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Experimental investigation of moisture freeze-out in a cryogenic heat exchanger for helium purification

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Abstract. Purification of helium gas is vital for reliable operation of cryogenic systems operating at 4.5 K or below. Any extraneous matter will freeze, causing performance degradation or damage of cryogenic equipment. Removal of low-level (1-100 ppm_v) moisture contamination by freeze-out provides excellent control over a wide range of concentration. However, freeze-out processes require heat exchangers designed for contaminant solidification with minimal impact on flow distribution and heat exchange, and thorough understanding of frost formation on heat exchanger surfaces and the underlying heat and mass transfer. Frost formation in a cryogenic (300-80K) multi-pass tube-in-tube heat exchanger, integrated in a commercial helium purifier, is studied. A test bench incorporating the purifier and a novel low-level (5-100 ppm_v of moisture) controlled contaminated gas generator is developed. The moisture freeze-out process in the test heat exchanger is studied under real-world and controlled conditions. Transient data relating heat exchanger performance, pressure drop, and deposited frost mass are collected. The effect of moisture concentration and flow imbalance on heat exchanger performance degradation and frost deposition are investigated. A 1D numerical model developed at FRIB was used to study the frost formation profile and capacity in the test heat exchanger, and its performance degradation.

1. Introduction

Helium cryogenic systems operating at 4K (or below) require very high purity helium to operate reliably, efficiently, and safely. All other substances in the helium will freeze along the process path before reaching the operating temperatures. These solids can cause degradation of heat exchangers, create leaks through valves, and damage high-speed rotating machinery (turbines, compressors). Therefore, ensuring the helium is pure (<1 ppm_v of impurities) is of utmost importance to the safe and reliable operation of a helium refrigeration system. Impurities are introduced to the system typically during commissioning and maintenance of equipment [1], by air and moisture intrusion in sub-atmospheric systems, by new helium being added, and a refrigerator's compressor oil. Additionally, helium is most commonly available commercially for these purposes as Grade A purity (or 99.997% pure, or 30 ppm_v of impurities) [2]. The main components of this contamination are nitrogen, oxygen, volatile organic compounds, and water. Nitrogen, oxygen, and volatile organic compounds are typically removed by an adsorption process using an activated charcoal or silica bed [3]. That leaves the main challenge of helium purification, removing water, for which there are several methods. One is adsorption using a silica bed, but this has been shown to be ineffective for low-level contamination over the long term [4] and have complicated, time-consuming, and unreliable regeneration processes [5]. Freeze-out purification systems, when properly designed, can remove very low-level moisture contamination (up to fraction of a ppm_v) due to the low partial pressures of the moisture. Optimal design of the freeze-out heat exchanger in these systems is a significant factor, as it must retain a large amount of moisture, without significantly impacting heat exchange, flow distribution, and pressure drop. The design should also support easier removal of the contaminants for a quicker regeneration process. However, design procedures and associated fundamental correlations (e.g., heat and mass transfer) for these heat exchangers are very limited in the literature. Frost deposition from humid air streams with relatively high humidity (>10% relative humidity) has been studied extensively [6-8], but that from a helium stream with low levels of impurity is unavailable in the literature. Gas purifiers with the capability to remove low levels of contamination are used in very specific industries (e.g., medical, cryogenic) and are uncommon. Optimal process design of such systems can be challenging but poses the potential to carry out novel research.

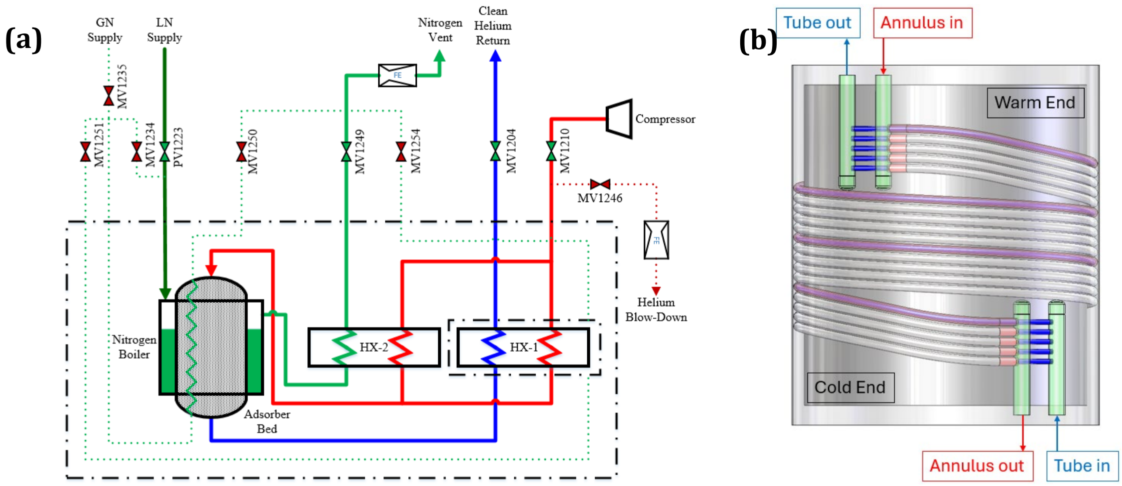

Figure 1. a) Schematic of FRIB purifier and b) Simplified geometrical model of freeze-out HX

One such purifier is available commercially. The industrial helium purifier has four main components, and several associated components for operation and collecting the relevant data. The components are a heat exchanger used for freeze-out (HX-1), a heat exchanger used to recover the cooling from the nitrogen boil-off (HX-2), a nitrogen boiler heat exchanger utilizing liquid nitrogen to cool the helium close to 80K, and an adsorber bed to remove the components of air. The air collection capacity of the purifier was estimated, but this estimation showed that it would not be the limiting factor in purifier operation (air capacity is not the limiting capacity in operation time, water capacity is) [9]. A schematic of the purifier is shown in Figure 1a. HX-2 is a relatively small heat exchanger (about 4% the size of HX-1). HX-2 can be isolated from the system in order to send all the contaminated helium through HX-1. This results in the flow thermal capacity being approximately the same on both sides of HX-1, hereon described as HX-1 being 'balanced'. While HX-2 is in operation, HX-1 is considered 'unbalanced'.

HX-1 has 10 parallel passes of coaxial tube-in-tube geometry, coiled in a helix, as represented in Figure 1b. The cold returning gas flows through the inner tubes (also called the low side), while the contaminated gas flows through the annular space between the two coaxial tubes (also called the high side). A header (green) splits the gas flow into these 10 tubes.

To understand and characterize the operating envelope and performance of a freeze-out out heat exchanger, the commercially procured helium purifiers at the Facility for Rare Isotope Beams (FRIB) at Michigan State University (MSU) are considered. They are widely used in large scale cryogenic systems for particle accelerators at several national research facilities in the United States, and are representative of the present state of the art. Although this system is not specifically designed for the freeze out process, the overall configuration of the purifier unit makes it attractive for studying moisture freeze-out in a heat exchanger. There are four such purifier systems available at FRIB. These purifiers are designed to purify up to 60 g/s (nominally at 13.0 bar) of helium. They can sustain up to several hundred ppm_v of nitrogen (air) for several days, but only a fraction of that in moisture. Liquid nitrogen is the cooling medium for 80 K adsorption of nitrogen/air and moisture collection in the 300-80K heat exchanger being used for freeze-out.

The moisture collection capacity and performance of the heat exchanger being used for freeze-out have not been precisely measured and there is no information available for how they react to different operating conditions. As such, a series of tests were conducted to measure this.

2. Testing setup

A series of tests were performed to find the capacity for moisture collection under various conditions in order to better understand the driving forces that contribute to increased moisture collection capacity and operating time and how these factors contribute to HX-1 performance.

Tests were performed under two sets of conditions. One during a nominal, practical operation of the purifier at FRIB, and the second with a controlled injection of moisture into the purifier, for component performance evaluation. The test data was analyzed to understand the influence of the various process parameters on the moisture collection capacity and operational efficiency.

2.1 Data measurement

In order to properly measure the moisture collected by the heat exchanger and the performance of the freeze-out heat exchanger, the following measurements were taken. A classical venturi flowmeter equipped with a Rosemount 3051 series differential pressure transmitter was connected to the nitrogen vent line, to measure the amount of nitrogen utility usage. The liquid nitrogen (LN) consumption rate is an indication of the cooling required by the purifier and is directly related to the operating cost of the system. There are several temperature and pressure sensors throughout the purifier, measuring the operation and characterization of the purifier components. The helium flow rate is measured on the purifier (recovery) compressor skid. The moisture collected by the purifier was measured during regeneration in a cold trap attached to the vacuum port used to pump out all the substances in the purifier at the end of an operating cycle. The frost in the cold trap is melted and its volume is measured in a graduated beaker. The inlet purity of the helium to the purification system is measured (and constantly monitored) using a digital hygrometer (Panametrics Dew.IQ) in ppm_v. The water entering the purifier was measured by gravimetric and visual (through a liquid-level port) measurements on the humid gas generator.

2.2 Humid gas generator

A controlled contamination study is required to understand the influence of the various process parameters on the moisture collection process. Controlling contamination level and flow rate allows for isolation and better study of specific variables and their effects on frost deposition in the freeze-out heat exchanger. Equipment to achieve constant and controllable low-level contamination is not available commercially. As such, a low-level moisture generator was designed and fabricated. Tests were conducted at controlled inlet moisture contamination conditions were performed to estimate the moisture collection capacity of the purifier.

A schematic diagram of the moisture generator set-up is shown in Figure 2. The recovery compressor supplies clean helium to the moisture generator from helium gas storage tanks for this test. The moisture generator set-up uses two helium streams – one 'dry' (moisture volume fraction < 0.1 ppm_v) and the other saturated with moisture. Balanced mixing of the two streams through two valves (MV112 and MV101) is used to achieve a target contamination (moisture) level (typically between 5 – 100 ppm_v). Helium bubbled through a sintered metal filter in a water-filled vessel generates the saturated helium stream. The 10 liter vessel was filled with a more than sufficient amount of water (3-5 times the projected collected mass) before each test.

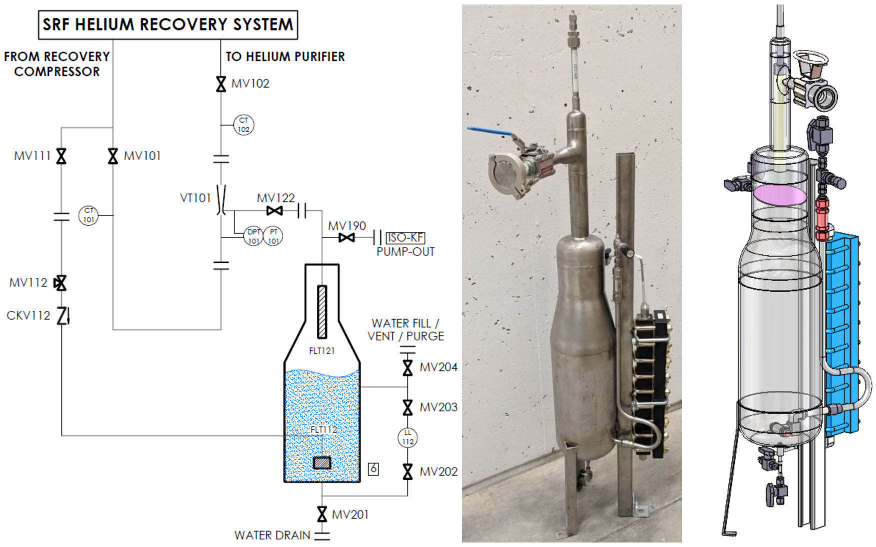

Figure 2. Schematic diagram, picture, and 3D model of a constant low-humidity gas generator setup

The water vessel and surrounding piping were fabricated at FRIB. The valves (and their flow coefficients) were selected specifically to allow for generation of a desired range of low-level moisture contamination. MV112 and MV101 finely tune the amount of helium going through the vessel. Figure 2 also shows a photograph of the humid gas generator and a 3D model of the details of the inside of the vessel. This includes the graduated liquid level viewing port (blue in the model, black in the picture), which is used to measure liquid leaving the vessel. This method of measurement was calibrated by introducing known volumes of water into the vessel and noting the readout at 150 mL intervals. The humid gas generator was tested with nitrogen, showing it can achieve the desired controlled levels of contamination. The vessel uses two sintered metal filters. The first (FLT112) splits the helium stream into small bubbles. This maximizes the total surface area of the helium, increasing the mass transfer of water into the helium bubbles. The second (FLT121) covers the helium outlet from the vessel, right above a deflector plate. These work in tandem to prevent unabsorbed water from being carried with the helium stream.

3. Results and discussion

3.1 Testing during nominal operating conditions

The first test was done during a period of maintenance on the FRIB main refrigerator. During nominal operation of the purifier the inlet contamination and necessary helium flow rate can vary widely depending on the helium being processed (from equipment being commissioned, or make-up helium being added to the system). The moisture contamination level increased as the cryogenic components warmed up, releasing impurities, sending them to the purifier. Measurements under such variable conditions are shown in Figure 3.

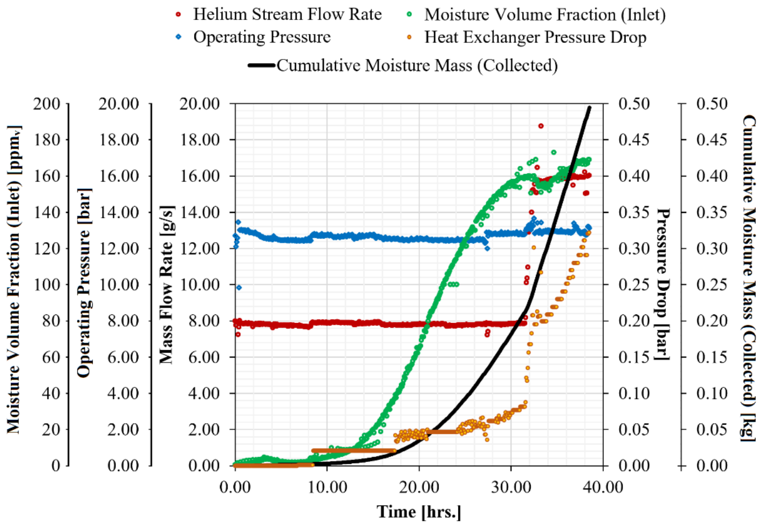

Figure 3. Testing during controlled operating conditions

The purifier was operated until it reached a pressure drop of greater than 0.25 bar, which represented the beginning of an exponential increase in pressure drop in this case, showing that the heat exchanger flow is or will soon be blocked by frost. The test resulted in approximately 0.5 kg of moisture being collected over the 38.5-hour period.

3.2 Testing during controlled operating conditions

With the use of the low-level humid gas generator, the purifier was tested under controlled operating conditions. Four tests were performed, as described in Table 1. Helium with ~30 ppm_v and ~60 ppm_v, both with balanced and unbalanced flow in HX-1 was tested. The helium mass flow rate was approximately 14 g/s, the inlet pressure was approximately 14 bar, and the starting pressure drop in the purifier was approximately 0.1 bar. Humidity level was chosen as an independent variable because it can shift where frost begins depositing by changing the partial pressure profile. A higher humidity will begin depositing at warmer temperatures. For example, 60 ppm_v of water vapor in helium will begin frosting at ~251 K, while 30 ppm_v will begin frosting at ~245 K. Flow imbalance was chosen as variable because it can shift where frost deposits in the heat exchanger by shifting the cooling curves (temperature profiles) in the heat exchanger.

The tests were run until pressure drop reached ~0.6 bar, which signifies a point in the test at which the pressure drop is suredly increasing exponentially, which means the purifier is almost plugged with frost. This increased pressure drop was possible because of the increased observation of the purifier during testing. The measurements of temperatures, pressures, pressure drop, and mass flow rates of nitrogen and helium were used to calculate performance parameters of HX-1, namely the number of transfer units (NTU). HX-1 has a base (no frost deposition) NTU of 16, then degrades throughout the tests as frost deposits on its surfaces.

The mass collected during these tests is a direct function of the contamination level, the mass flow rate, and the time. Therefore, the 60 ppm_v cases took significantly less time. Test 1 (30 ppm_v, balanced) had the most mass collection, while the other 3 cases collected roughly the same amount, although test 2 (30 ppm_v, unbalanced) had the least mass collection. Figure 4a shows the pressure drop for all four tests normalized in time (t/t_f). It shows how the slope of the pressure drop varied between the tests. The 60 ppm_v tests show the same flat, then exponential curve. The 30 ppm_v tests show a more linear increase in pressure drop, with the 30 ppm_v balanced test being the most linear. The 60 ppm_v tests used more nitrogen toward the end of the tests than the 30 ppm_v tests, further suggesting that the heat exchanger degraded more. The balanced cases used more nitrogen than the unbalanced cases, as they were not utilizing the nitrogen boil-off to cool the incoming helium and needed more nitrogen to make up for this lost cooling. This effect decreases over time, as the flow in HX-2 is blocked by frost long before HX-1, due to the frost capacity of HX-2 being relatively quite small, approximately 12 g. Therefore, the unbalanced cases become more balanced over the length of the test, which manifests in the heat exchanger duty and nitrogen vent temperature.

These observations lead to several hypotheses. First, the 30 ppm_v balanced test deposited frost more spread out throughout the heat exchanger, allowing for more area of frost deposition with less localized pressure drop. This means that the cooling curves in the heat exchanger shifted gradually over time, allowing frost to deposit over a larger heat exchanger area. Second, the 60 ppm_v cases plugged with frost more locally. This is shown by the pressure drop rising rapidly and suddenly. The more spread-out frost deposition causes pressure drop to rise gradually, as it constricts more of the flow area of the annulus over time, but evenly throughout the tube, so as not to completely block flow.

Table 1. Controlled test results

Test	Inlet Moisture [ppm_v]	HX-1 Flow [g/s]	Final NTU [-]	Final ΔP [bar]	HX-1 Mass Collected [g]	Starting N_2 flow rate [g/s]	Final N_2 flow rate [g/s]	Time of Test [hrs.]
1	32.0	Balanced	9.7	0.66	800	5.63	7.72	103.5
2	32.1	Unbalanced	5.8	0.61	538	4.02	11.74	71.4
3	68.6	Balanced	3.6	0.98	581	6.06	16.41	36.8
4	58.3	Unbalanced	3.2	0.61	591	4.15	17.39	46.3

The 30 ppm_v tests have much more variation between the balanced and unbalanced conditions than the 60 ppm_v tests because the lower real test value ppm_v (58 vs 68 ppm_v) in the balanced test counteracts the effect of the unbalanced heat exchanger. The partial pressure curve is nearer to the inlet for the 60 ppm_v tests, so it could be plugging with frost in the header when the cooling curve shifts toward the entrance to the heat exchanger over time.

The mechanical construction of the ten parallel tube sets in the heat exchanger makes a large difference on the flow distribution between the tubes and its impact on the heat exchanger performance. If the flow is not balanced between the tubes (biased by the header design), one tube may get more flow, resulting in it collecting more mass and plugging before the other tubes. If the flow is blocked in one of the tubes, the refrigeration from the low side going through that tube is wasted. This flow blockage may cause frost to build up closer and closer to the header in that tube, due to cold temperatures reaching the header. Blockages in one tube would cause the mass flow rate in the other tubes to increase, taking on the additional flow. This would cause the warm end temperature difference to increase, resulting in the frost depositing closer to the cold end of the heat exchanger over time, shifting further as more tubes plug. Flow blockage in certain tubes causes the NTU to decrease greatly, as the low side flow is wasted. This effect can be seen in tests 2, 3, and 4, in which NTU dropped much more. In order to achieve this magnitude of NTU degradation, flow distribution must be affected.

3.3 Simulation results

A one-dimensional heat and mass transfer computational model was developed to characterize the properties and performance of HX-1 [9]. It takes inputs of helium purifier inlet mass flow rate, pressure, temperature, humidity, HX-1 cold-end temperature difference, HX-1 pressure drop, nitrogen flow rate, pressure, and vent temperature. At a single point in time, it calculates all purifier temperatures, pressures, and mass flow rates (for unbalanced conditions). It then uses these inputs and calculated values to calculate heat and mass transfer, temperature profile, UA profile, NTU profile along the length of HX-1, and frost profile along the length of HX-1.

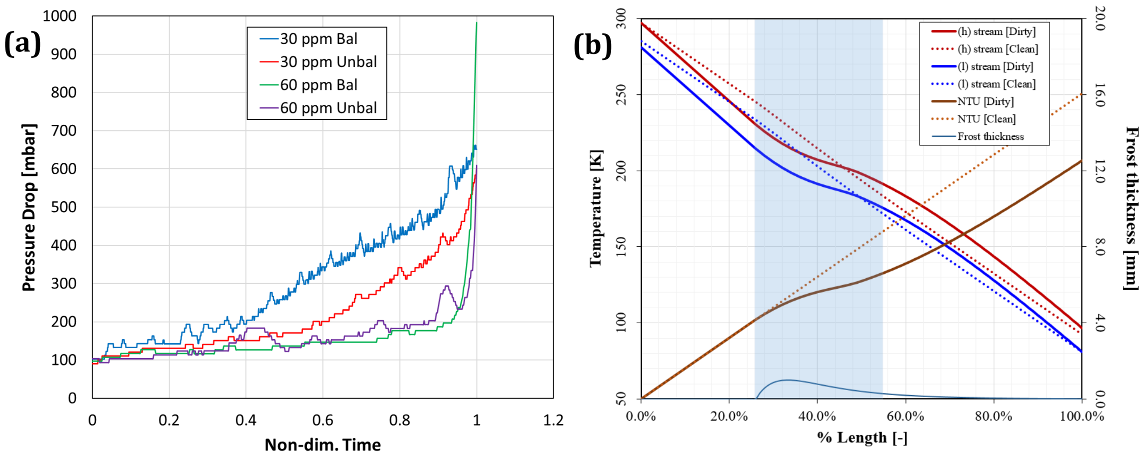

Figure 4. (a) Pressure drop normalized for time and (b) Simulation results: Gas temperatures, NTU, and frost formation profile along length of heat exchanger at beginning and end of test duration

Test 1 (30 ppm_v, balanced) was simulated using the model, with inputs from the test data. Notably the final pressure drop, which determined the maximum frost height. Figure 4b shows the HX-1 cooling curves and NTU both at the beginning (clean) and end (dirty) of the test time-period, and the max. frost thickness at the end of the test time-period. The NTU of the clean HX-1 was corrected by a factor of 0.95 to match the value of approximately 16 obtained from the test data. This accounts for HX-1 inefficiencies not accounted for in the model (heat in-leak, etc.). The model predicts the final NTU to be 10.2, compared to a measured value of 9.7. The model predicts the total mass collected to be 713g, compared to the measured value of 800g.

Other tests (2, 3, and 4) did not match as well with the simple computational model, overpredicting frost collection and underpredicting NTU degradation. This is due to flow mal-distribution that is more pronounced during those tests [9]. The model assumes equal flow distribution, which is why there is reasonable matching for case 1. This indicates that this model can predict the behavior of the heat exchanger when the real conditions match this assumption. This finding is critical in showing that flow mal-distribution is a significant factor in moisture capacity of a freeze-out heat exchanger, and explaining why the tests . A heat exchanger design that limits or removes flow mal-distribution can make better use of its available surface area and volume for collecting frost. The model is also able to show the expected region in which frosting occurs and how it shifts during different inlet conditions. Higher inlet ppm_v results in higher saturation temperature, which makes frost deposit closer to the warm end of HX-1. Increasing high-side flow (relative to low-side flow) moves the frosting region closer to the cold end of HX-1.

4. Summary

A tube-in-tube heat exchanger was tested for its ability to serve as a freeze-out heat exchanger for helium purification application. It was tested under nominal operating conditions, during commissioning of equipment at FRIB. It was found that the capacity under these unsteady conditions was approximately 500g of water. It was then tested under controlled conditions, using a humid gas generator. It works by sending helium through a bubbler in a water vessel and diluting it, and was able to produce a controllable, finely-tuned moisture contamination in a helium gas stream between 5 and 100 ppm_v. The heat exchanger was tested under controlled conditions at 30 and 60 ppm_v, with balanced and unbalanced flow. Pressures, temperatures, mass flow rates, and mass collected were measured and heat exchanger metrics including transient UA, NTU, and pressure drop were reported and discussed. During these tests, the moisture capacity of the heat exchanger was 500-800g. It was determined that this heat exchanger design has significant flow mal-distribution between the 10 tubes, which results in uneven frost deposition and premature plugging of the heat exchanger. Additionally, a one-dimensional computational model was developed to predict the behavior of the heat exchanger. It shows reasonable matching for tests with minimal flow mal-distribution. Due to its overprediction of frost collected in cases with significant flow mal-distribution, this result suggests that eliminating flow mal-distribution with operational or design changes can result in a larger heat exchanger frost collection capacity.

Acknowledgements

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