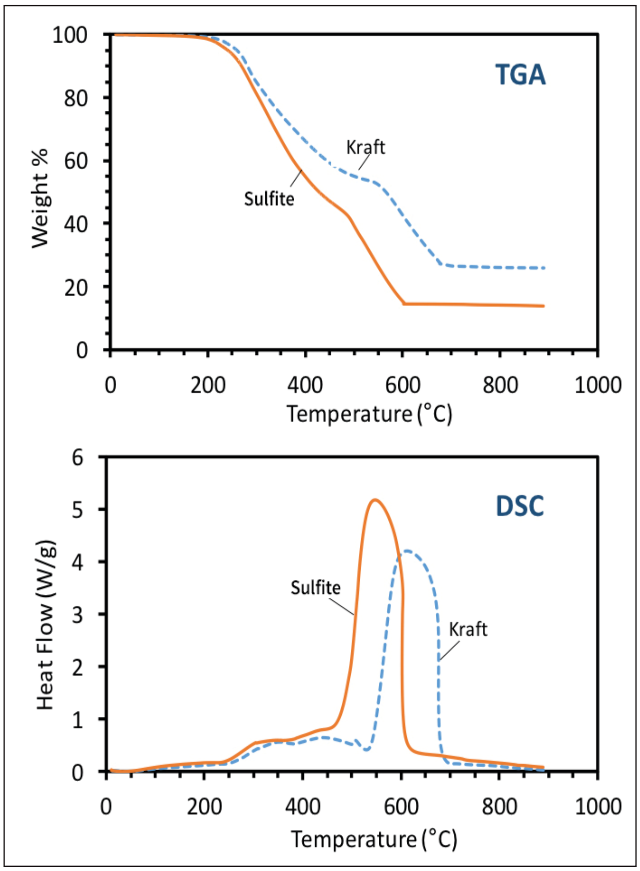
RESULTS AND DISCUSSION

Sludge analysis

The compositions of sulfite mill and kraft mill biosludges are shown in **Table I**. On a dry basis, the sulfite mill biosludge had a higher volatile content, higher heating value, and lower ash content than the kraft mill biosludge. After dewatering, a solids content of 13% could be achieved for both types.

A simultaneous thermogravimetric analysis/differential scanning calorimetry (TGA-DSC) analyzer was used to examine the combustion behavior of the sludge samples, which were first dried overnight at 105°C. The samples were heated in the analyzer from room temperature to 900°C at a heating rate of 20°C/min. The results are depicted in **Fig. 2**. Both samples have no change in weight up to about 180°C, at which point they began to lose weight. At 250°C, the heat flow rate and weight loss rate increase for both samples as volatiles are emitted and burned. This is followed by a continuing decrease in weight and a slight decrease and plateau in heat flow, indicating that the rate of heat absorbed equals the rate of heat released. This is a result of continued volatile burning occurring simultaneously with the heating of the solid particles.

At 460°C for sulfite mill biosludge sludge and 535°C for kraft mill biosludge, there is a large exothermic peak accompanied by a sharp reduction in sample weight as the solids



2. Thermal behavior of kraft and sulfite mill biosludges: thermogravimetric analysis (TGA, upper), and differential scanning calorimetry (DSC, lower).

burn (char burning). The amount of fixed carbon was similar for both biosludge types. The total heat flow during combustion was determined by calculating the area under the exothermic peaks. The total heat flow was found to be 1324 kJ/kg for sulfite mill biosludge and 1242 kJ/kg for kraft mill biosludge. This difference is most likely a result of the higher heating value of the sulfite mill biosludge, which contains more organic matter and less ash (Table I). After the char burning, the heat flow decreases for both samples and no changes in sample weight are observed. The plateau of the weight loss curve at high temperatures indicates the combustion is complete. Ash contents of the kraft mill biosludge and sulfite mill biosludge were 25 wt% and 14 wt%, respectively.

Drying rates

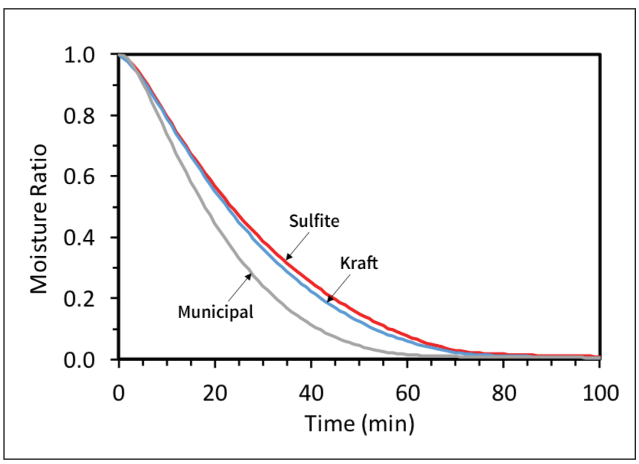
Upon drying, sludge samples lose weight as their moisture contents decrease. In this study, the change in sample weight during drying is expressed as moisture ratio to account for the difference in initial weight, as in Eq. (1):

$$\text{Moisture Ratio} = \frac{M - M_{Final}}{M_{Initial} - M_{Final}} \quad (1)$$

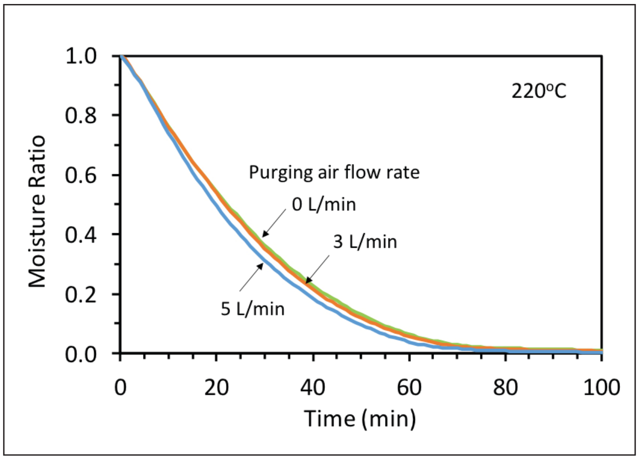
where M is the moisture content at time t (g H_2O/g dry solids), $M_{Initial}$ is the initial moisture content (g H_2O/g dry solids), and M_{Final} is the final moisture content (g H_2O/g dry solids).

Figure 3 indicates the drying curves for sulfite and kraft mill biosludges, which were mechanically dewatered to 13% solids, and municipal sludge, which could only be mechanically dewatered to 10% solids. The samples were dried at 220°C, with air flow rate of 1 L/min. The results indicate that the drying trends for sulfite and kraft mill biosludges are nearly identical and are similar to municipal sludge. The difference in drying rates can be attributed to the higher water content in the municipal sludge before drying.

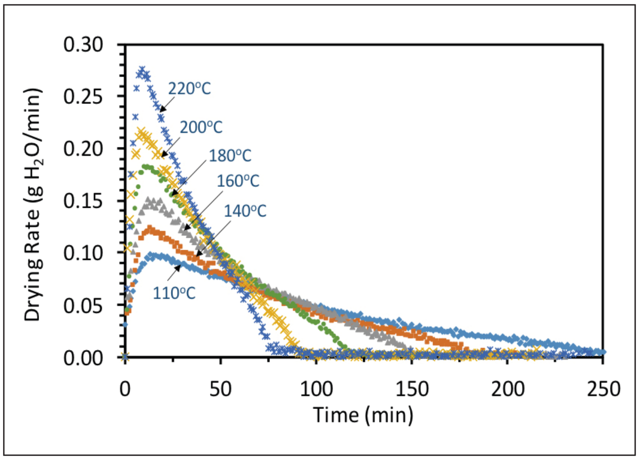
Figure 4 shows the effect of air flow rate on the drying of sulfite mill biosludge at 220°C. As expected, the biosludge drying rate increases as the air flow rate increases. This is because the greater velocity is more effective at removing the vaporized water from the sample, thus maintaining a concentration gradient.



3. Drying curve at 220°C and 1 L/min purging air flow rate for mechanically dewatered sulfite and kraft mill biosludges and municipal biosludge.



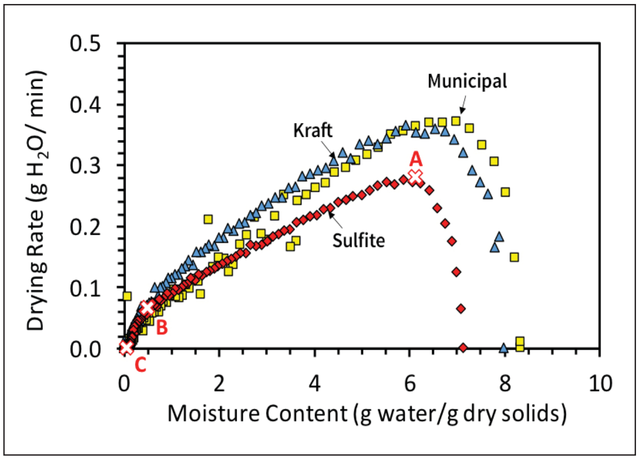
4. Effect of air flow rate on the drying curve of sulfite mill biosludge at 220°C.



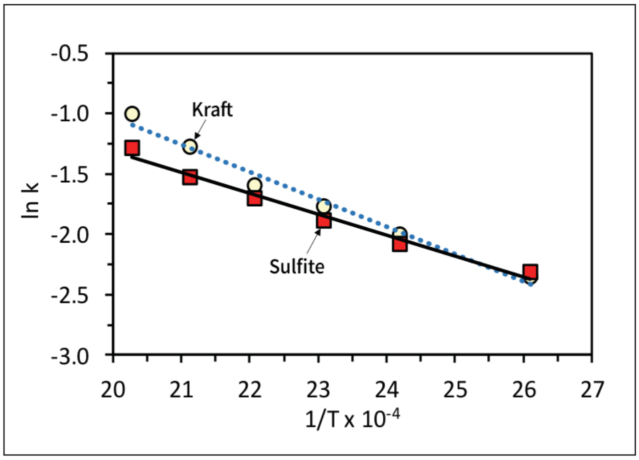
5. Drying rate curves of sulfite mill biosludge at temperatures between 110°C and 220°C and 1 L/min purging air flow rate.

The drying rate of sulfite mill biosludge dried at various temperatures from 110°C to 220°C is indicated in **Fig. 5**. The drying rate was determined by calculating the moisture content on a dry solids basis from the experimental data and then determined as the difference between two moisture contents in 1-min time intervals.

The drying rate at 220°C reached a maximum of 0.28 g H_2O/g dry solids at 9 min. During this time, the sample material undergoes heating as most of the thermal energy is absorbed by the water on the surface. Once enough thermal energy has been absorbed, the water on the sample surface will vaporize. The mass transfer rate will increase at the point where the initial vaporization briefly occurs. Once the maximum drying rate is reached, there is a sudden decrease in drying rate as the moisture content decreases. This is because the outer layer of the material has dried, forming a crust, and the water vapor must migrate from the sample interior to the sur-



6. Krischer curves showing the first and second falling rate periods for sulfite mill and kraft mill biosludges and municipal sludge at 220°C and 1 L/min purging air flow rate.



7. Arrhenius plot of sulfite and kraft mill biosludge at various temperatures with 1 L/min purging air flow rate.

face to escape. Because the distance the water must travel increases throughout the drying process, the rate is controlled by the diffusion of water toward the sample surface. Eventually, the drying rate reaches zero at 75 min because the moisture content is approximately zero and the concentration gradient between the sample and the hot air no longer exists.

Figure 6 shows the drying rates at 220°C of sulfite and kraft biosludges and municipal sludge as a function of moisture content. These curves, known as Krischer curves [8], suggest that the sulfite biosludge drying rate is lower than the rate for the kraft mill biosludge. This might be a result of the higher organic content in the biosludge, as suggested by Tuncal [9].

As suggested in previous studies [6,10], drying occurs primarily during two falling rate periods at two critical moisture contents. For the sulfite biosludge, for example, the first period is from A to B when the moisture content decreases from 5.8 to 0.3 g H_2O/g dry solids. The second period is from B to C when the moisture content decreases further from about 0.3 to 0 g H_2O/g dry solids. **Table II** summarizes the critical periods for all three types of sludge examined in this study.

The second critical moisture content of the pulp mill biosludges agrees with values reported in literature (0.4–0.8 g H_2O/g dry solids) [11]. However, the experimental value for municipal sludge is outside of this range and falls into the gelatin and gel category (>80 g H_2O/g dry solids). The critical moisture content is not solely a material property, but also depends on the drying conditions such as the temperature, drying rate, and humidity.

Critical Moisture Content	Sulfite Mill, g H_2O/g dry solids	Kraft Mill, g H_2O/g dry solids	Municipal, g H_2O/g dry solids
First (Point A)	5.8	6.5	6.7
Second (Point B)	0.4	0.4	0.4

II. Critical moisture contents of the tested biosludge.

The rate constant for the drying process can be obtained by considering drying to be first order as follows in Eq. (2):

$$\frac{dM}{dt} = M_o e^{-kt} \qquad (2)$$

where M is the moisture content at time t (g H_2O/g dry solids), M_o is the initial moisture content (g H_2O/g dry solids), t is time (min), and k is the rate constant.

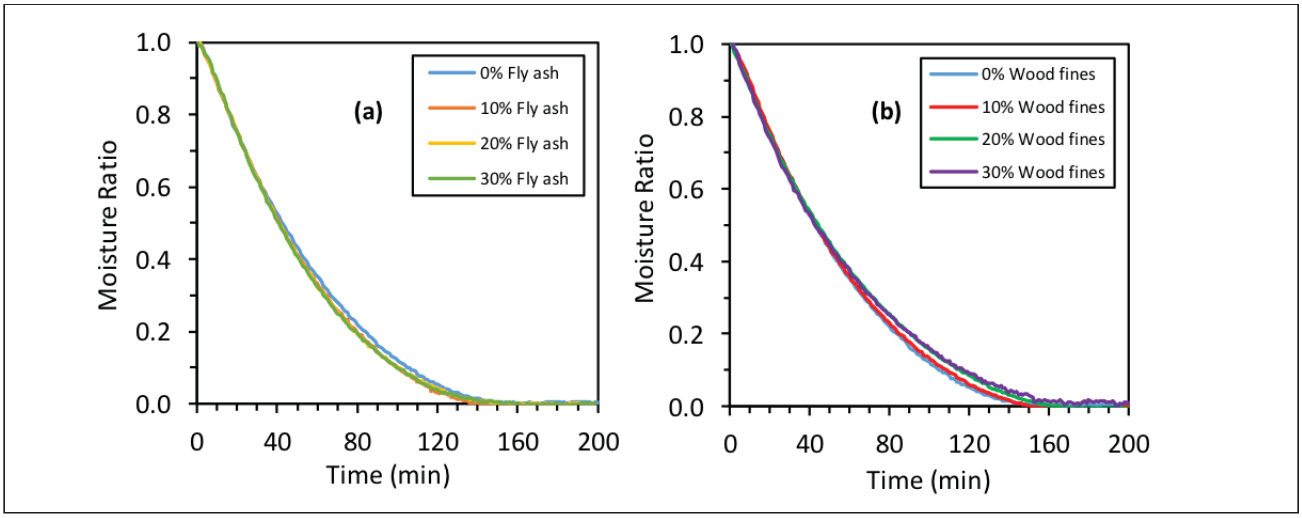
To obtain the drying rate constant for the sulfite biosludge, the natural logarithm of the maximum drying rate value for each temperature shown in Fig. 5 was plotted versus 1/T, where T is absolute temperature in °K. This Arrhenius plot was developed for both sulfite and kraft mill biosludges (**Fig. 7**).

Drying temperature clearly has a significant effect on the drying rate. The slope of this curve represents the activation energy, which is the minimum energy required for the process to proceed. The activation energy was determined to be 1742 kJ/mol for the sulfite mill biosludge and 2268 kJ/mol for the kraft mill biosludge.

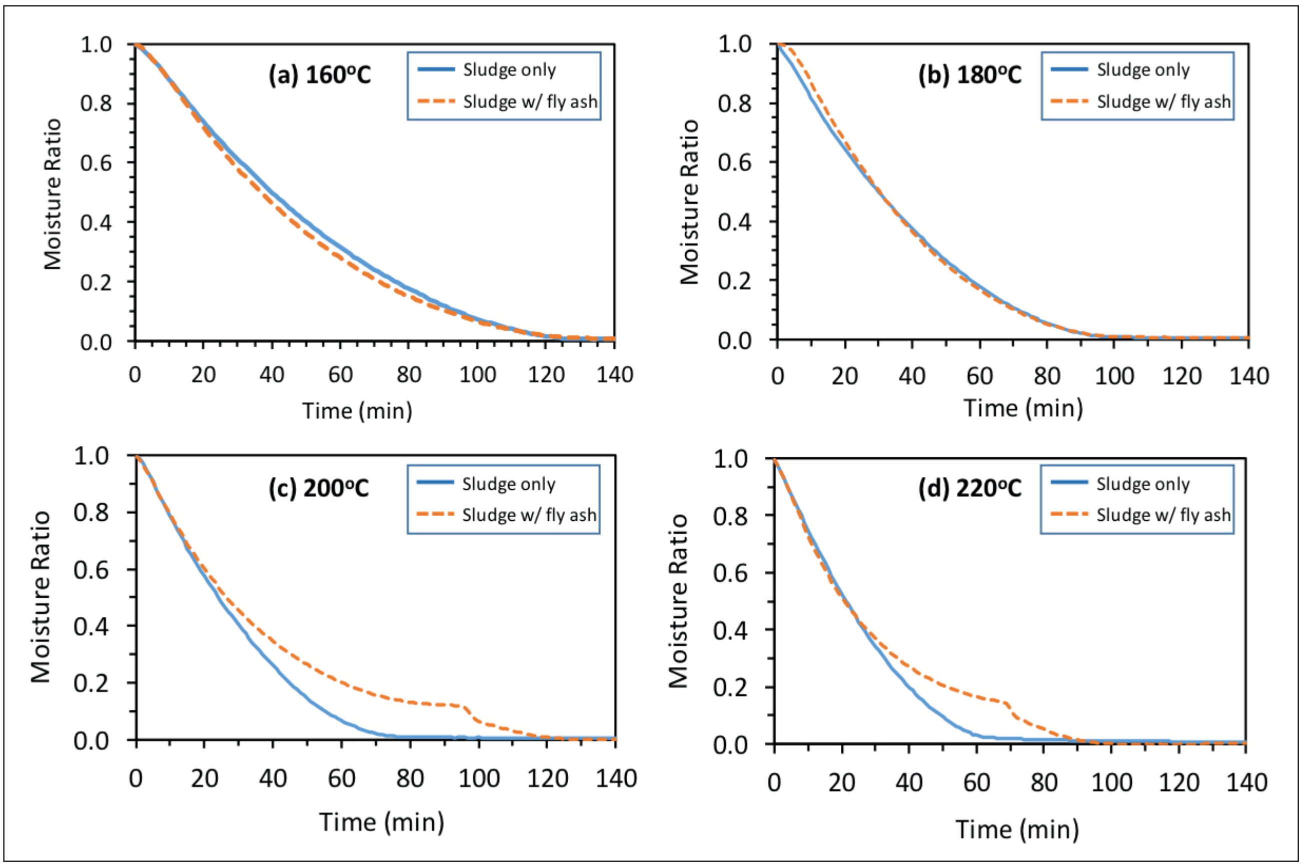
Effects of fly ash and wood fines addition

Fly ash and wood fines, common solid waste products at pulp mills, were added to the dewatered sludge mixture to test whether their addition can improve biosludge drying. These materials were oven dried and added to 10 g of biosludge in amounts of 10 wt%, 20 wt%, and 30 wt%. These tests were conducted at 140°C because higher temperatures would cause volatile burning of the wood fines.

Figure 8 shows the effect of fly ash and wood fines on the drying of biosludge. Fig. 8a indicates that the addition of fly ash has no effect. This could be caused by the low temperature (140°C), which did not allow the fly ash particles to reach high enough temperatures to alter the drying rate of biosludge. Fig. 8b indicates that the addition of wood fines adversely affects the drying of biosludge. This might be a result of the moisture-free wood fines absorbing moisture from



8. Effects of (a) fly ash and (b) wood fines on biosludge drying at 140°C with 1 L/min purging air flow rate.

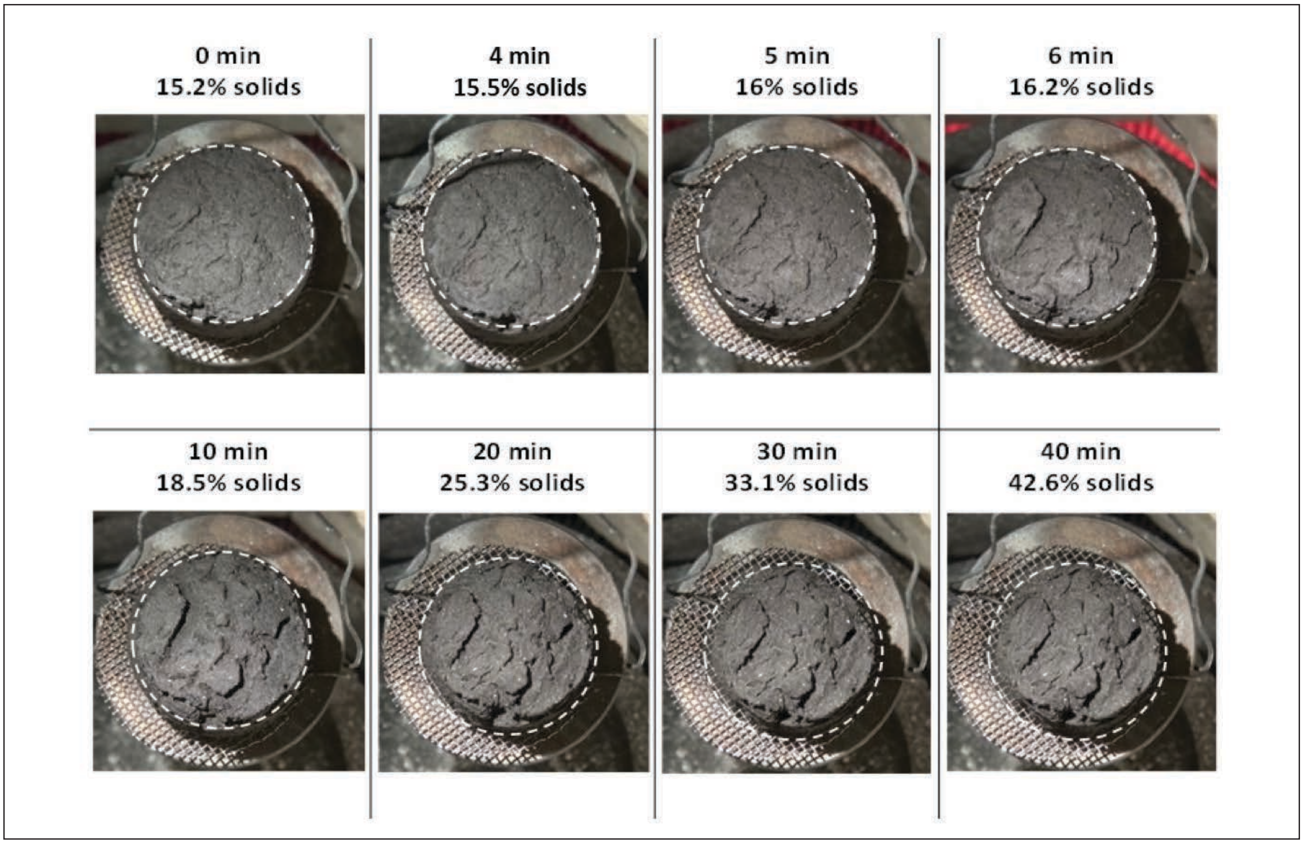


9. Effect of temperature on drying of biosludge mixed with 20 wt% fly ash with 1 L/min purging air flow rate.

the biosludge, as well as slower moisture transfer out of the wood fines than moisture transfer out of the biosludge. This agrees with findings on municipal sludge that suggest an increase in the organic content of the sludge adversely affects the drying potential [9].

From the results presented in Fig. 8, drying the sample at a low temperature was likely the reason the fly ash did not affect the drying behavior of biosludge. To test whether the drying temperature has an effect, 10 g of biosludge with 20 wt% fly ash were dried at various temperatures in the 140°C–220°C range. The results (**Fig. 9**) confirm that the fly ash does not have a significant impact on the drying behavior of biosludge.

At higher drying temperatures, 200°C and 220°C, sudden drops in the moisture content occur at 98 min and 70 min, respectively. This is caused by devolatilization of the sample,



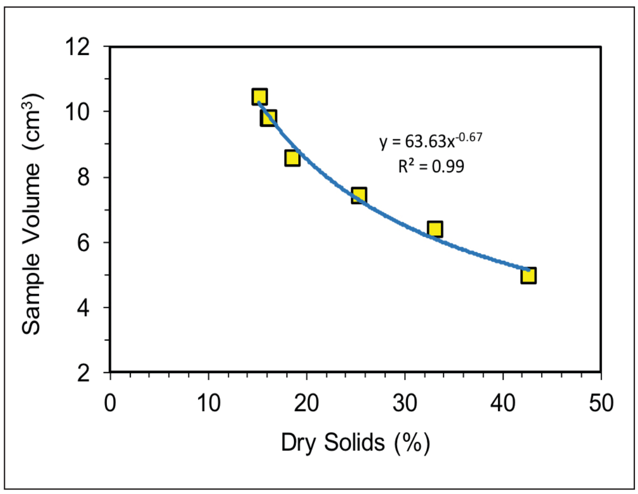
10. Crack development and shrinkage of the sulfite mill sludge during first 40 min of drying at 220°C with 1 L/min purging air flow rate.

where smoke was emitted. Devolatilization occurs initially at the surface and is reflected by the sudden weight loss indicated in Fig. 9c and Fig. 9d. Once the surface volatiles have escaped, the rate decreases as devolatilization occurs in the interior of the sample. In addition, the drying rates of the samples with fly ash at 200°C and 220°C were slower than the samples without fly ash, possibly because of the fly ash coating the sludge particles and preventing vaporization.

Sludge shrinkage during drying

The samples were prepared with dewatered sulfite mill biosludge, which had been molded into a cylinder using a cutout. During drying, the sludge passes through several physical stages and undergoes extensive cracking and shrinkage. **Figure 10** shows the shrinkage and cracking progression of biosludge during the first 40 min of drying at 220°C. After this, crack formation and shrinkage are difficult to notice visually.

Initially, the dewatered sludge is deformable with a soft pastelike consistency and shear thinning behavior. This was also noticed by Markis et al. in their study of biosludge rheology [12]. The drying behavior was comparable for all tests and sludge types. During the initial 5 min, a thin porous crust developed and the surface changed from slippery and shiny to rough and dull. As the moisture content decreases, the sludge cake shrinks and cracks form. Once the surface is no longer wet the thin porous crust develops into a hard impermeable layer, which continues to expand toward the center of the sample. The interior remains wet and malleable.



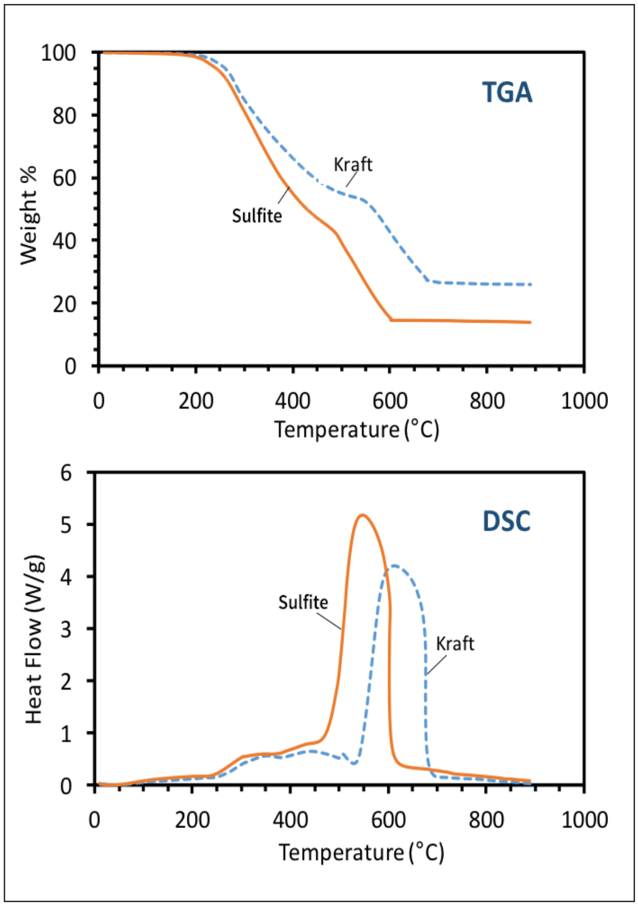
11. Change in volume of the sulfite mill sludge cake during the first 40 min of drying at 220°C with 1 L/min purging air flow rate.

The top surface of the sample (the portion visible in Fig. 10) shrunk faster and had more crust growth than the bottom surface. Cracking accelerates crust growth and drying because it exposes more surface area to the hot air. The edges dry faster because they have greater contact with the air flowing past. Significant shrinkage and cracking are observed up to 40–50 min at 55%–50% moisture, with only minor changes observed after this. These changes are random and unpredictable and can alter heat and mass transfer. Cracking is caused by nonuniform thermal stresses and an imbalance of surface capillary tension caused by shrinking [10,12]; it often begins in areas with superficial blemishes or low water content. Shrinkage occurs because of increasing particle-particle attraction as the water content decreases. Spalling, which results from tensile stresses and causes the sludge to lift, can also be observed. After approximately 100 min, the second drying period occurs and no shrinking is observed. As the sludge sample dries it became rigid, but brittle. The dried sample has many cracks and crevices, which can accelerate the drying process depending on their location and depth.

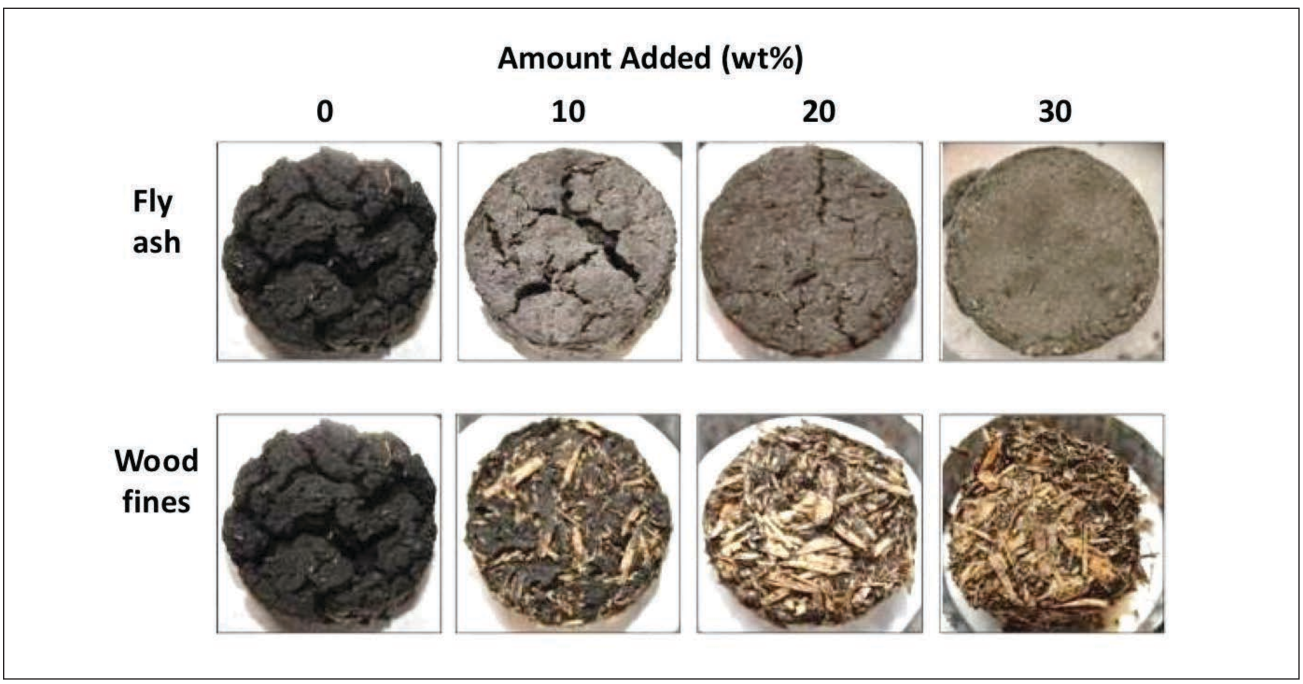
Figure 11 is based on the change in sample dimension shown in Fig. 10. The sample volume over the first 40 min of drying was estimated and plotted against the dry solids content of the sulfite mill sludge cake. The results indicate a rapid decrease in sample volume during the early stages of the drying process, when the sample is at higher moisture content, and corresponds to a higher drying rate. As the moisture content decreases, the rate of change of the sample volume also decreases because less water fills the voids within the sludge cake. This continues until the voids within the sample cake are empty.

Effect of fly ash and wood fines on shrinkage and cracking

We hypothesized that the addition of solids would support the solid sludge structure, increase available surface area, and reduce shrinkage. **Figure 12** shows the percentage de-



12. Effect of adding fly ash and wood fines on sludge shrinkage during drying at 220°C with 1 L/min purging air flow rate.



13. Comparison of crack formation between biosludge and biosludge mixed with various amounts of fly ash and wood fines. Samples were dried at 220°C with 1 L/min purging air flow rate.

crease in volume between samples of sludge with and without solids added. As expected, the sample shrinkage decreases as the amount of added solids increases. The samples mixed with wood fines shrink less than those with fly ash. The wood fines are much lighter than the fly ash, so for a given mass more wood fine particles would be present than fly ash. In addition, the wood fines have a larger volume and thus interact more with one another to form a stronger structure that is more resistant to shrinkage.

As indicated in **Fig. 13**, the addition of solids reduces cracking of the sample during drying. The cracking is further reduced as more solids are added to the sludge sample; this reduction is observed for both fly ash and wood fine additions. More cracking occurs in the samples with fly ash than those with wood fines, which are larger and more rigid and thus provide more support for the sludge.

CONCLUSIONS

A systematic study was performed to investigate the drying behavior of biosludges from a sulfite mill and a kraft mill. The results show that the drying behavior of sulfite and kraft mill biosludges is nearly identical, and the drying behavior is similar to that of municipal sludge. The drying rate of biosludge with low solids content increases initially and then decreases mostly linearly with an increase in solids content. Increasing the drying temperature increases the drying rate as expected, and the addition of fly ash and wood fines does not facilitate drying but reduces shrinkage and cracking. Fly ash facilitates devolatilization of sludge at temperatures greater than 200°C, and higher organic content may decrease the drying potential of biosludge. These findings provide a better understanding of how pulp and paper mill biosludge behaves as it is dewatered and dried. **TJ**

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ABOUT THE AUTHORS

For many mills, biosludge disposal is a problem that will only worsen as landfill space declines. We wanted to explore the fundamentals of biosludge drying and provide insights into making biosludge a more attractive fuel for combustion in biomass boilers

Previous research focused primarily on mechanically dewatering biosludge. Although drying is used at some mills as part of the disposal process, a fundamental understanding of the drying process is lacking. This research aims to characterize this process and provide insight into improving biosludge disposal.

One of the more challenging aspects of the research was the length of some of the experiments. We addressed this by preparing a detailed experimental plan and extending working hours as needed. One of the researchers was a new graduate student with limited knowledge of the pulp and paper industry and waste water systems. This research served as an introduction to both subjects that also provided a new appreciation for the industry.

It was interesting to see the similarities between the drying curves of biosludge from pulp and paper mills and sludge from municipal waste water treatment systems. The sudden decarbonization of biosludge when mixed with fly ash and dried at higher temperatures was also surprising.

Hovey

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Tran

For mills, this research provides additional insight into biosludge disposal, in particular the use of biosludge as a fuel in biomass boilers. Possible next steps would be to dry biosludge in a laboratory-scale dryer to better simulate industrial drying processes and to test biosludge from different mill types to observe any differences.

Hovey is a graduate student; Allen is professor and chair; and Tran is Frank Dottori Professor of Pulp and Paper and director of the Pulp and Paper Centre, Department of Chemical Engineering and Applied Chemistry, at the University of Toronto, Toronto, ON, Canada. Email Tran at honghi.tran@utoronto.ca.

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