

Pilot scale black liquor concentration using pressure driven membrane separation

SAM RAE, ELLA V. RICHARDS, MAX KLEIMAN-LYNCH, BRENT D. KELLER,
AND BRANDON I. MACDONALD

ABSTRACT: Black liquor concentration using pressure driven membrane separation has long been proposed as a means of achieving energy savings and breaking production bottlenecks. To date, limitations in membrane performance and stability under black liquor process conditions have prevented those promises from being realized outside of tightly controlled laboratory settings.

In this work, we describe the first successful pilot scale field deployments of a membrane system for black liquor concentration. Using a purpose-built system and commercial sized, spiral wound graphene oxide membrane elements, we have logged nearly 6000 h of runtime across deployments to multiple mill sites. We demonstrate concentration of black liquor from 14% to >20% total solids, while generating permeate water comparable in quality to that of evaporator condensate and an 81% reduction in energy consumption relative to evaporation. At a commercial scale, these results translate to \$2 M/year in net energy savings for a typical mill, as well as an opportunity to support production increases or mill expansions. These results represent a significant leap forward in the ability of membrane systems to deliver substantial value via black liquor concentration.

Application: In this work, the authors describe the first successful pilot demonstration of black liquor concentration using a membrane system. This technical demonstration paves the way for commercial sized systems achieving substantial cost and energy savings.

Black liquor (BL) concentration using multi-effect evaporation accounts for 30% of overall kraft mill energy consumption [1]. Augmenting this process through the incorporation of membrane driven separations provides an opportunity to reduce the energy costs associated with BL concentration while simultaneously offering a route to increased pulp production in evaporator limited mills. Pressure driven membrane separations such as reverse osmosis are up to 90% more energy efficient than thermal evaporation, as they do not require a liquid-to-vapor phase transition [2]. As a practical example, using membrane systems to increase black liquor solids from 10%–15% to 20%–30% prior to evaporation can save an estimated 23 trillion Btu/year industry-wide, resulting in millions of dollars per year in costs savings for the average mill [3,4].

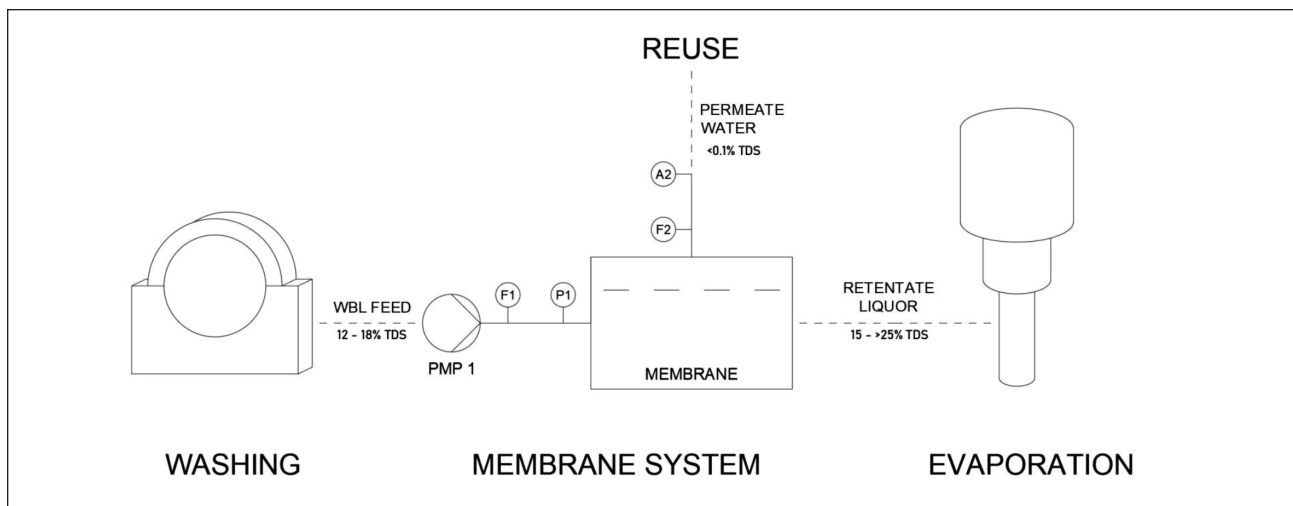
The concept of using membranes for BL concentration dates back several decades, with a variety of polymeric and ceramic membrane materials having been evaluated [5–7]. However, these efforts have universally failed to reach commercial scale adoption due to the unique challenges provided by BL processing. Any materials used in such a process must be able to withstand operating conditions of 70°C–90°C and pH 12–13 without degradation. The high input solids concentration (12%–18%) can lead to irreversible fouling of the membrane surface, diminishing separation performance. Finally, BL contains a diverse range of chemical components, including lignin, organic acids, and both monovalent and divalent inorganic salts [8]. Most previous studies have focused exclu-

sively on lignin recovery from black liquor rather than concentration of all organic and inorganic constituents. To produce permeate water quality comparable to that of evaporator condensate, any membrane system must be able to efficiently separate all these components.

Membranes based on graphene oxide (GO) have enabled an expansion of the operating conditions accessible to pressure driven separations. Graphene oxide is a two-dimensional material formed through the oxidation of graphite [9,10]. It is exceptionally robust, able to readily withstand high temperature, high pH environments without degradation [11,12]. Chemical modification of the oxygen containing functional groups on the GO surface allows for a broadly tunable set of membrane performance properties. For example, by adjusting the hydrophilicity or hydrophobicity of the membrane surface, fouling due to high solids concentration can be mitigated [13,14]. In addition, tuning the surface charge and spacing between adjacent GO layers allows the membrane to be optimized for performance with specific process feed streams. By leveraging these properties, promising results for BL concentration using GO membranes have been achieved by us and others using laboratory-scale, flat sheet samples [15–17].

To be a commercially viable process, BL membrane concentration must be as reliable as the incumbent technology of evaporation. To date, no BL membrane systems, GO based or otherwise, have been able to achieve this rigorous standard in continuously operating, field-based trials of ei-

RECOVERY CYCLE



1. A simplified process flow diagram for the black liquor concentrating membrane system. The weak black liquor (WBL) feed from the washing step is fed to the membrane system and separated into a clean permeate stream and concentrated retentate black liquor stream. Retentate is fed directly to the evaporation stage while permeate is reused as process water in the mill (PMP 1 = WBL pump).

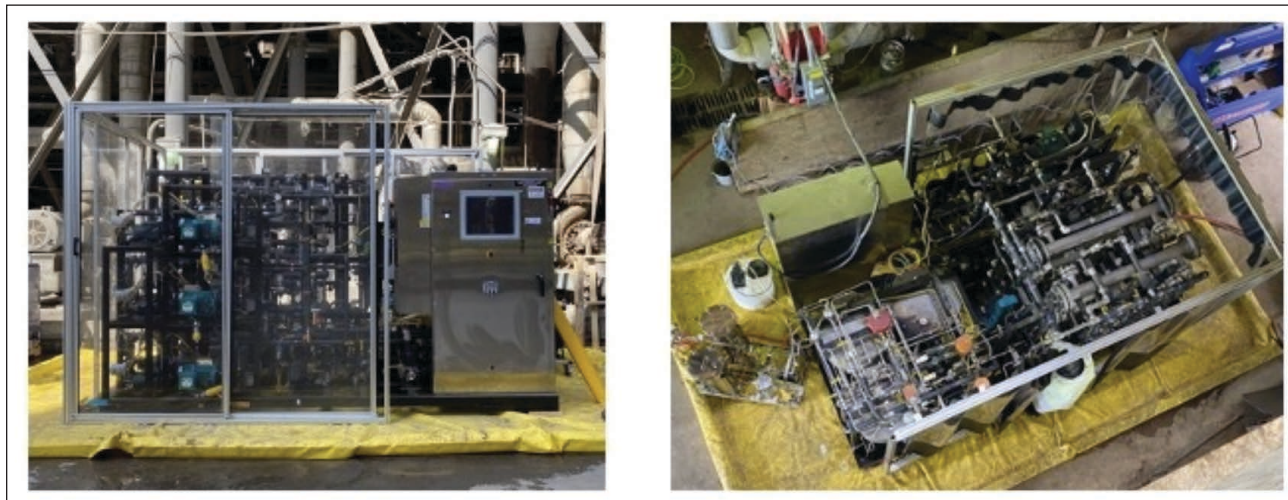
ther lab or pilot scale equipment. Here, we describe results from several successful pilot scale field deployments using spiral wound GO membrane modules, the same form factor used in commercial membrane operations globally. With nearly 6000 h of operational runtime across three different mills, including a single deployment of >2900 h, we demonstrate consistent membrane based black liquor concentration at the multiple gallons per minute scale. This work successfully provides technical validation of advanced membrane technology for black liquor processing, proving readiness for full commercialization of this technology.

METHODOLOGY

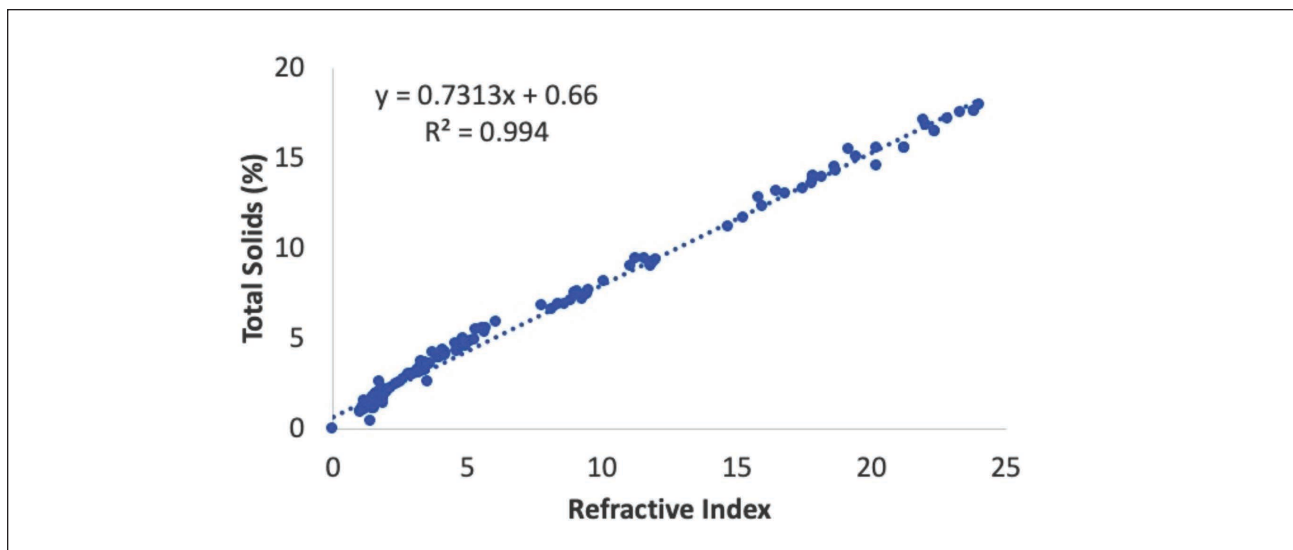
System overview

The pilot scale membrane system is designed to utilize commercial scale, spiral wound GO membrane modules to extract clean permeate water from black liquor (BL). The sys-

tem consists of twelve individual membrane housings and can operate in single stage, multi-stage, or multi-pass configurations. The system draws 2–3 gal/min of BL, typically between 12% to 18% total dissolved solids (TDS). This feed stream is drawn from directly before the multi-effect evaporator, as it would be in a full-scale commercial system. Using pressure driven filtration, the weak black liquor (WBL) feed is separated into permeate and retentate streams. Permeate is the clean, reusable water stream between <0.01 and 0.1% TDS, analogous to evaporator condensate. Retentate is the concentrated black liquor outlet stream, between 15% and >25% TDS. In a commercial system, retentate will be fed directly to the multi-effect evaporator, while permeate water reuse cases such as brownstock washing will be mill-dependent. A simplified process flow diagram for the pilot system is shown in **Fig. 1**. Photographs of the system are included in **Fig 2**.



2. Optical photographs of the black liquor concentration membrane system at a deployment site.



3. An example of a calibration curve used to determine black liquor solids from refractive index.

The pilot system is designed to collect all data required to design a full-scale system. To optimize system performance, operating pressure (P_1), feed flow rate (F_1), and other key metrics are controlled using variable frequency drives, solenoid control valves, and automatic control sequences. Permeate flow rate (F_2) and quality (A_2) are monitored with inline telemetry, with data recorded every 15 s and uploaded to a data historian for real-time performance monitoring. For more detailed offline analysis, the system is equipped with manual sample ports for collection of permeate from each individual membrane vessel. Membrane cleaning is performed using a clean-in-place (CIP) process using a dedicated tank, pump, and control valve. In the CIP process, the membrane(s) to be cleaned are isolated from the WBL flow path, then cleaned using a low pressure and high flow rate feed from the CIP tank that generates high turbulence across the membrane surface, removing or redissolving surface foulants. Once finished, the cleaned membrane(s) are then returned to service. In the pilot system, due to design constraints, the contents of the CIP tank are sent to the sewer. In a commercial system, the CIP fluid will be returned to the process stream.

Performance quantification

Feed, retentate, and permeate quality is measured inline using conductivity and refractive index probes. Refractive index values are converted to total solids using calibration curves developed offline. As BL composition varies between mills, individual calibration curves are built for each test site. Calibration curves are built by collecting samples and directly measuring solids content by taking aliquots of known volume and evaporating to dryness. A sample calibration curve is presented in **Fig. 3**.

In addition to inline and total solids measurements, samples of permeate are sent to a third-party laboratory for independent characterization. Black liquor solids were

measured according to TAPPI Standard Test Method T 650 “Solids content of black liquor,” and chemical oxygen demand (COD) was measured following digestion under acidic conditions. In addition, methanol concentration was measured by gas chromatography flame ionization detection (GC-FID), and sulfate, carbonate, and elemental concentrations of carbon, sodium, potassium, and silicon were measured by inductively coupled plasma optical emission spectroscopy (ICP-OES). The ICP-OES method was also used to measure potassium and silicon.

Power consumption is quantified using inline telemetry data. In a membrane system, pumps consume the vast majority of the power. A pump’s power consumption can be calculated from its applied pressure differential, flow rate, and efficiency. The power consumption of PMP 1 in Fig. 1 can be calculated with Eq. 1:

$$Power = \frac{Q_1 \cdot P_1}{\epsilon_1} \quad (1)$$

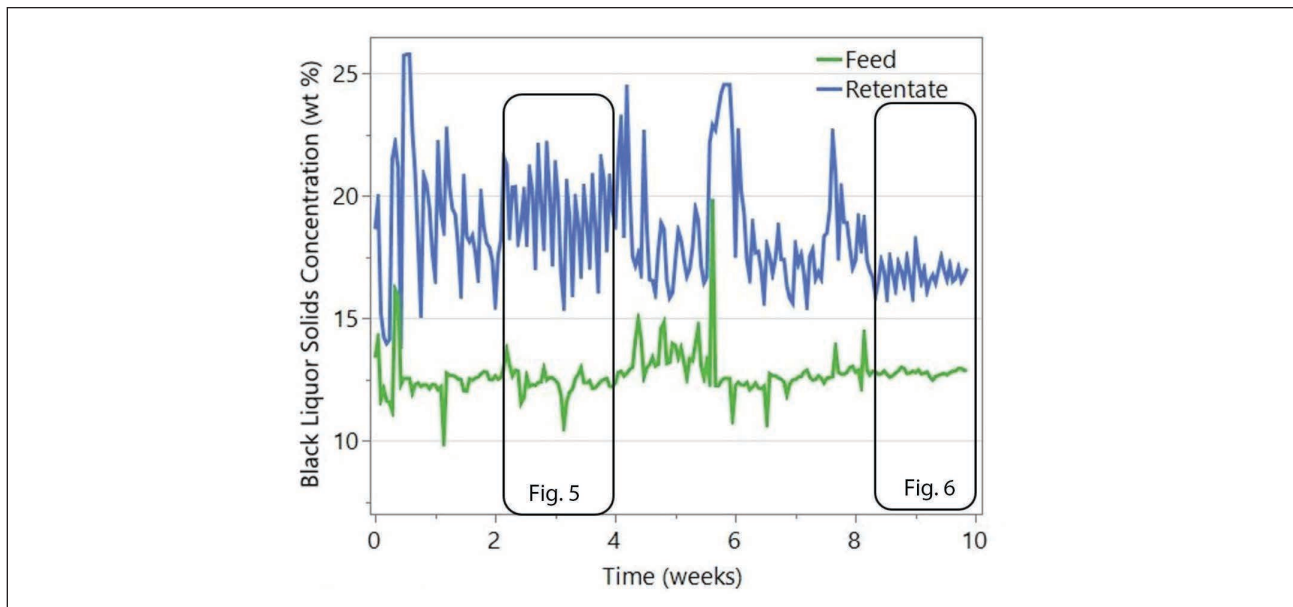
where Q_1 represents flow, P_1 represents pressure, and ϵ_1 represents pump efficiency. Using Eq. 1, the power consumption of all pumps in the process can be calculated.

Steam savings and BL throughput are calculated using an Excel-based model of a 6-effect evaporator system, as described previously [4]. Gross cost savings enabled through carbon pricing offsets are calculated using Eq. 2:

$$Annual\ savings = \frac{SS \cdot CP \cdot EF \cdot ED}{\eta} \quad (2)$$

where SS is the steam savings enabled by a membrane system, CP is the carbon price, EF is the emission factor of the fuel source, ED is the energy density of steam, and η is the boiler efficiency. Any carbon tax or credit savings should be calculated net of the electrical requirement of the mem-

RECOVERY CYCLE



4. Feed and retentate total solids over a span of 10 weeks continuous operation, as measured using inline telemetry. The highlighted subsections display the time ranges used in Fig. 5 and Fig. 6. The fluctuations in retentate solids are due to CIP events and would be diminished as the number of modules in the system was increased in translating to commercial scale. The pilot system clearly demonstrated concentration of BL by at least 4% solids for the entire duration.

brane system and net of any reduced electricity generation. Due to regional variations in electrical grid emissions (including up to 95% carbon free grids in pulp producing areas with abundant hydro or nuclear power), we present the gross savings here.

RESULTS AND DISCUSSION

To augment or replace an evaporator, a membrane BL concentration system must increase the total solids of the feed liquor by removing sufficiently clean water and do so with stable, reliable performance. We thus focus on three key topics: the solids difference between feed and retentate streams, permeate water quality, and system stability as measured by consistent permeate flow rates. Success criteria for each metric were determined in advance through consultation with mills, based on techno-economic analysis using mill production data to determine requirements for economic viability of a scaled-up commercial unit. Across deployments to three kraft mills and nearly 6000 h of cumulative runtime, we have successfully met or exceeded all target requirements, as described in detail in the following sections.

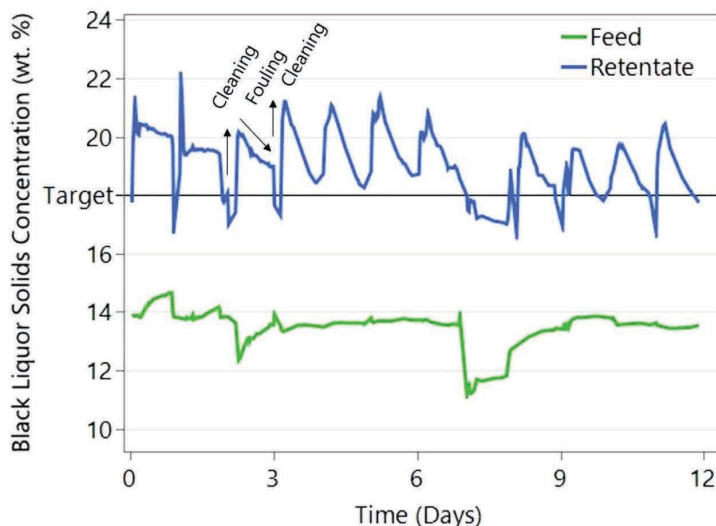
Black liquor concentration

For a fiber line producing a steady flow rate and concentration of BL feed, the difference between the feed and retentate black liquor solids (BLS) determines the reduction in evaporator steam demand and the subsequent energy savings from implementing a membrane system. An increase in BLS of only 2% can result in an up to 15% reduction in steam demand [4]. Shown in **Fig. 4** are feed and retentate

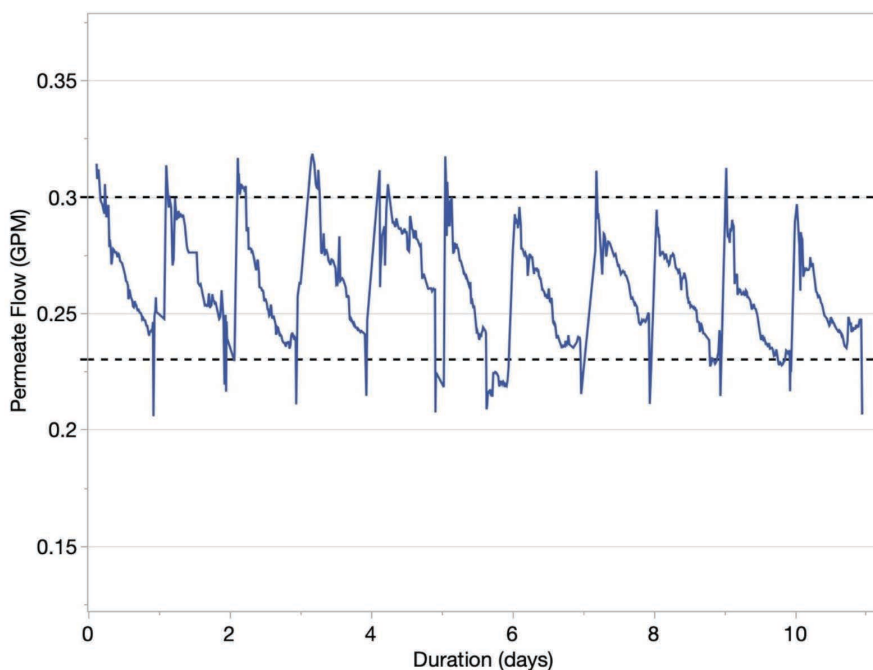
solids from our GO membrane system across ten weeks of continuous operation, where an average increase of 5% in solids was achieved. The ability to effectively provide BL concentration over an extended period of time despite normal process variations demonstrates the robustness of the membrane system in a real-world environment.

Figure 5 shows a 12-day period where the system was configured to target a retentate solids threshold of >18%. Tuning of retentate solids can be achieved through a combination of process conditions, such as applied pressure and flow rates, as well as the number of membranes in operation, with more membranes resulting in greater BL concentration. Over the course of this test, a consistent retentate solids output of 18%–22% was achieved—an average increase in BLS of 5% and a peak increase of 8%. The brief excursions below the target threshold occurred during membrane cleaning cycles, discussed in detail below, and during one overnight period when inlet solids dropped sharply from 14% to <12%. The pilot system does not have any process controls to respond to changes in feed solids, and a larger system could easily adjust process conditions to maintain a steadier retentate solids in such an event.

The periodic oscillations recorded in retentate solids are driven primarily by membrane cleanings and the pilot scale of the system. As membranes foul, retentate solids decrease due to a decrease in permeate flow rate. Following cleaning, the flow rate is restored, leading to an increase in retentate solids. In a larger system, membrane cleanings can be efficiently staggered to minimize output fluctuations. In addition, advanced process controls can be used to adjust system pressure and feed rate to maintain a fixed retentate



5. Feed and retentate solids levels across 12 days of testing. Cleaning cycles are employed to maintain retentate solids above the target threshold.



6. Permeate flow rate for the highlighted region within Fig. 4. In this study, each module was cleaned once per day. Permeate flow remains inside the same operating window (dashed lines) for the duration of the test, indicating stable membrane performance.

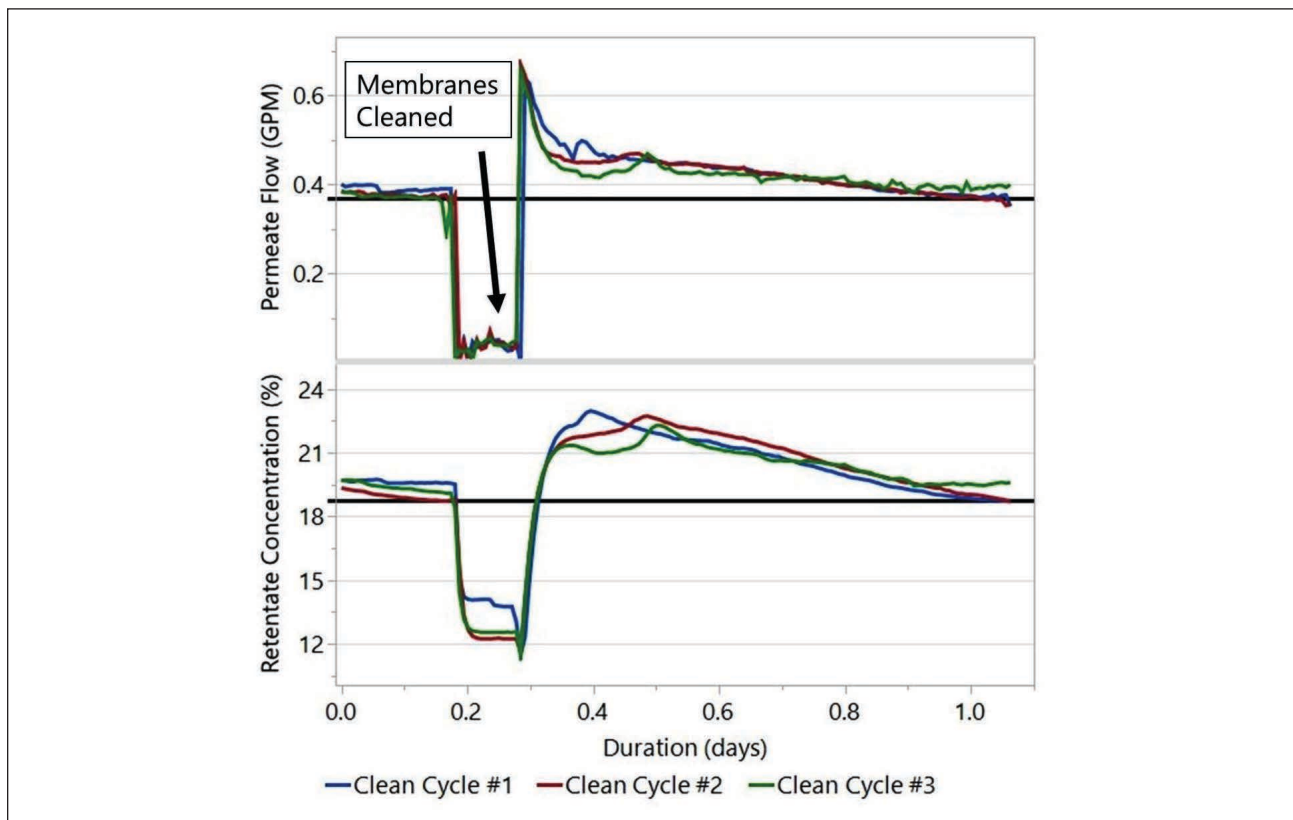
solids. By doing so, we estimate that the difference between minimum and maximum retentate solids can decrease by more than 80%.

Performance stability

Maintaining stable performance is an essential requirement for economical BL membrane concentration. An inability to maintain consistent performance results in unacceptable

system downtime to replace failed, fouled, or underperforming membranes. Previous attempts to scale membranes for BL processing, even in well controlled laboratory environments, have struggled to maintain stable performance due to irreversible fouling of the membrane surface, leading to permanent permeate flow loss over the course of hours to days [18]. While all membranes are susceptible to fouling, this challenge is exacerbated with BL, given its solids

RECOVERY CYCLE



7. Overlay of permeate flow rate and retentate concentration following three cleaning cycles performed several days apart. The membrane clean is repeatable and effective with similar peak performance and subsequent slope, which is ideal for steady operation in large scale systems with cleans distributed across many modules.

loading (12%–18%) and diverse range of chemical constituents [19]. Graphene oxide membranes are naturally resistant to fouling due to their surface charge, hydrophilicity, and smooth coated surfaces [12,13]. Here, we leverage those material properties, as well as a clean-in-place (CIP) system for foulant removal, to obtain reliable performance over the course of several weeks of continuous operation.

Membrane fouling consists of two different types: reversible, caused by the deposition of materials on the membrane surface that can be removed by physical or chemical cleaning; and irreversible, caused by partial or complete blockage of internal membrane pores that cannot be recovered. **Figure 6** shows a detailed view of permeate flow rate over a 250-h time period. As the membranes are operated, a slow decrease in permeate flow rate is observed, caused by the buildup of deposited material on the membrane surface [20] until it approaches the lower dashed line. When a cleaning sequence is performed, the permeate flow is restored to its original level (upper dashed line). The ability to repeat this cycle many times without a loss in baseline flow rate demonstrates that the membrane fouling is a reversible process.

Due to the large volumes of BL feed required to foul GO membranes (>10,000 gallons of BL per week even at pilot scale), CIP experiments are difficult to accurately evaluate on a laboratory scale. As such, this work is the first conclu-

sive demonstration of the extent of GO resistance to BL fouling. However, even small deviations in permeate flow rate after a CIP event can indicate incremental amounts of irreversible fouling, which may build up over months or years of operation. A more detailed look at the CIP process is depicted in **Fig. 7**. As the membranes are removed from the BL flow path for cleaning, the permeate flow from those modules drops to zero. Immediately after returning to service, there is a sharp spike in permeate flow. Simultaneously, there is an increase in the retentate concentration, from a baseline of 19% to approximately 22%. This is a direct result of the increased rate of permeate removal. The three cleaning cycles shown were performed several days apart under the same process conditions. The agreement in permeate flow rate, retentate solids, and similar slopes following CIP indicates no changes in the fouling behavior over time, demonstrating the ability to successfully remove foulants and return membrane performance to its original level. This consistency ensures that commercial scale systems can be designed to perform reliably over the full life of the membrane modules, which for initial installations, is expected to be approximately one year.

Permeate quality

The ability to match permeate quality with that of evaporator condensate is a requirement for a membrane system to

Characterization	Permeate	Combined Condensate
Conductivity ($\mu\text{S}/\text{cm}$)	231	200 - 1000
COD (mg/L)	672	1716
Methanol (mg/L)	463	760
Total Carbon (w %)	0.1	0.21
Sodium (ppm)	<42.7	<49.9
Potassium (ppm)	<42.7	<49.9
Silicon (ppm)	61.1	<49.9
Sulfate (ppm)	<20	<20
Carbonate (ppm)	1500	5000
Chlorine (ppm)	<20	<20



8. (a) Chemical characterization of the membrane permeate and a combined condensate sample from the same mill, and (b) a picture of the feed black liquor (left) and the output permeate (right). The permeate is visibly clear and of comparable composition to the combined condensate.

integrate with existing mill infrastructure. If the permeate produced by a membrane system is less clean than condensate streams, mill water balances would need to change, which could increase overall mill water consumption. To achieve the required purity of permeate, our system leverages a multi-pass filtration approach to first remove larger organics species and then the smaller salts. **Figure 8** displays the chemical composition of the final permeate along with that of combined condensate from the same mill, as well as a photograph of both the feed and permeate water. The permeate is visibly clear and free of any color bodies that might interfere with reuse.

In addition to excellent visible clarity, the chemical analyses performed by an independent, third-party laboratory indicate that permeate would be a good substitute for condensate. Sodium is one of the most important permeate constituents, as any sodium lost in permeate reuse or treatment must be made up with purchased chemicals. Both the combined condensate and our permeate had sodium levels below the test detection limit (50 ppm), indicating negligible amounts present in either stream. Many mills do not measure sodium levels in condensate frequently and instead rely on conductivity as a practical measurement for condensate purity. Here, the permeate (231 $\mu\text{S}/\text{cm}$) also compares favorably to that of the combined condensate (200–1000 $\mu\text{S}/\text{cm}$). The COD level of the permeate (672 mg/L) is below that of the condensate (1716 mg/L) as well as many mill effluent streams, which is relevant for mills that treat part of their condensate as effluent. There was a moderate amount of methanol in permeate samples (463 mg/L), which is likely the source of most of the COD. If desired, the permeate could be fed to a stripping column to recover the methanol or directly used as process water in methanol tolerant locations such as hooded washers.

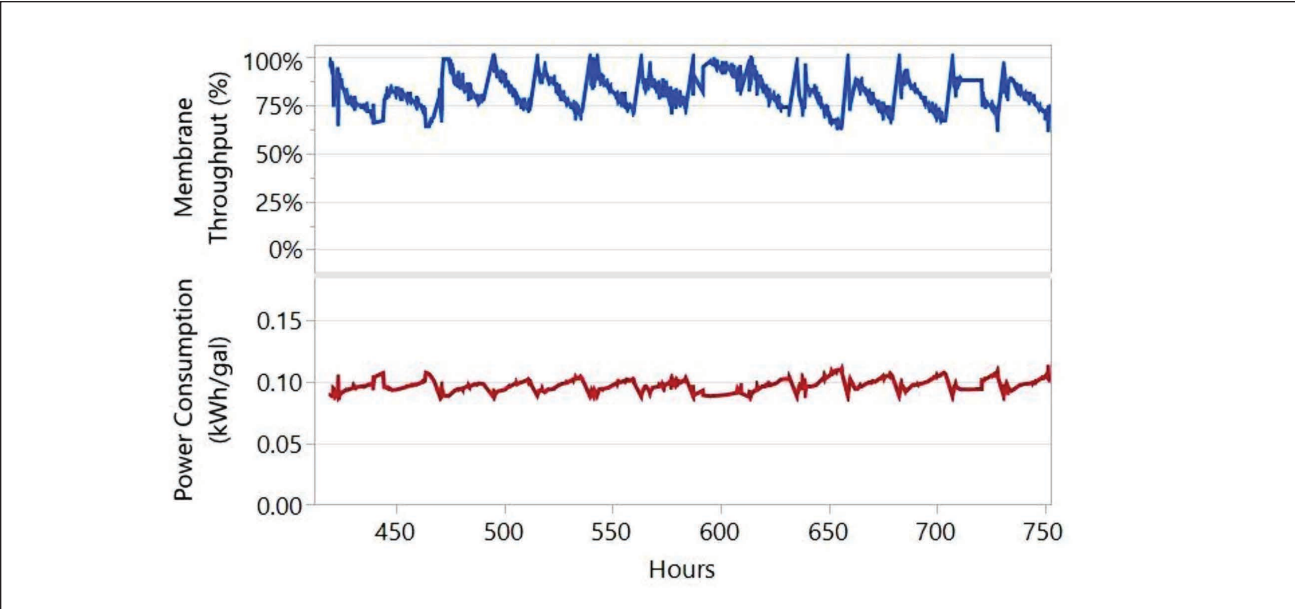
Energy and economic impact

Membrane systems capable of treating black liquor will drastically reduce energy consumption of BL concentration to improve mill profits and achieve sustainability goals. The data collected during the demonstration enables calculation of the energy savings from a commercial scale membrane system.

First, we calculate the energy required by a full-scale version of the system based on the results of this work. The power consumption of a commercial membrane system is dominated by pumps, specifically those used to achieve high operating pressures and flow rates across the membrane surface. Because the pilot system is comprised of full-scale membrane subunits, we can directly compare its performance with a commercial scale system on a per unit of water produced basis. Using Eq. 1, we calculate the estimated power consumption of the pilot system pumps in real time, then normalize by the permeate flow rate to track the power consumed per gallon of permeate produced (**Fig. 9**). There are some fluctuations in this figure of merit over time that are accounted for by changes in membrane permeate throughput and are also shown.

Based on this pilot system performance data, the steady state pumping power required to extract clean water from WBL is 0.11 kWh/gal permeate. At the commercial scale, auxiliary systems such as CIP will add an additional 25% to the power consumption, bringing the total to 0.14 kWh/gal permeate. For a mill processing 2000 tons of BLS/day from 14% to 20% concentration, the total power requirement for a membrane system would be approximately 6.6 MW. These utility expenses can be easily compensated for by evaporator steam savings in mills with favorable power pricing and/or relatively high fuel costs for generating steam.

RECOVERY CYCLE

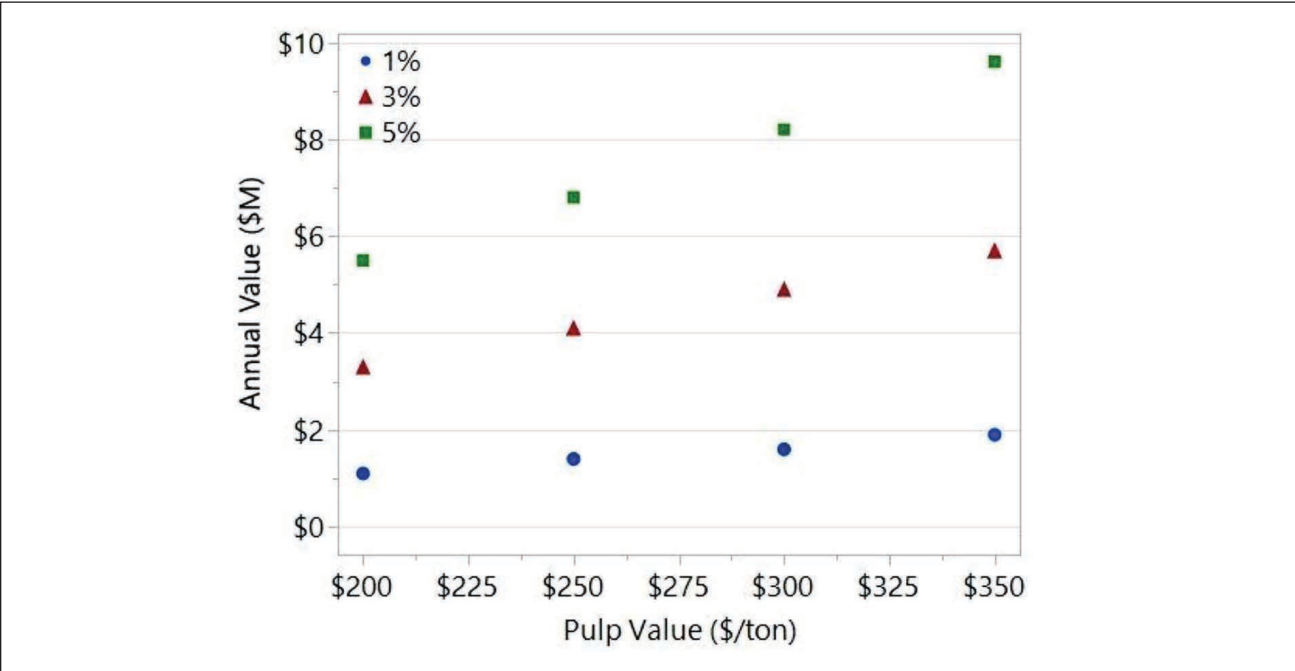


9. Normalized membrane throughput and power consumption as a function of time for the pilot membrane system. Power consumption fluctuates between 0.08 and 0.11 kWh/gal of permeate as a result of membrane throughput fluctuations.

To compare most directly, we calculate the typical energy requirement of a multiple effect evaporator on a condensate unit basis. To correctly account for the full energy impacts of adding a membrane system, we first compute both the amount of steam heat (generated as high pressure steam) required by a typical set of evaporators and the associated power generation from a backpressure or extraction turbine. Assuming a steam temperature and pressure of 500°C and 800 psi, respectively, and a boiler efficiency

of 75%, a multi-effect evaporator with a steam economy of 5 requires 0.85 kWh of heat per gallon of condensate. However, this high-pressure steam is first run through a turbine to generate approximately 0.10 kWh of electrical power per gallon of condensate (assuming a typical turbine yield of 8 tons of steam per MWh). Thus, the net amount of energy required by evaporation is 0.75 kWh/gal of condensate, making the membrane system 81% more efficient.

In previous works, we have performed in-depth model-



10. Calculated values of pulp production increases of 1%, 3%, and 5% at different pulp prices, ranging from \$200/metric ton to \$350/metric ton.

ing of the impacts of membrane concentration on mill economics [4,21]. Here, we apply the results obtained from our pilot system as direct inputs to these models, with a focus on three key areas: steam savings, carbon pricing, and increased BL throughput.

First, membrane concentration reduces the load on the evaporator train, thereby reducing steam demand. By increasing the inlet solids concentration from 14% to 20%, as demonstrated here, an evaporator steam reduction of 36% is achieved [4], which is more than 10% of the average mill wide energy consumption [1]. For a mill processing 2000 metric tons of BLS/day, this translates to 8.8 kg/s of steam savings or \$5 MM in annual gross savings at a steam cost of \$18 per metric ton of low-pressure steam (inclusive of fuel costs and net of power generation). Subtracting the power required by the membrane system, a mill can expect \$2 MM/year in net energy savings if the membrane system consumes power at \$50/MWh. Increasing the membrane system size to obtain even higher retentate solids would enable further net energy savings.

Second, for jurisdictions that have adopted carbon pricing, membrane-based BL concentration presents an opportunity for savings through reduced carbon emissions. In Canada, the above 2000 metric ton BLS/day mill, if using supplemental natural gas as fuel (emission factor of 53.06 kg CO₂/MMBtu [22]) and assuming a boiler efficiency of 80%, would save a further \$2.2 MM annually at the 2022 carbon price of \$50/metric ton of CO₂ equivalent and \$7.4 MM in 2030 when the price will increase to \$170/metric ton.

Lastly, for mills that are evaporator limited in their production capacity, membrane systems offer an affordable route to increased capacity by raising both BL throughput and inlet solids. Increasing the inlet BLS from 14% to 20% with membrane concentration yields a potential solids throughput increase of up to 57%. More realistically, we estimate that adoption of membrane concentration could allow mills to increase pulp production by up to 5% above current production levels. The value created by doing so is estimated in **Fig. 10** for a mill producing 1500 metric tons/day of pulp. At \$350/metric ton of marginal pulp production, a 5% production increase translates to a \$9.6 MM annual benefit.

CONCLUSIONS

We have designed, built, and deployed the first pilot black liquor membrane concentration system, using commercially sized, graphene oxide spiral wound membranes. Across deployments to three separate sites and nearly 6000 h of operational runtime, we have achieved consistent black liquor concentration, produced clean permeate water, and overcome the stability limitations of conventional membrane materials. By increasing BLS from 14% to >20%, we have demonstrated the ability of membrane systems to augment existing evaporator trains and unlock value in steam

savings, carbon pricing offsets, and increased production throughput. Simultaneously, we have produced clean clear permeate comparable to condensate with < 50 ppm sodium, conductivity of <250 µS/cm, and <1000 ppm of COD.

These pilot demonstrations conclusively prove that the long-theorized value of membrane black liquor concentration can successfully be translated to real world results. This work paves the way for the deployment of full-scale commercial membrane systems in the immediate future. **TJ**

LITERATURE CITED

1. BCS Inc./Oak Ridge National Laboratory, "Materials for separation technologies: energy and emission reduction opportunities," Report for U.S. Department of Energy Efficiency and Renewable Energy Industrial Technologies Program, BCS Inc./Oak Ridge National Laboratory, Columbia, MD, USA/Oak Ridge, TN, USA, 2005. Available [Online]: https://www1.eere.energy.gov/manufacturing/industries_technologies/imf/pdfs/separationsreport.pdf <03April2023>.
2. Sholl, D.S. and Lively, R.P., *Nature* 532(7600): 435(2016). <https://doi.org/10.1038/532435a>.
3. Appti, "Black liquor concentration technology roadmap," Report for Alliance for Pulp & Paper Technology Innovation, Appti, Washington, DC, 2016. Available [Online] https://www.appti.org/store/p4/Black_Liquor_Concentration_Roadmap.html, <03April2023>.
4. Kleiman-Lynch, M., Rivera, D.F., Frayne, S.H., et al., *TAPPI J.* 20(5): 331(2021). <https://doi.org/10.32964/TJ20.5.331>.
5. Kirbawy, S.A. and Hill, M.K., *Ind. Eng. Chem. Res.* 26(9): 1851(1987). <https://doi.org/10.1021/ie00069a022>.
6. Tanistra, I. and Bodzek, M., *Desalination* 115(2): 111(1998). [https://doi.org/10.1016/S0011-9164\(98\)00030-7](https://doi.org/10.1016/S0011-9164(98)00030-7).
7. Wallberg, O., Jönsson, A.-S., and Wimmerstedt, R., *Desalination* 156(1): 145(2003). [https://doi.org/10.1016/S0011-9164\(03\)00337-0](https://doi.org/10.1016/S0011-9164(03)00337-0).
8. Keyoumu, A., Sjö Dahl, R., Henriksson, G., et al., *Ind. Crops Prod.* 20(2): 143(2004). <https://doi.org/10.1016/j.indcrop.2004.04.017>.
9. Hummers, W.S. and Offeman, R.E., *J. Am. Chem. Soc.* 80(6): 1339(1958). <https://doi.org/10.1021/ja01539a017>.
10. Marcano, D.C., Kosynkin, D.V., Berlin, J.M., et al., *ACS Nano* 4(8): 4806(2010). <https://doi.org/10.1021/nn1006368>.
11. Ma, J., Ping, D., and Dong, X., *Membranes* 7(3): E52(2017). <https://doi.org/10.3390/membranes7030052>.
12. Rashidi, F., Kevlich, N.S., Siquefield, S.A., et al., *ACS Sustainable Chem. Eng.* 5(1): 1002(2017). <https://doi.org/10.1021/acssuschemeng.6b02321>.
13. Bano, S., Mahmood, A., Kim, S.-J., et al., *J. Mater. Chem. A* 3(5): 2065(2015). <https://doi.org/10.1039/C4TA03607G>.
14. Han, J.-L., Zhang, D., Jiang, W., et al., *J. Membr. Sci.* 576: 190(2019). <https://doi.org/10.1016/j.memsci.2018.12.055>.
15. Wang, Z., Ma, C., Siquefield, S.A., et al., *ACS Sustainable Chem. Eng.* 7(17): 14915(2019). <https://doi.org/10.1021/acssuschemeng.9b03113>.
16. Frayne, S., MacLeod, M., MacDonald, B.I., et al., U.S. pat. 11097227B2 (Aug. 24, 2021).
17. MacLeod, M., Frayne, S., MacDonald, B.I., et al., U.S. pat. 11123694B2 (Sept. 21, 2021).

RECOVERY CYCLE

18. Jönsson, A.-S. and Wimmerstedt, R., *Desalination* 53(1): 181(1985). [https://doi.org/10.1016/0011-9164\(85\)85060-8](https://doi.org/10.1016/0011-9164(85)85060-8).
19. Zaman, A.A. and Fricke, A.L., *Ind. Eng. Chem. Res.* 35(2): 590(1996). <https://doi.org/10.1021/ie950202h>.
20. van der Marel, P., Zwijnenburg, A., Kemperman, A., et al., *J. Membr. Sci.* 348(1): 66(2010). <https://doi.org/10.1016/j.memsci.2009.10.054>.
21. Rivera, D.F., Kleiman-Lynch, M., Keller, B.D., et al., *TAPPI J.* 20(7): 467(2021). <https://doi.org/10.32964/TJ20.7.467>
22. EPA, "Emission factors for greenhouse gas inventories," Report for United States Environmental Protection Agency, EPA, Washington, DC, 2022. Available [Online]: https://www.epa.gov/system/files/documents/2022-04/ghg_emission_factors_hub.pdf<03April2023>.

ABOUT THIS PAPER

Cite this article as:

Rae, S., Richards, E.V., Kleiman-Lynch, M., et al., *TAPPI J.* 22(4): 223(2023). <https://doi.org/10.32964/TJ22.4.223>

DOI: <https://doi.org/10.32964/TJ22.4.223>

ISSN: 0734-1415

Publisher: TAPPI Press

Copyright: ©TAPPI Press 2023

[About this journal](#)

ABOUT THE AUTHORS

Membrane based black liquor concentration has the potential to enable significant cost savings and emissions reductions, but it has previously proven to be technically very difficult due to the challenging process conditions. Demonstrating pilot level performance is a key step in proving that, with the right system and materials, this technology is feasible at scale. This research builds on our previous modeling work published in *TAPPI Journal* and demonstrates a reduction to practice of the technology. It also builds upon work by other groups who have shown the potential of graphene oxide membranes for use in high temperature, high pH applications.

Membrane fouling in a field environment is very different from that which occurs at a benchtop scale. Learning how to address this with effective cleaning strategies was a major challenge, but also a necessary one in the development of this technology. Demonstrating that the measured energy reductions on a kWh/gal basis approach the theoretical limits determined with modeling was very satisfying and provided great validation of the system design and technical approach.

Each of the three mills where our system was deployed presented unique challenges. Determining how to operate our system to accommodate these

challenges while maintaining system stability and product quality forced us to think creatively and provided essential insights into what will be required of commercial scale systems.

In this work, we share the results of more than 8 months of field testing using our pilot system containing commercial scale membrane modules. We describe the ability of our system to concentrate black liquor, produce clean, clear permeate and to do so while overcoming the stability limitations of previous generations of membrane technologies. Building upon our previous work, we translate these results into economic impacts, demonstrating predicted steam savings and increased pulp production for evaporator limited mills.

This work validates that membrane systems for black liquor concentration are ready for commercialization and lays out how mills can benefit from the adoption of this technology. Our next steps are to continue the scaling process and to build commercial sized black liquor concentration systems.

Rae is chemical engineer, Richards is Materials and Testing team lead, Kleiman-Lynch is process engineer, Keller is CTO and co-founder, and MacDonald is director of Product for Via Separations in Watertown, MA, USA. Email MacDonald at bmacdonald@viaseparations.com.



Rae



Richards



Kleiman-Lynch



Keller



MacDonald