PNNL- 38899

Novel Method for Domestic Stable Isotope Production (CRADA #667) Final Report

January 2026

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Pacific Northwest National Laboratory
Richland, Washington 99354

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CRADA Title: Novel Method for Domestic Stable Isotope Production

Responsible Technical Contact at DOE Lab (PNNL):

Dan R. Bottenus

Provide a list of publications, conference papers, or other public releases of results, developed under this CRADA:

While this research has not yet been published externally, the following technologies, techniques, and methodologies have been developed:

1. Microchannel distillation column design, fabrication, and commissioning methods.
2. Experimental test protocols for distillation performance evaluation (startup/shutdown procedures, steady-state runs, sampling/analysis approach).
3. Experimental test protocols for isotachophoresis performance evaluation (operating procedures, analysis approach).
4. Process modeling methodology using COMSOL Multiphysics to simulate microchannel distillation column behavior and isotachophoresis separation performance.

Provide a detailed list of all subject inventions, to include patent applications, copyrights, and trademarks:

No subject inventions were generated under this CRADA.

Executive Summary of CRADA Work

This research advances understanding of how to practically obtain chlorine enriched in the isotope ^{37}Cl, a capability widely viewed as enabling for the next generation of molten chloride salt reactors. Pacific Northwest National Laboratory investigated and directly measured chloride isotope separation using two distinct approaches, microchannel distillation (MCD) and isotachophoresis (ITP).

MCD applies an intensified form of distillation: by pushing liquid and vapor through very thin passages and a porous microwicking material, it greatly increases gas–liquid contact area, allowing even very small boiling point differences to be exploited with significantly smaller equipment than conventional distillation. The demonstrated system was designed for batch processing of hydrogen chloride (HCl) gas, and information collected from the column was used to determine what column length would be needed to reach a target of 90% ^{37}Cl enrichment—information that is essential for realistic scale-up and cost estimation.

ITP, in contrast, is an electrically driven separation method: chloride ions are arranged between a leading electrolyte and trailing electrolyte so that ions “stack” into distinct, sharpened bands based on small differences in their tendency to migrate under an applied voltage. A counterflow is used to hold those bands in place to continuously withdraw an enriched ^{37}Cl stream. By showing that both MCD and ITP can produce measurable isotope separation and by comparing their experimental results and scale-up considerations, this project provides a clearer, evidence-based path toward industrially relevant ^{37}Cl enrichment—supporting cleaner molten salt reactor operation and ultimately benefiting the public through improved nuclear energy options.

Summary of Research Results

This report presents the progress achieved on our chlorine isotope separation efforts in the CRADA #667 agreement. We used a microchannel distillation (MCD) packing structure to enrich HCl isotopes (H35Cl, H37Cl) and an isotachophoresis (ITP) separation to enrich ionic chloride species (^{35}Cl- and ^{37}Cl-). A 2 m tall MCD column was constructed and achieved 264 separation stages over several days, which is well above the goal of 50 stages outlined in the CRADA. The resulting height equivalent to a theoretical plate (HETP) was 0.76 cm. ITP tests were run ~ 24 hours and achieved a single-stage separation factor of 1.559.

1. Project Overview

Chloride isotope separation is a highly demanded technology that is needed to enable the next generation of molten salt reactors. Molten chloride salt reactors require chloride that is enriched in the ^{37}Cl isotope. The major isotope of chlorine, ^{35}Cl, has a strong (n, proton) threshold reaction that will poison the reactor and also gives rise to ^{36}Cl as an activation product, which is a long-lived, energetic beta source that complicates waste disposal [1]. Pacific Northwest National Laboratory (PNNL) has investigated two different methods to enrich the ^{37}Cl isotope using two different techniques reported here: microchannel distillation (MCD) and isotachophoresis (ITP). The objective of this research was to obtain measurements of the separation of chloride isotopes to compare the performance and feasibility of each method.

MCD uses microwicking technology to enhance the separation of liquid components based on differences in boiling point. The main difference from conventional distillation is that a microchannel distillation column makes use of thin liquid and vapor flow passages to increase the interfacial area and minimize mass transfer resistances. The MCD concept described here uses a thin, porous material that greatly increases the interfacial area of the gas/liquid interface to reduce the size of the equipment needed to perform the desired separation. This material allows even tiny differences in boiling point to be used for gas separation in reasonably sized equipment. Our system is designed for semi-automated batch distillation of hydrogen chloride (HCl) gas to demonstrate feasibility and determine the column length needed for 90% enrichment.

ITP separates ions based on their electrophoretic mobility, which may be different for different isotopes. In contrast to conventional electrophoresis, where bands of ions separate and broaden with time, ITP brackets the desired ions between a leading electrolyte (LE) and a trailing electrolyte (TE). The ions in between “stack up” into separate bands based on their mobility. A counterflow can be used to keep the bands stationary, which allows us to continually withdraw enriched ^{37}Cl isotope from its respective band.

We successfully demonstrated chloride isotope separations using both MCD and ITP. In this report, we first describe the background and experimental design for each method. Then we present results of our separation measurements on each system. Finally, we compare the two methods and analyze scale-up design considerations.

2. Microchannel Distillation Technical Approach

Microchannel distillation is a technology that has been advanced by PNNL for use in isotopic enrichment [2]. Distillation is a method of separating volatile liquid mixtures based on the relative volatility of the liquid components. In distillation columns, a counterflow of liquid and vapor is used to accomplish multiple separation stages in a single process. Microchannel distillation makes use of thin, porous materials and flow channels inside the distillation column to increase the interfacial area for vapor liquid equilibrium to enhance the performance of the distillation process. The use of microchannels significantly reduces the size of the equipment needed to achieve the desired separation. Several examples of microscale porous column packing materials are shown in Figure 1.

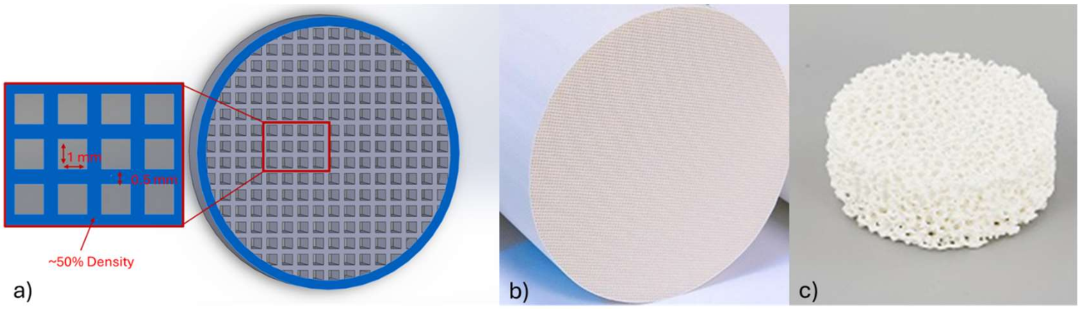

Figure 1. Three types of microchannel packing. a) 3D printed packing structure with gas channels and partially dense liquid channel walls (not evaluated but have parts to evaluate with additional funding). b) Alumina extruded honeycomb packing (not evaluated here). c) Expanded alumina foam packing (evaluated here).

For an isotopic separation, the separation performance depends on the relative volatility of each isotopologue, and the number of distillation stages used. According to literature, the relative volatility of $H^{35}Cl/H^{37}Cl$ at its normal boiling point is between 1.0004 and 1.0006 [3]. The low volatility difference implies a large number of separation stages are needed to perform isotopic enrichment.

The minimum number of stages (N) required to perform a given separation [4] is given by

$$N = \frac{\ln\left(\frac{x_d(1-x_b)}{x_b(1-x_d)}\right)}{\ln(\alpha_{avg})},$$

where x_d is the mole fraction of the more volatile component in the distillate, x_b is the mole fraction in the bottoms product, and α_{avg} is the average relative volatility. For example, separating $H^{35}Cl$ from $H^{37}Cl$ requires at least 6680 stages to achieve 90% pure $H^{37}Cl$ in the bottoms product, assuming the distillate mole fraction remains at the natural abundance of 24.2%.

Each stage requires a length of column referred to as the height equivalent to a theoretical plate (HETP). A given column packing can be characterized by its HETP. Commercial packing has HETPs typically in the range of 0.3-0.5 m [5], which would result in a column length of at least 1336 m to perform the desired 90% chloride separation. The best available commercial packing is the Sulzer Laboratory packing [5] reported to have an HETP as low as 2 cm but is available only in diameters of 2 to 8 cm. The Sultzer laboratory packing would still require a column of 134 m in length. Microchannel distillation has been shown to reduce the HETP to below 1 cm [2], which could significantly reduce the amount of column length required to achieve this separation.

Our MCD setup is shown in Figures 2 and 3. The column is 79 in long, or approximately 2 m, and has an inner diameter of 1 in. Packing consists of alumina (85%, porosity 80-90%) open cell foams (1 in diameter, 1 in length). This material has additional micropores and mesoscale pores that increase mass transfer by liquid wicking into these features.

The system was assembled in-house using components compatible with anhydrous HCl and operated at near atmospheric pressure and temperature close to the boiling point of HCl (-85°C). Experiments processed ~ 130 g of HCl gas per run. The feed gas entered at the top of the column and was liquefied using a 16 W (@ 77K) Sunpower CryoTel GT cryocooler attached to a custom condenser heat exchanger fabricated from stainless steel for compatibility. At the operating temperature (-85°C), the lift provided by the cryo chiller is ~ 45 W. The distillation system was encased in a vacuum chamber to prevent heat leaks and keep the system adiabatic. Thus, no liquid cryogens were used to operate the system. As distillation was performed, ^{37}Cl was enriched at the bottom of the column over time, and ^{35}Cl was enriched at the top of the column.

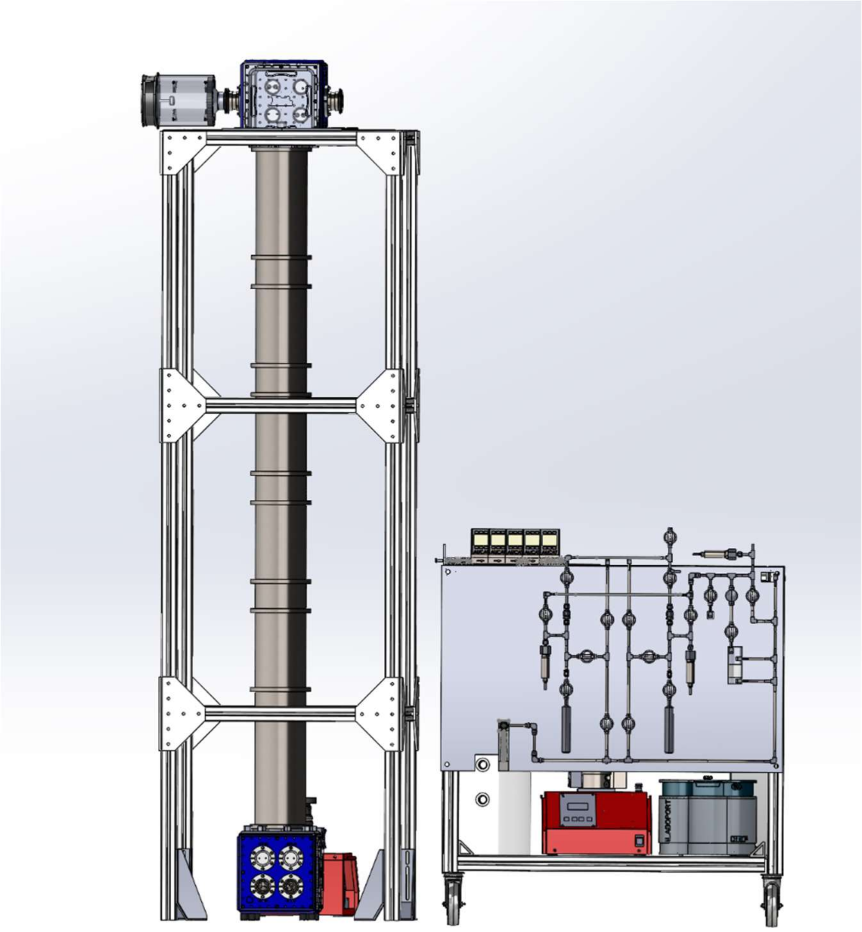

Figure 2. SolidWorks representation of the distillation system used for the HCl isotope enrichment.

Figure 3. Photo of microchannel distillation column and system.

During operation, small gas aliquots from the top and the bottom of the column were periodically collected for offline analysis to measure the amount of enrichment and determine the performance from the distillation column. Gas samples were analyzed off-line using an Agilent 8900 triple-quadrupole-ICP-MS/MS using previously developed methods to analyze chloride ratios [6]. After the experiment, the remaining HCl in the column was vaporized and absorbed in an abatement canister.

2.1. CFD design aid

COMSOL Multiphysics (version 6.3) was used to guide the design of the distillation packing. The main variable studied for the packing component was the size of the vapor channels; it was found that both smaller vapor channels and thinner walls improve separation. These geometric parameters are constrained and determined by manufacturing capabilities. Based on the results of the COMSOL simulations, we chose the expanded alumina foam depicted in Figure 1c) as our packing material for the initial test, since it minimizes the vapor channel dimensions and wall thickness while also offering improved wicking compared to other available materials.

The COMSOL model was also used to determine the operational flow rates. The cryo-chiller lift determined a maximum flow rate of 0.042 g/s, and a minimum flow rate of 0.005 g/s was estimated as the limit below which the fluid in the porous walls will stagnate and halt the distillation. Figure 4 shows the HETP predicted by the COMSOL model across a range of internal column flowrates. The simulations used the dimensions of the alumina foam packing. Lower operation flowrates produce a smaller HETP, down to a minimum value of 0.028 cm at the lowest flowrate. This is the ideal situation, and experience has determined that performance is regularly at least 10X worse than simulation predictions, or an HETP between 0.3 and 0.6 cm for this packing material.

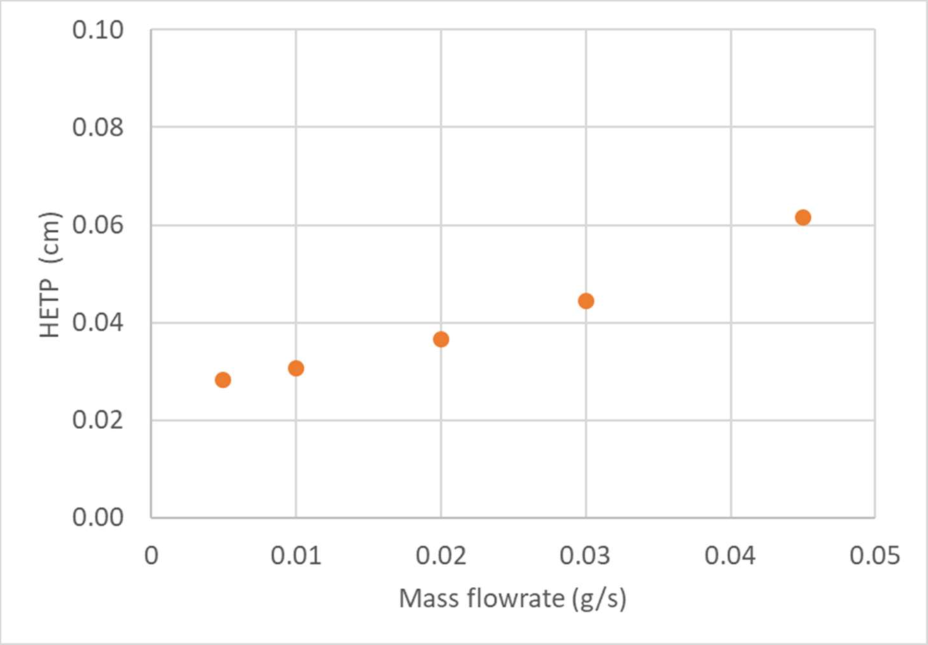

Figure 4: HETP predicted by COMSOL simulations at different mass flowrates in the MCD column.

Figure 5 shows a prediction of the ^{37}Cl mole fraction as a function of the length of the distillation column at an average internal column flowrate (0.02 g/s) and using dimensions of the 3D printed packing material referenced in Figure 1. Figure 5 shows approximately 3% (from 25% to 28% mole percent) enrichment for 0.1 m contact length, or a height equivalent to a theoretical plate (HETP) of 0.099 cm. If we again use our 10X heuristic, then we predict an HETP closer to 1 cm on a real column.

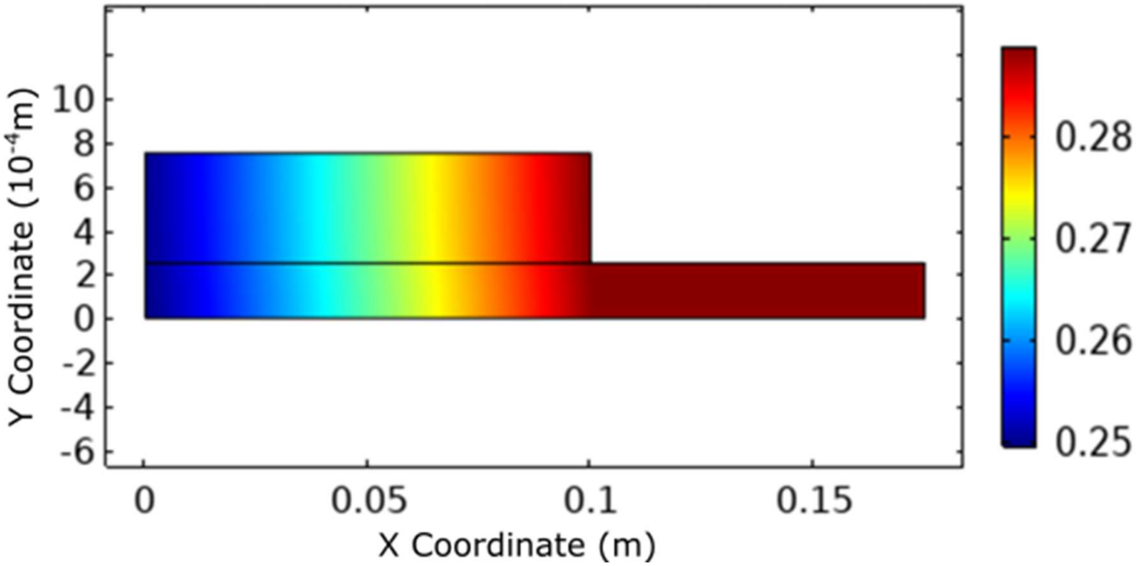

Figure 5. A COMSOL simulation showing the mole fraction of ^{37}Cl (represented by the heat map) as a function of length down the distillation column at a flowrate of 0.02 g/s. The simulation includes a vapor channel (top) and a porous wall for the liquid to flow through (bottom), with extra length to represent the reboiler.

2.2. MCD experimental overview

We performed two experiments in the MCD apparatus. The first experiment was run for 48 hours at a reboiler duty of 12.5 W. At this reboiler duty, the internal mass flowrate in the column is 0.028 g/s, which is in the middle of the range we can operate in; much lower will cause the column to stagnate, and much higher will exceed the reboiler and condenser power. The second experiment was run at the same reboiler duty for 168 hours to ensure that the column had time to reach steady state.

3. Isotachophoresis Technical Overview

Electrophoretic isotope separation relies on the small but measurable differences in the electrophoretic mobilities of different isotopes. Stationary isotachophoresis traps the isotope ions between a leading electrolyte and a trailing electrolyte, which causes each species to stack up in bands according to their electrophoretic mobility. A flow counter to the direction of migration of the ions keeps the bands stationary, which allows us to withdraw samples from the enriched ^{37}Cl isotope band from a stationary sample port. We used a solution of ammonium chloride (NH_4Cl) for our chloride ions, ammonium thiosulfate ($(NH_4)_2S_2O_3$) as the leading electrolyte (LE), and HEPES ($C_8H_8N_2O_4S$) as the terminating electrolyte (TE). Each species is titrated with ammonium hydroxide to carefully control pH conditions.

Figure 6 shows the bench-scale device used to perform our ITP separation, known as the VpHor and described previously [7]. The instrument is a vertical column containing fluid in the annulus between a boron nitride rotating rotor and a Plexiglas stationary stator. The rotation creates radial mixing by creating vortices (as shown in Figure 6), which is a form of undesired dispersion, but reduces the axial dispersion overall by eliminating electroosmotic flow. Cooling fluid is recirculated through the center of the hollowed-out rotor to remove Joule heat during experiments. The total annulus volume is ~ 25 mL. Electrodes located at the top and bottom of the VpHor are separated from the annulus by dialysis membranes that are continuously swept with LE (bottom) and TE (top) during experiments to remove electrolysis products and prevent buildup of hydrogen at the cathode and oxygen at the anode. A series of 21 sample ports are machined into the back of the chamber along the axial length of the column to remove samples for analysis after an experiment is performed.

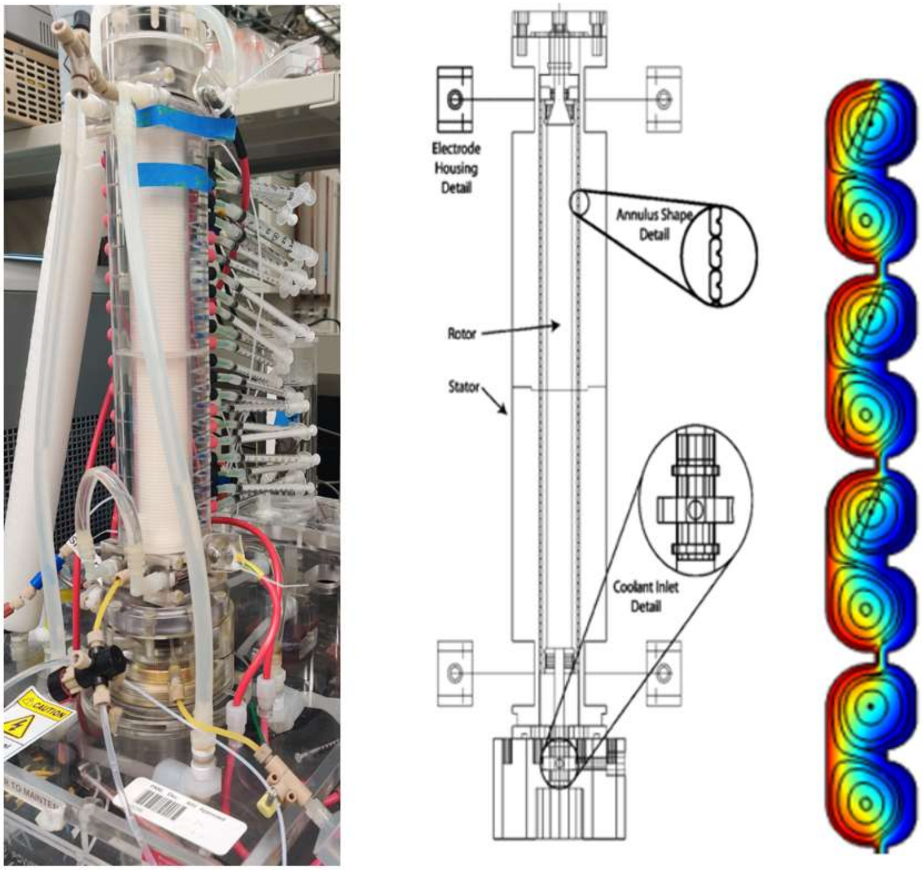

Figure 6: (Left) A depiction of the bench-scale electrophoresis unit known as the VpHor. (Center) A detailed drawing of individual components of the VpHor, including the electrode housing, rotor, stator, and coolant inlet. The string of kidney bean-like structures is the annulus between the rotor and the stator. (Right) Velocity profile in the VpHor annulus, with red indicating high velocity near the rotor wall, which spins at 50 rpm, and blue indicating low or zero velocity near the stationary stator wall.

3.1. COMSOL simulations

We used COMSOL Multiphysics to select the operating conditions for our ITP device. We used a one-dimensional (1D) model that used the Nernst-Planck equation to simulate the electric potential and the concentration profile of each ionic species under different concentrations and flowrates, and then we selected the conditions that gave the best separation.

The simulations used 10 mM thiosulfate as the LE, 5 mM HEPES as the TE, and ammonium as the counterion to match our experimental conditions. Simulations were run for an initial 4 hours simulation time to approach steady state, and then an additional 20 hours with a continuous chloride feed. The plateau concentration [8] (the ITP zone concentration determined by ionic properties) of chloride is 19 mM at this LE concentration, so both the initial chloride fill and the continuous chloride feed were set to that concentration. Our simulations showed that if a lower concentration is used in the initial fill, then the chlorine band will shrink as the ions concentrate over time until the system reaches steady state, and if the feed is set to a lower concentration, then the chlorine band will again shrink over time as more chloride is withdrawn than is being fed.

Previous batch experiments gave a ^{37}Cl/^{35}Cl ratio of about 0.5 on the heavy side and about 0.25 on the light side (compared to a ratio of 0.32 in natural chlorine) after 24 hours. In our simulations, we used the dispersion in the VpHor as a fitting parameter and tuned it until the heavy side ratio reached 0.64 and the light side reached 0.18 after 24 hours, which we considered to be sufficiently close to the previous batch data to inform our experimental design.

Figure 7 shows the concentrations of each species after 24 hours as a function of location in the VpHor device, with zero being at the top of the column. After an initial 4 hours to equilibrate, chloride was fed at a rate of 2 μL/min at a position of x = 14 cm and drawn off at positions x = 23.5 cm (light end) and x = 4 cm (heavy end) in a 3:1 ratio (i.e., 1.5 μL/min on the light end and 0.5 μL/min on the heavy end). The ions form into bands of LE (right side), chloride isotopes (middle), and TE (left side). The ^{37}Cl isotope is enriched on the left side of the chloride band (i.e., the top of the column), and the ^{35}Cl isotope is enriched on the right side (or bottom of column).

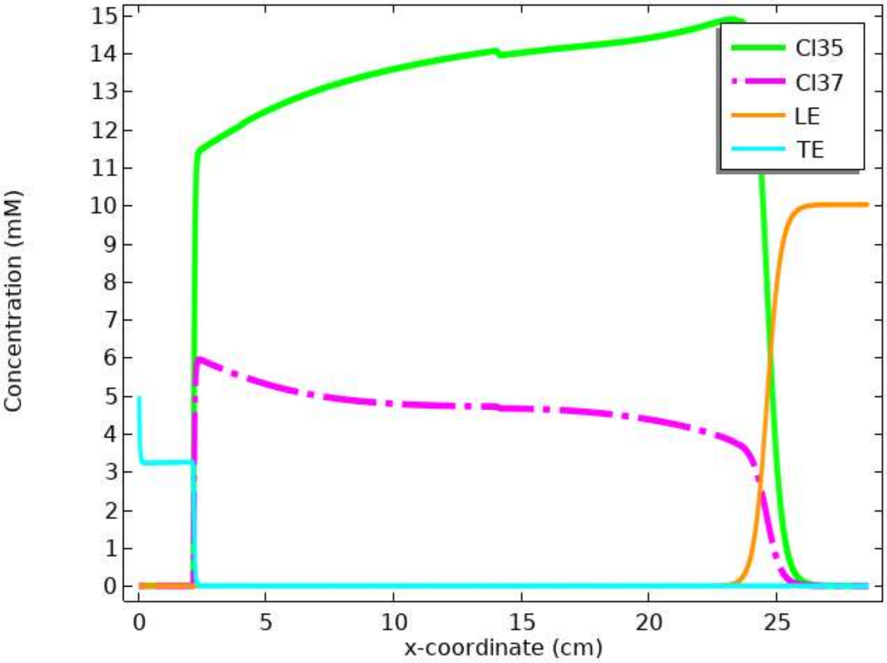

Figure 7: Concentrations of each ionic species as a function of location in the VpHor column, predicted by our COMSOL model. The concentrations of ^{35}Cl, ^{37}Cl, the leading electrolyte (LE) and the trailing electrolyte (TE) are shown at the end of a 24-hour period. After an initial 4 hours to equilibrate, chloride was fed into the middle of the column (at x = 14 cm) at a rate of 2 μL/min and drawn off at a 3:1 ratio, meaning 1.5 μL/min on the light end and 0.5 μL/min on the heavy end. The simulation was run for an additional 20 hours to predict the profile at the end of a 24-hour period.

This simulation was repeated at different feed rates and draw ratios and extended so that the initial equilibration period lasted 24 hours and the feed and draw period lasted an additional 48 hours, with the resulting isotope ratios shown in Figure 8. With no feed or draw, the isotope ratio continues to increase with time, taking longer than 72 hours to fully equilibrate. When the feed and draw streams are added, the ratio levels off more quickly or even decreases slightly. Lower feed rates give better separation, and higher draw ratios (meaning more is drawn off the light end than the heavy end) also give better separation. This creates a clear tradeoff between the purity and the production rate of the ^{37}Cl-enriched product. Based on these simulations, we chose to run our experiments at a 2 µL/min withdrawal rate at a 3:1 draw ratio, which was predicted to give high separation while also ensuring that we had enough sample to analyze.

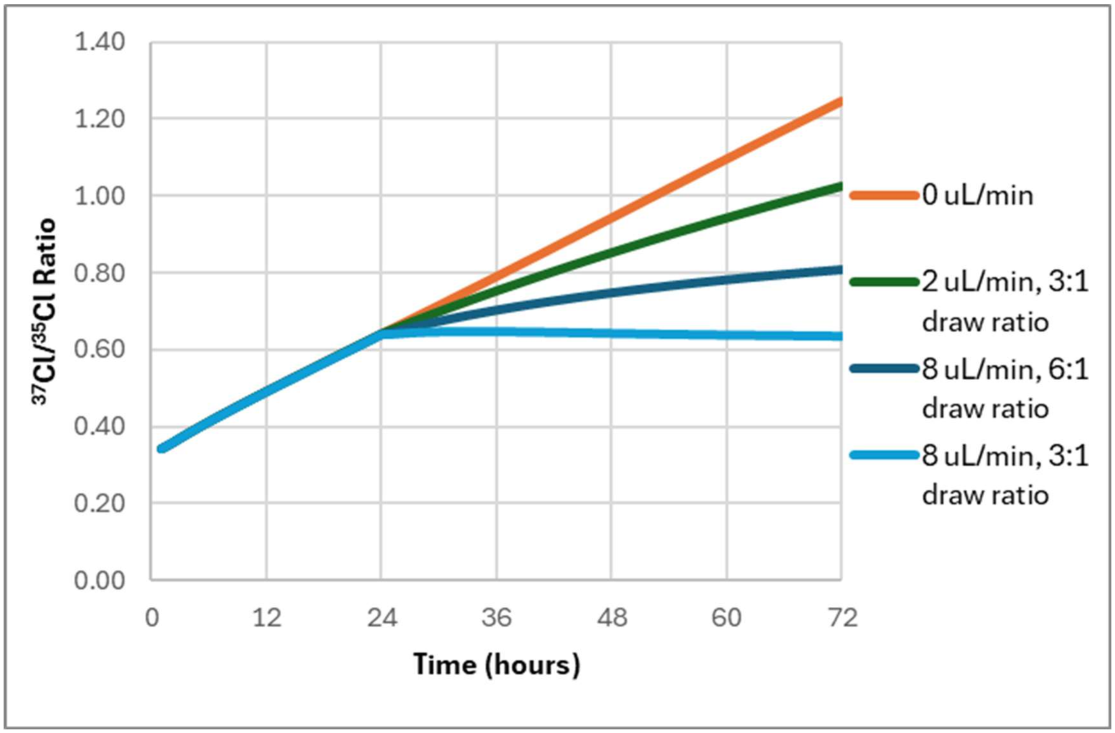

Figure 8: The ratio of ^{37}Cl to ^{35}Cl isotopes is shown as a function of time for four different combinations of feed rate and draw ratio.

3.2. ITP Experimental Overview

Table 1 summarizes the operating conditions of each of our two experimental ITP runs. Each experiment was run for 24 hours. The LE was 10 mM ammonium thiosulfate, titrated with ammonium hydroxide to a pH between 9 and 10. The TE was 5 mM of either TAPS or HEPES. In our first run, we used TAPS as our TE, and we also added 5 mM barium hydroxide to react away any carbonated species that might be present in the device that could disrupt our measurements. In our second run, we switched to HEPES and we did not add the barium hydroxide to match more closely with our previous batch experiments. A small quantity of OrangeG dye was also added to the TE to serve as an indicator of where the TE band ended and the chloride band began. A sample of 19 mM ammonium chloride was used to fill the column between the TE and LE.

Experiments were run with a voltage slightly over 1 kV, and with a current draw close to 30 mA, as given in Table 1 and Figure 9. The counterflow was set to a flowrate that would maintain the OrangeG indicator band in a constant position, which was approximately 0.03 mL/min. After waiting several hours to ensure the OrangeG band did not move, we began to continuously withdraw 1.5 µL/min from a sample port near the bottom of the column (enriched in ^{35}Cl) and 0.5 µL/min from a sample port near the top of the column but below the OrangeG indicator band (enriched in ^{37}Cl), while feeding 2 µL/min of 19 mM ammonium chloride to the center of the column. Samples were collected every hour.

As shown in Figure 9, the system exhibits an initial conditioning phase followed by a prolonged period of stable operation. A mid-run disturbance is observed as a transient decrease in current while the applied voltage remains stable. This behavior is attributed to bubble formation at the top of the column, which temporarily disrupted current flow. The issue was subsequently mitigated, after which stable operation resumed, and the experiment continued until 24 hours had passed.

Table 1. *Operational conditions of ITP experiments.*

Experiment number	Leading electrolyte	Terminating electrolyte	Voltage (kV)	Est. time (hr)	Meaningful sample numbers
1	10 mM $(NH_4)_2S_2O_3$, pH 9.3 w/ NH_4OH	5 mM TAPS, 5 mM $Ba(OH)_2$ pH 10 w/ NH_4OH	1.20	24	S2, S5-S22
2	10 mM $(NH_4)_2S_2O_3$, pH 9.3 w/ NH_4OH	5 mM HEPES, pH 10 w/ NH_4OH	1.04	24	S9-S23

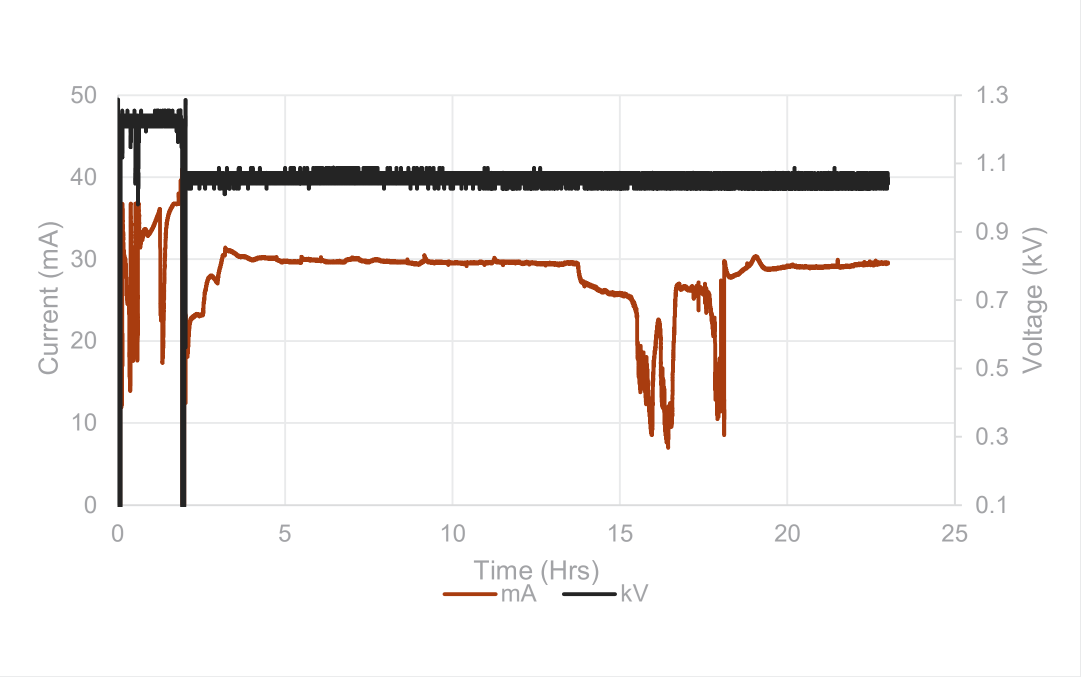

Figure 9. Current and voltage profiles recorded over a 24-hour experimental run.

4. Results for MCD and ITP

In this Sections 4.1 and 4.2, we present the separation performance measured on our MCD column and on our ITP device. In Section 4.3, we compare the two techniques and analyze scale-up considerations.

4.1. Microchannel distillation separation

We used our MCD column, which contained ~ 2 m of alumina foam packing, to separate HCl isotopes at total reflux. For our first test, we ran the column for 48 hours at a reboiler duty of 12.5 W, collecting samples periodically throughout the duration of the experiment. The isotope ratio of each sample is shown in Figure 10. At the end of the 24 hours, the $^{37}Cl/^{35}Cl$ ratio was 0.3337 at the reboiler and 0.296 at the condenser, which corresponds to a HETP of 0.84cm. Thus, 238 separation stages were achieved in the distillation column, which surpasses the goal of 50 separation stages outlined in the CRADA by nearly a factor of five.

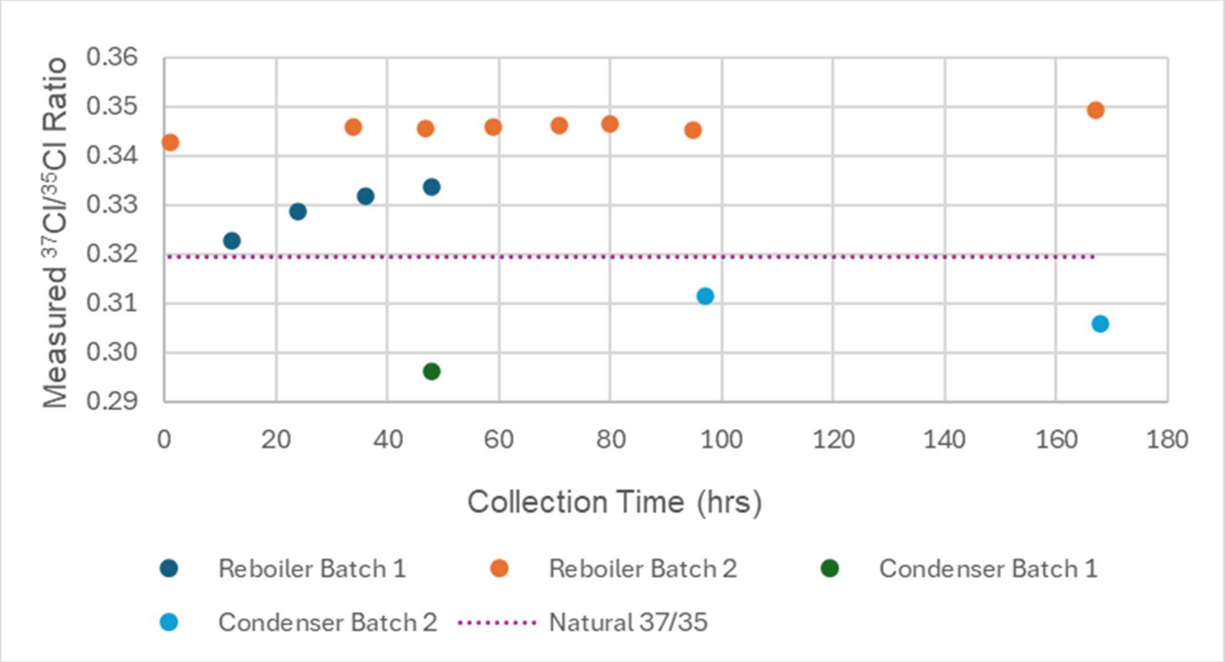

Figure 10. Measurements of the ^{37}Cl/^{35}Cl isotope ratio for samples collected during each of our two MCD runs.

For our second test, we ran the column for 168 hours to ensure the column had enough time to reach steady state. The isotope ratio of each sample is also shown in Figure 10. At the end of this experiment, we measured a ^{37}Cl/^{35}Cl ratio of 0.349 at the reboiler and 0.306 at the condenser, which corresponds to 264 separation stages and an HETP of 0.759 cm. This indicates slightly better separation than our first measurement, which is consistent with the column having reached steady state.

As stated previously, producing 90% pure ^{37}Cl from natural chlorine would take 6680 separation stages. Using our measured HETP value from the 168-hour test, we predict that we would need a column length of 50.7 m to perform this separation, which is significantly more compact than what can be achieved with any commercial packing. For comparison, argon isotopic separation has been performed on a 26 m tall column [9], and petroleum separations use columns 60 m and taller [10]. Alternatively, the separation could be achieved using multiple smaller distillation columns in series.

With additional testing, the setup may be optimized further. Our simulations suggest that decreasing the column flowrate could decrease the HETP even further; however, this was difficult to do in the current configuration because of the minimum power requirement of the condenser cryocooler. Also, it is possible that the relative volatility of the isotopic compounds could change with temperature, meaning that operating at a higher or lower pressure could plausibly improve the separation as well.

4.2. Isotachophoresis results

Figure 11 shows the ^{37}Cl/^{35}Cl ratio at different points in the column at the end of each of our two runs. Sample 0 is taken from the top of the VpHor device and sample 25 is at the bottom. The first run did not achieve significant separation; we attributed this to a leak in the dialysis membrane at the bottom of the VpHor device.

The second run achieved similar separation to our earlier batch experiments, producing a ^{37}Cl/^{35}Cl ratio of up to 0.5. However, as shown in Figure 12, the chloride was significantly lower in the column than what was expected, and so the portions that were drawn off for analysis during the run did not collect a measurable amount of sample. This means there was some unknown species collecting in the column over time and moving the chloride isotopes vertically downward. In a subsequent run this could be corrected by drawing off the unknown species as it builds up, however we did not have the time and budget to pursue this proposed additional test.

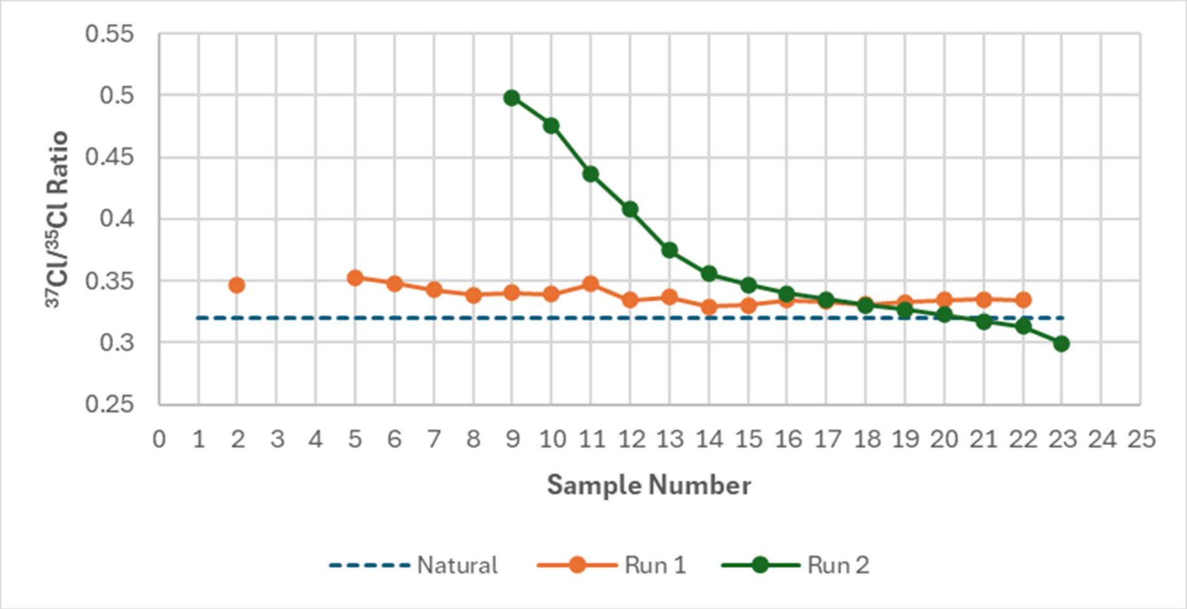

Figure 11. ^{37}Cl/^{35}Cl isotope ratio at the end of each 24-hour ITP experiment.

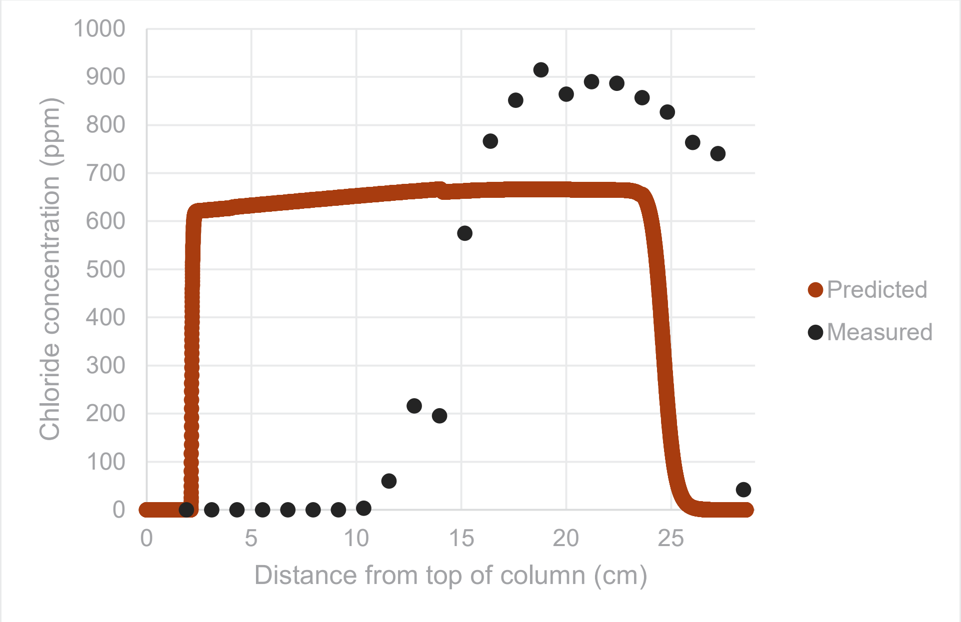

Figure 12. Chloride concentration profile predicted by our COMSOL simulations (red) and measured at the end of our second 24-hour experimental run (black).

4.3. Comparison

Both MCD and ITP achieved a significant degree of chloride isotope separation. To compare different separation methods, it is common to use the separation factor, defined as

$$q = \frac{x_f(1 - x_i)}{(1 - x_f)x_i},$$

where q is the separation factor, x_f is the final mole fraction of the component that gets enriched, and x_i is the initial mole fraction of the same component. For ITP, the separation factor gives the separation that can be achieved by a single stage. The separation that can be achieved by N stages is given by q^N.

The MCD single-stage separation factor is defined by the relative volatility, which is equal to 1.0005. Our 2 m tall MCD column achieved 264 separation stages. Since ITP does not have an equivalent definition for a separation stage, we will compare our ITP results to distillation stages so we have a way to compare the two techniques.

Our ITP device achieved a ^{37}Cl concentration of 33.3%, or a separation factor of 1.559. This is equivalent to 888 distillation stages, meaning that one 30 cm tall VpHor device gives 3.4 times more separation than a 2 m tall MCD column. However, there are likely large differences in the throughput. In our ITP experiments we withdrew 2 μL/min of 600 ppm chloride, which would be 0.63 g/yr. In our simulations, higher withdrawal rates significantly reduced the amount of separation, and so this value should be representative of the typical operational scale of our ITP separation. The MCD column was operated batch rather than continuously, so we do not have an empirical withdrawal rate to compare against, but we can still estimate a possible value for the throughput. We estimated the production rate using a mass balance for the column, combined with the Underwood equation [4] to determine the minimum reflux ratio required to enrich the ^{37}Cl to 90%, and assuming that the column's total vapor flow is fixed. Using this procedure, we estimate that a MCD column at the vapor flowrate used in our tests could produce 84 g/yr, significantly more than a single ITP device.

The MCD column and ITP device used similar amounts of power. The MCD column used 12.5 W in the reboiler and 70 W in the cryocooler. However, our cryocooler required a minimum input power of 70 W and was therefore not optimized for this distillation. Assuming that a cryocooler more appropriate for the temperature range of HCl was used, we estimate that it would use closer to 35 W, for a total power consumption of 47 W. The ITP separation ran at 1.2 kV and 30 mA, for an operating power of 36 W. Therefore, the MCD test device required slightly more power than the ITP test device. However, since the two devices achieved different amounts of separation and have different expected production rates, these power figures are intended only to indicate operating scale.

MCD is closer to being ready for deployment than ITP as well. In our ITP experiments, we identified an unknown species that built up in the device over time. It would require additional research to identify and eliminate the source of this species, or else careful control to withdraw it as it builds up, if we wanted to run the device for longer than 24 hours. We have also noticed that the pH of the LE and TE changes over time, and it would need to be regenerated or use a single-pass approach. By contrast, our MCD experiments allowed us to collect enriched ^{37}Cl without any additional issues being identified.

MCD also compares favorably to other methods of Cl isotope separation. As a point of comparison, PNNL has previously used thermal diffusion columns to separate chlorine isotopes using an imposed temperature gradient [11]. According to data collected during that research, thermal diffusion columns could perform an equivalent separation to our MCD column (i.e., a ^{37}Cl/^{35}Cl ratio of 0.334 from natural feed) using a column length of 0.31 m at a hot side temperature of 400°C, with a power consumption of 280 W. Assuming that the separation and power scale linearly with length, it would take thermal diffusion columns 24 m long that use 21 kW of power to enrich natural HCl gas to 90% pure ^{37}Cl. This is several orders of magnitude more power consumption than a MCD column, where the power does not increase with column length. Therefore, MCD would require more column length but significantly less power than thermal diffusion columns to separate chlorine isotopes to a useful level.

In summary, MCD and ITP can both be used to efficiently separate chlorine isotopes. A single ITP device gives much higher separation than a MCD column, but it is also expected to give much lower throughput given the current state of the technology. ITP would also require additional development to resolve some of the observed experimental complications.

5. Conclusion

We designed, constructed, and tested two different methods to separate chlorine isotopes: microchannel distillation, which separates species based on their relative volatility, and isotachophoresis, which separates species based on their electrophoretic mobility. It was found that a 2 m tall MCD column achieved 264 separation stages, which is far above our goal of 50 stages, and an HETP of 0.76 cm. Our ITP experiments achieved a separation factor of 1.559, which was more than 3 times higher than the separation achieved by our MCD column. However, an ITP device would likely have lower throughput, and some additional development would be required before ITP is ready to scale up, while an MCD system is closer to being ready for deployment. We conclude that both ITP and MCD are highly promising techniques for the separation of chlorine isotopes, but MCD is the more practical near-term path for high-throughput enrichment, while ITP offers a compelling longer-term route to higher selectivity if throughput and scale-up challenges can be addressed.

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