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Membrane-Based Air Dehumidification Using Organic Ionic Liquid Desiccant

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Abstract

Membrane Ionic Liquid Desiccant Air Dehumidification (MILDAD) is a promising alternative to conventional dehumidification systems that employ mechanical compressors and refrigerant-based cooling with evaporator and condensing coils. However, its application has been limited due to high manufacturing and installation cost, durability issues, concerns with corrosion due to leakage and salt carry over into the downstream air and onto surfaces. This paper reviews substantial progress made with new systems based on combining advanced ionic membranes that are pressure resistant, thin walled with high water permeability, and non-porous with synthetic organic, non-crystalline ionic liquid desiccants. Alternative design configurations are discussed comparing cross flow shell-and-tube membrane contactors employing ionic membrane tubes through which ionic liquid desiccant air flows. Performance data for MILDAD prototypes is provided. Dehumidification of the incoming supply airstream and regeneration into exhausted building air is demonstrated. The latent effectiveness of the MILDAD system will be discussed along with some future steps to improve performance of the system.

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Keywords: Ionic Liquid Desiccants, Pervaporation, Membrane ;

1. Introduction

According to U.S. Energy Information Administration [1], the world's energy consumption grew 54% between 1990 and 2015. Buildings accounted for 20% of the total energy consumption in the world, and 40% in the United States. About 55% of the energy consumed in U.S. buildings is to keep the indoor environment thermally comfortable [2]. Humidity level is one of the important indicators of the quality of the indoor air environment (IEQ), which directly affect people's productivity and health. ASHRAE standard 55-2013 and 62.1-2016 request that HVAC system must be able to maintain a humidity ratio of below 0.012, and the relative humidity should be controlled below 65% to keep a good IEQ. Dannemiller et al. (2017) and APAFrankel et al. (2012) observed that water provides a major breeding ground for the growth of bacteria and fungi in buildings, thus, affecting people's health [3][4]. In many regions, air dehumidification is necessary to reduce the moisture level in the indoor environment.

Conventional air dehumidification methods include mechanical cooling and sorption (adsorption with solid desiccant or absorption with liquid desiccant). With mechanical cooling, air is cooled down to its dew point temperature and water vapor in the air is condensed. To provide acceptable air temperature for air-conditioning or ventilation, the cold air usually needs to be warmed (reheat), which wastes energy. Sorption method uses solid or liquid desiccant to absorb water vapor in the air [5]. In conventional Liquid Desiccant Air Dehumidification (LDAD) system, the liquid desiccant is in direct contact with the wet air to be treated and absorbs water vapor in the wet air. With LDAD, the air temperature and humidity can be independently

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regulated, and the cooling system can work more energy efficiently with higher evaporating temperature. However, since air flow directly contacts with liquid desiccant in conventional LDAD systems, small solution droplets can be carried over by the air flow and spread into ductwork or the conditioned space. It not only negatively impacts the health of occupants, but also results in corrosion of the air supply ductwork. In addition, dehumidification performance of the liquid desiccant could degrade due to exposure to the pollutants in the ambient environment.

To eliminate the carry-out or entrainment of tiny desiccant droplets, we plan to employ a liquid-to-air membrane energy exchanger (LAMEE) developed at Xergy, Inc. The study reported here investigates the new membrane-based ionic liquid desiccant air dehumidification (MILDAD) system by using the aqueous solution of an ionic liquid as the desiccant. The first innovation of the system is using membrane-based mass-heat exchanger to eliminate the carry-over problem of the typical LDAD. The second innovation of the MILDAD is the use of ionic liquid as desiccant. Ionic liquid is synthesized liquid salts which are in the liquid phase when exposed to room temperature and atmospheric pressure. They have high thermal stability, negligible or no vapor pressure, high solubility in water, low or no corrosion to metals, and low driving temperatures to release water vapor (for regeneration). Ionic liquid, therefore, are ideal substitutes for traditional desiccant liquids because they can reduce or eliminate the corrosion and save the liquid desiccant used in the operation. Using Ionic liquids has potential to remarkably reduce the operation and maintenance cost, as well as improve system performance.

2. Technology Description

2.1. Membrane Contactors

A MILDAD system uses semi-permeable membranes to separate the liquid desiccant and the air so that the problem of carryover is avoided [6]. A porous membrane is a thin material with pores in it that allow some substances to penetrate through it while preventing others from entering. A non-porous membrane is a thin material that incorporates (typically a polymer) material that transmits water. The driving force between the air and the liquid desiccant is the difference in water vapor pressure on either side of the membrane. There are two main parameters to describe a membrane: water transmission rate and selectivity. The water transmission rate (sometimes referred to as “Permeability”) is the rate at which vapor permeates through a membrane per unit area or per unit driving force; selectivity is the ratio of the permeability coefficient of vapor and the others in the humid air through the membrane [5]. The degree of selectivity is largely based on the membrane properties. There are some technical challenges to using MILDAD, such as biological fouling on the membrane; low moisture permeability, poor strength, and high cost of the membrane [7]. Yang et al. (2015) reported that the uneven distribution of liquid desiccant mainly caused by membrane deflection will significantly reduce performance [8]. In this work, we report data for non-porous i.e. gas impervious membranes, developed by Xergy Inc. [9]. Xergy Inc. has developed thin, strong, advanced composite membranes and tubes based on functional polymers such as ionomers. In order to minimize membrane deflection and leakage issues (which is typical of systems incorporating flat sheet membranes) Xergy developed a tubular construct which also reduces the uneven distribution of desiccant. It should be noted that while porous membranes show higher initial rates of moisture permeability compared to non-porous (i.e. impervious or dense or composite membranes), Xergy’s non-porous membranes provide much better selectivity, eliminate cross-contamination, and no biological fouling; thus even at a lower permeation rates, they maintain stable performance and exceed the moisture permeation rates of porous membranes over time.

2.2. Ionic Liquid Desiccant

The most frequently used liquid desiccants are lithium chloride, triethylene glycol, and calcium chloride [10]. However, the traditional desiccant liquids have problems of corrosion, high cost and so on. Thus, alternative liquid desiccant is highly desirable. Ionic liquids are salts comprised of organic cations and inorganic anions or organic anions. In this study, the aqueous solution of an Ionic liquids--[EMIM][OAc]—is used as the liquid desiccant. It has high thermal stability and with low or no corrosion to metals [11]. Moreover, it does not have any crystallization problem. Fig. 1(a) shows the constant [EMIM][OAc] concentration lines in a psychrometric chart, which indicate the equilibrium conditions for air in contact with an [EMIM][OAc] solution at the specified concentrations. Fig. 1(b) shows the commonly used concentration ranges of various

liquid desiccants at 20 °C for typical air dehumidification operation, which is indicated by the blue band of equilibrium vapor pressure.

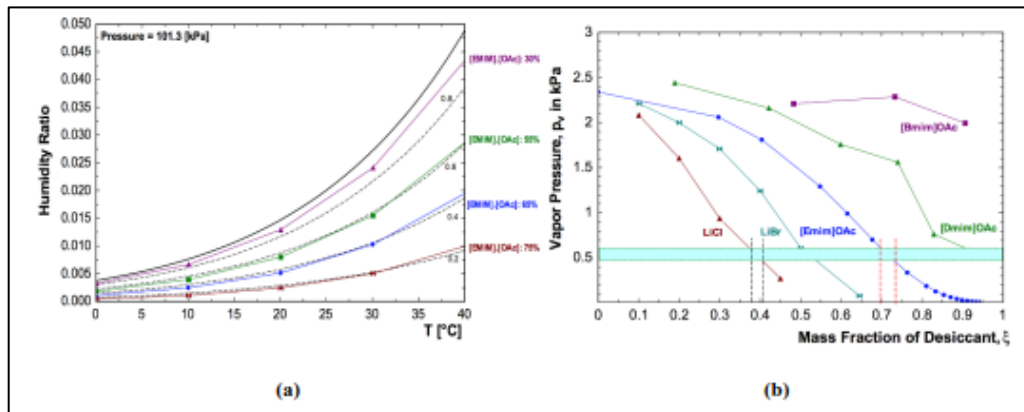


Figure 1: Properties of [EMIM][OAc] (a) dehumidification capacities at different concentrations, (b) commonly used concentration ranges of different desiccants

2.3. Membrane-based Liquid Desiccant System

Fig. 2 shows the construction of a counter-flow membrane-based liquid desiccant air dehumidification system. A MILDAD is generally comprised of two supply air streams and two modules—a dehumidifier and a regenerator. A liquid desiccant solution is pumped through the permeate side channels, and an air stream is drawn to the feed side channels at the same time. Because of the difference between the temperature and humidity of the solution stream and air stream, heat and mass are exchanged through the membrane. In this process, the concentration of the solution is reduced by absorbing water vapor. Meanwhile, the solution absorbs heat generated by phase transformation of water vapor and then its temperature rises. The water vapor pressure on the surface of the solution increases with the decrease of the concentration of liquid desiccant and the increase of temperature, and the moisture absorption capacity of the solution decreases. The hot dilute solution is pumped into the regenerator to exchange heat and mass with a hot and dry air stream to evaporate the moisture. After that, the solution becomes concentrated again, and the vapor pressure becomes lower as well. It will be sent to the dehumidification module to keep continuous operation of the MILDAD.

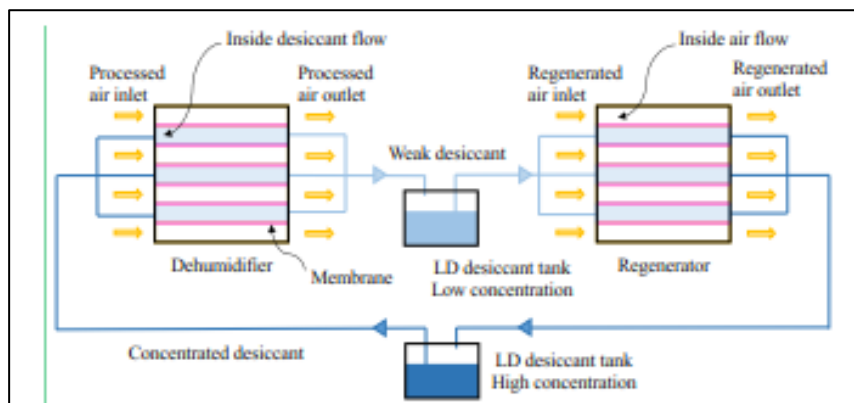


Figure 2: Schematic diagram of basic procedure of membrane-based liquid desiccant system

3. Design of a Small-Scale Prototype MILDAD

A small-scale prototype MLDAD was developed by Xergy. It uses a bundle of membrane tubes made with an ionic (nonporous) membrane instead of multiple membrane plates as in the referenced studies. The membrane tubes have better mechanical strength than the membrane plates and thus are expected to work at higher flow rates on both the air and solution sides of the membrane without any leakage. The schematic of the new prototype is shown in Figure 3.

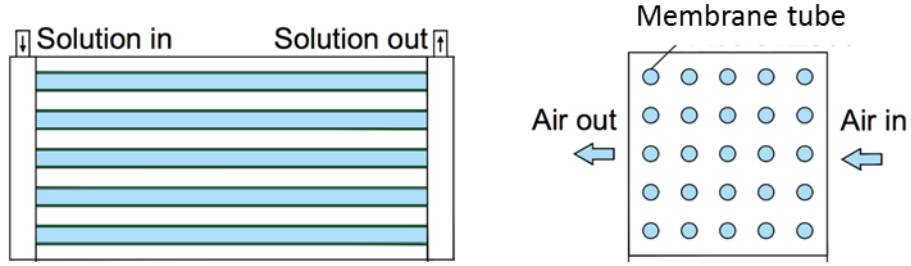


Figure 3. Concept design of a MILDAD using membrane tubes: a side view (left) and a section view (right).

The small-scale MILDAD prototype is shown in Figure 4. It measures 2.5 inches (64 mm) long, 4 inches (102 mm) wide, 4 inches (102 mm) high. It has 26 tubes made with an ionic (nonporous) membrane. The tubes are laid out in a staggered arrangement. Each tube has an outer diameter of 0.13 inch (3.3 mm) and an inner diameter of 1/8 inch (3.2 mm). The distance between two adjacent tubes is 1/4 inch (6.4 mm). The staggered arrangement could provide better convection heat and mass transfer but could also result in a higher air-side pressure drop. The end of each tube is connected to a header containing ILD solution. The ILD solution flows inside the tubes from the top header to the bottom header, while air is drawn through the outer surface of the tubes perpendicularly to the solution flow to exchange heat and water vapor with the ILD solution, forming a cross-flow pattern.

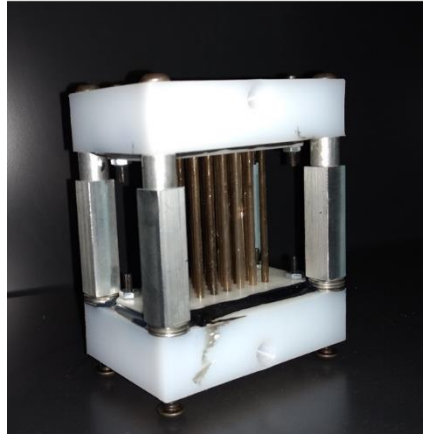


Figure 4. A small-scale prototype of the MILDAD built by Xergy Inc.

4. Experiments

The performance of both the dehumidification and humidification (regeneration) of the small-scale prototype MILDAD was evaluated at Oak Ridge National Laboratory (ORNL) under various operating conditions, including inlet air temperature and humidity, air and ILD solution flow rate, and ILD solution temperature.

Several performance metrics are evaluated based on the measured data, including moisture removal rate (m_v), specific vapor transportation rate (J), effective permeance (P), latent cooling (Q), and latent effectiveness (ϵ_L).

The moisture removal rate (m_v) is the product of the absolute humidity difference (ΔAH) and the mass flow rate of air (m_{air}), which represents the quantity of water vapor removed within a unit time, as expressed in Eq. (1).

$$\dot{m}_v = \Delta AH \cdot \dot{m}_{air} \quad (1)$$

The specific vapor transportation rate (J) is expressed with Eq. (2):

$$J = \frac{\dot{m}_v}{A_{eff}} \quad (2)$$

where m_v is the moisture removal rate, and A_{eff} is the effective contact area.

The effective permeance of the MILDAD (P) is calculated with Eq. (3).

$$P = \frac{J}{\Delta P_v} \quad (3)$$

where ΔP_v is the vapor pressure difference between two surfaces of the membrane. The latent cooling capacity of the MILDAD (Q) is calculated with Eq. (4):

$$Q = \dot{m}_v \times L \quad (4)$$

where L is the latent heat of vaporization of water, 2260 J/g at 20 °C.

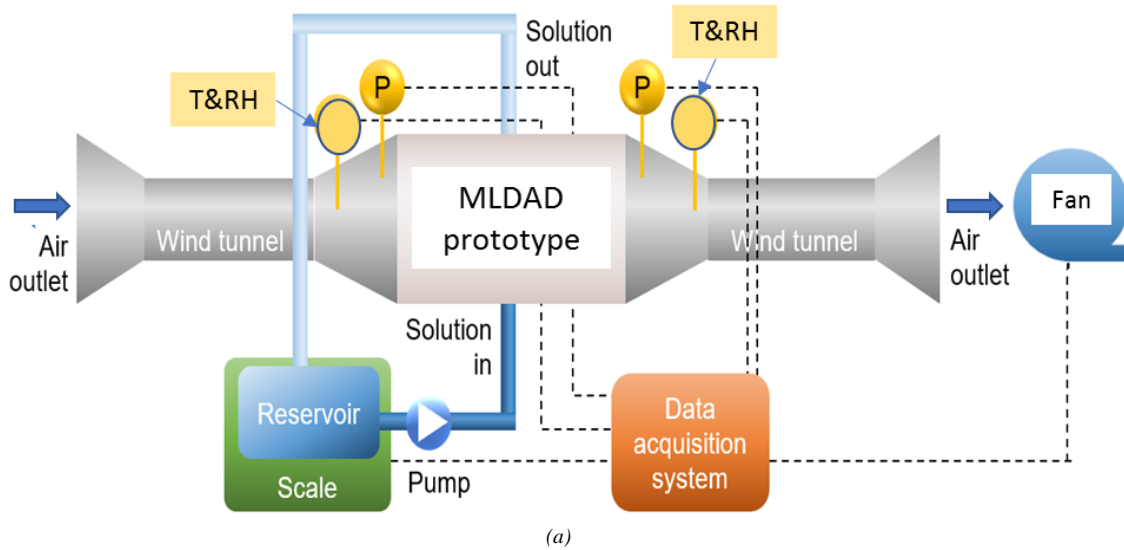
The latent effectiveness (ε_{lat}) is the ratio of the actual to the maximum possible water vapor transfer rates of the MILDAD, as expressed with Eq. (5):

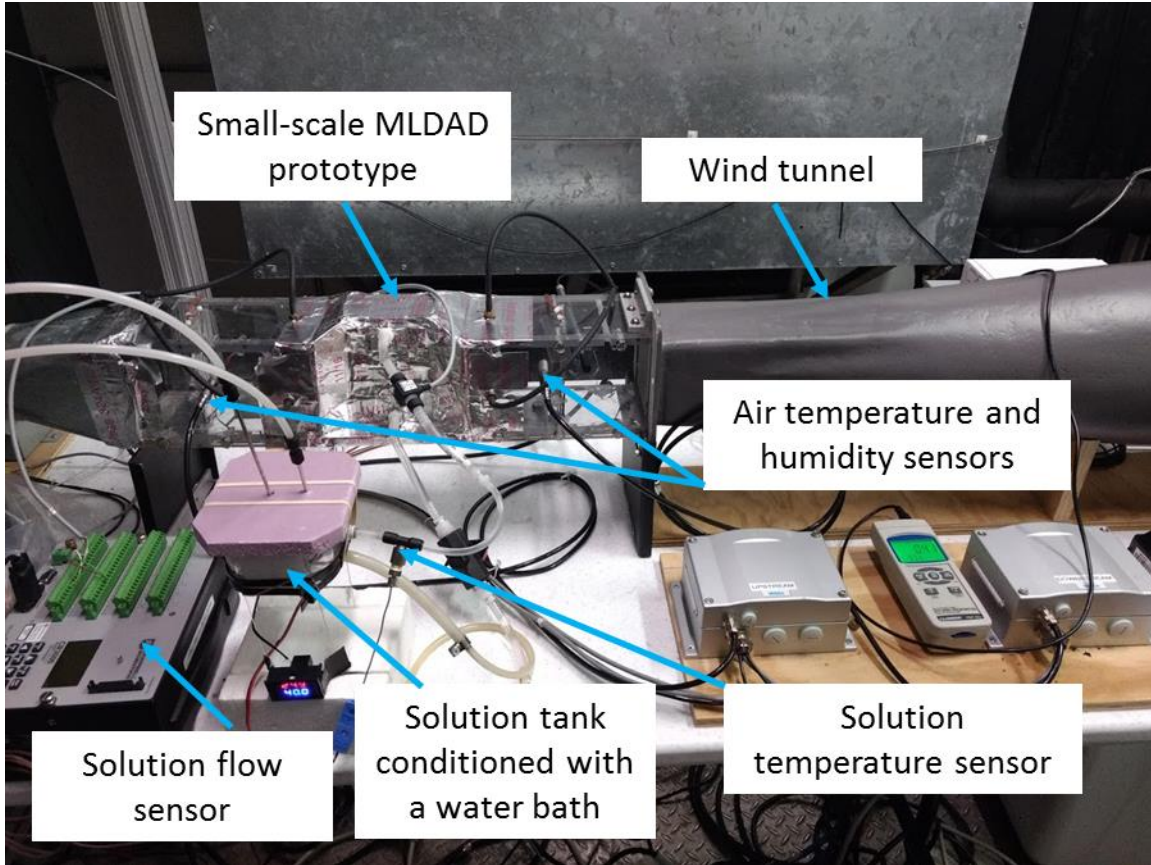
$$\varepsilon_{lat} = \frac{w_{air,in} - w_{air,out}}{w_{air,in} - w_{sol,in}}, \quad (5)$$

where w is the humidity ratio referring to the moisture content in the air (i.e., grams of water vapor in each kilogram of dry air), and for liquid desiccant, w_{sol} represents the equilibrium humidity ratio of the liquid desiccant solution, which is determined by the temperature and concentration of a given liquid desiccant solution.

4.1. Experimental Apparatus

Figure 5 shows the experimental apparatus for the tests, which were performed in a climate chamber at ORNL. The prototype was placed in the middle of an Omega WT4401-D wind tunnel, which measures the airflow rate going through the prototype based on the measured pressure drop across the prototype. The airflow rate can be adjusted by regulating the wind tunnel's variable speed fan. The wind tunnel was in a climate chamber, in which the air temperature and relative humidity could be maintained within a small range ($\pm 0.5^\circ\text{C}$ temperature and $\pm 1\%$ relative humidity) of specific conditions. During a test, room air in the precisely controlled climate chamber was drawn through the small-scale prototype MILDAD. The air pressure, temperature, and relative humidity at the inlet and outlet of the MILDAD were measured by individual sensors. The ILD solution was circulated through the prototype at a fixed flow rate by a solution pump. A small reservoir was placed in the solution loop. The solution temperature in the reservoir was maintained at a specified setpoint by a refrigerated/heated circulating water bath. A high solution electronic scale was applied to weigh the solution reservoir in order to calculate the vapor absorbed by the ILD solution. A data acquisition system was used to collect and save data from the sensors during each test. The measurement devices and corresponding accuracies are shown in Table 1.





(b)

Figure 5. The experimental apparatus for testing a small-scale MILDAD prototype: (a) schematic diagram; and (b) actual setup

Table 1. Sensor Specifications

Target parameters	Measurement devices	Corresponding accuracies	Range
Temperature	Vaisala HMT330	± 0.25 C	-15~ +40 C
Relative humidity	Vaisala HMT330	$\pm (1.0 + 0.008 \times \text{reading})$ %	0 – 95%
Differential air pressure	Omega PX02K1-16A5T	0.25%	0 – 32 in. Hg
Scale	Torbil AG4000	0.01 gram	4,000 grams

4.2. Test Results of the small-scale prototype

The tests were separated into the following five groups based on the conditions of the solution in the membrane tubes and the room air in the climate chamber. Each group included two to three tests with different airflow rates. Test results are summarized in the Table 2. The results of test group 5 were not included because leakage of the ILD solution was observed during the tests.

- **Case 1:** Humidification (membrane tubes filled with pure water) at 35°C and 20% relative humidity (RH) chamber condition, with 0.49 liters per minute (LPM) solution flow rate
- **Case 2:** Humidification (membrane tubes filled with pure water) at 25°C and 80% RH chamber condition, with 0.49 LPM solution flow rate
- **Case 3:** Humidification (membrane tubes filled with pure water) at 25°C and 80% RH chamber condition with lower water flow, with 0.30 LPM solution flow rate
- **Case 4:** Dehumidification (membrane tubes filled with EMIMOAc solution with 75% concentration) at 25°C and 80% RH chamber condition, with 0.15 LPM solution flow rate
- **Case 5:** Regeneration (membrane tubes filled with EMIMOAc solution with 75% concentration) at 35°C and 20% RH chamber condition, with 0.30 LPM solution flow rate

The test results indicate that the small-scale prototype MILDAD can humidify (Cases 1 through 3) and dehumidify (Case 4) air. The specific vapor transportation rate ranged from 53 to 105 g/(h-m²) for humidification, and it was between 32 to 47 g/(h-m²) for dehumidification. Figure 6 shows that the specific vapor transportation rate of the MILDAD was affected by the airflow rates, but the trend was determined by

other factors, including chamber condition and solution flow rate. For humidification, when the chamber condition was very dry (Case 1), increasing the airflow resulted in a decrease in the specific vapor transportation rate. However, when the chamber condition was humid (Cases 2 and 3), the specific vapor transportation rate was not as sensitive to the airflow rate as it was under dry condition. Furthermore, under the same chamber condition and with the same airflow rate, lowering the solution flow rate decreased the specific vapor transportation rate. For dehumidification (Case 4), increasing the airflow from 4.6 to 10.6 m³/h increased the vapor transportation rate, but further increases in the airflow rate resulted in a decrease in the vapor transportation rate.

The measured overall permeance of the small-scale prototype MILDAD ranged from 0.4 to 0.6 E-8 kg/(m²-s-Pa) in dehumidification mode and from 0.4 to 2.9 E-8 kg/(m²-s-Pa) in humidification mode.

Table 2. Test conditions and results

Case	Test Conditions (Measured Data)								Measured Performance	
	Air				Air Flow		Solution		Permeance kg/(m ² -s-Pa)	Specific Vapor transportation Rate g/(hr-m ²)
	T _{Air,in} C	T _{Air,out} C	RH _{Air,in} %	RH _{Air,out} %	Rate CFM	T _{Sol} C	Con _{Sol} %	Flow Rate GPM		
Case 1-1	35.22	35.15	19.37	19.81	4.8	35	0	0.13	4.3E-09	74.4
Case 1-2	35.25	35.12	19.57	20.27	3.3	35	0	0.13	4.5E-09	77.8
Case 1-3	35.27	35.11	19.52	21.04	1.8	35	0	0.13	6.1E-09	105.1
Case 2-1	25.14	25.10	79.21	80.28	2.9	25	0	0.13	2.6E-08	63.4
Case 2-2	25.14	25.09	79.84	80.51	6.0	25	0	0.13	2.7E-08	64.2
Case 2-3	25.20	25.16	80.21	80.71	8.7	25	0	0.13	2.9E-08	66.5
Case 3-1	25.13	25.09	80.07	80.55	8.6	25	0	0.08	2.3E-08	53.4
Case 3-2	25.07	25.03	80.02	80.66	5.9	25	0	0.08	2.6E-08	62.9
Case 4-1	25.07	25.51	79.81	77.21	2.7	25	75	0.04	4.8E-09	36.1
Case 4-2	25.6	25.36	79.49	78.26	6.3	25	75	0.04	6.2E-09	46.7
Case 4-3	25.20	25.44	79.92	78.65	8.5	25	75	0.04	4.2E-09	32.2

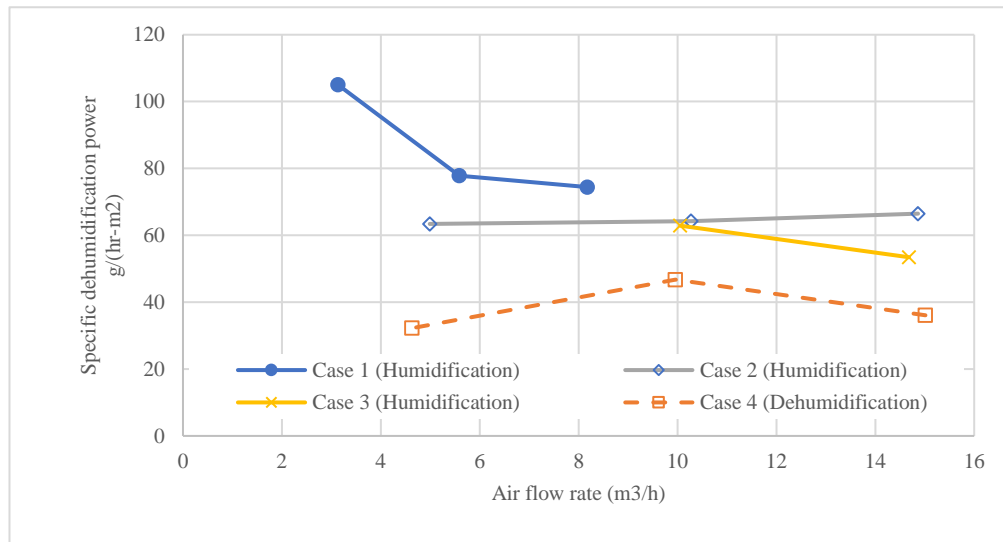


Figure 6. Measured specific vapor transportation rate of the small-scale MILDAD prototype in dehumidification and humidification operation at various airflow rates.

4.3. Test results of a medium-scale prototype

A medium-scale MILDAD prototype is tested in a wind tunnel built at Xergy, Inc as shown in Fig. 7. The air velocity; and temperature, relative humidity at the inlet and outlet of the MILDAD are measured. A data acquisition system is used to collect and save data from all sensors during each test. The measurement devices and corresponding accuracies are shown in Table 3. The volumetric flow rate of the system is calculated by multiplying the cross-sectional area of the air channel with the air velocity. To smooth the air circulation in

the test chamber, louvers are added at the air-side inlet and outlet of the MILDAD. The air circulation inside the wind tunnel is maintained at a rate of 10 CFM (0.28 m³/min) with a fan. Humidity was measured both upstream and downstream of the MILDAD prototype.



Figure 7: Experimental Apparatus for characterizing the performance a medium-scale MILDAD prototype

Table 3. The measurement devices and corresponding accuracies

Target parameters	Measurement devices	Corresponding accuracies	Range
Temperature	Vaisala HMT330	± 0.25 F	-15 ... +40 F
Relative humidity	Vaisala HMT330	$\pm (1.0 + 0.008 \times \text{reading})$ %	0 – 95%
Flow Rate	PerfectPrime WD9829, Precise Sensitive Hotwire Thermal Anemometer Probe	Resolution: 0.01 m/s $\pm (0.05 \times \text{reading})$	0.1 ~25.0 m/s

The results of the dehumidification test are as shown in Table 4. The relative humidity upstream and downstream of the MILDAD prototype are plotted with respect to time as shown in Fig. 8. The specific vapor transportation rate of the medium-scale prototype, 111 g/(m²-hr), is about 3 time higher than that of the small-scale prototype. It is thought to be due to higher flow rate and the longer tube length, which increase the contacting time for heat and mass exchange between ILD and air.

Table 4. Results from dehumidification test

Total testing time*	79.33	minutes
Air Flow Rate	0.28	m ³ /min
Total Volume of Air Processed	22.5	m ³
Average Δ AH	1.3	g/m ³
Total Moisture Removed*	29.68	g
Specific Water Removal Rate	0.03	g/m ² /s
Cumulative latent cooling	72,843	J
Latent cooling power	15.3	W
Power consumption of fan and pump	14.4	W
Estimated COP of latent cooling	1.06	W/W

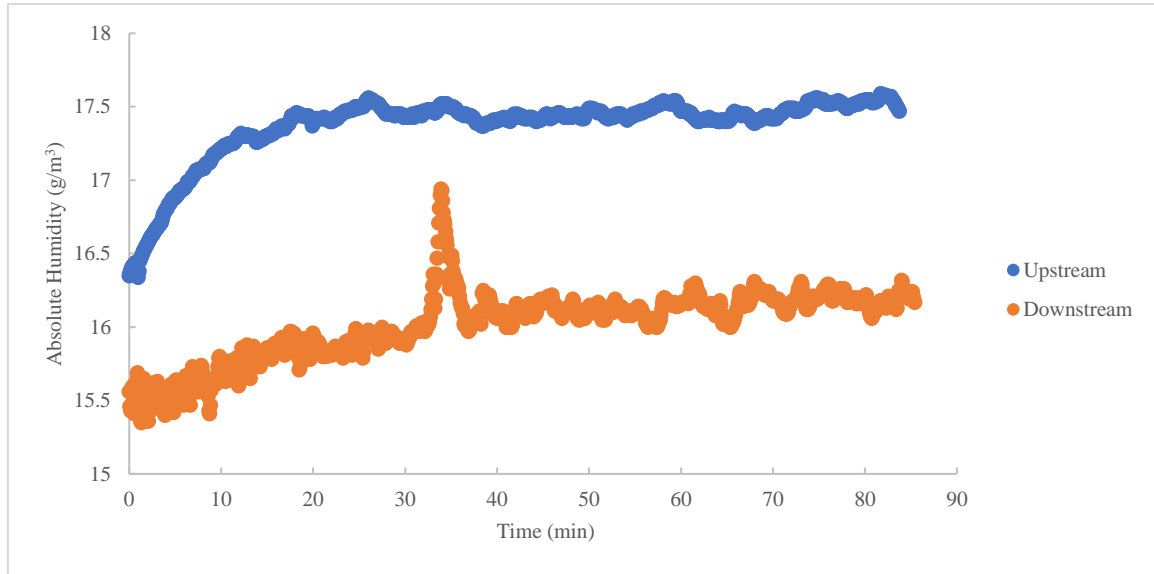


Figure 8: Dehumidification performance of dehumidifier module at 0.28 m³/min. *Note that the spike in AH between 30 and 40 min was caused by a leak in the system. This leak was resolved during the test and these data points were disregarded in the analysis.

5. Design and Expected Performance of a Full-sized MILDAD

It is calculated that 1,371 membrane tubes are needed to have the same latent cooling capacity of a conventional 5,000 Btu (1.5 kW) air-conditioner—0.3 kW latent cooling capacity, or 0.12 g/s water vapor removal rate. Dimensions of a MILDAD with 0.3 kW latent cooling capacity are listed in Table 5.

The humidity ratio of typical entering air, $w_{air,in}$, of a room air-conditioner is 10.95 g/kg (26.7°C and 50% relative humidity). The airflow rate of a 5,000 Btu (1.5 kW) air-conditioner is 170 CFM, or 0.095 kg/s. With a 0.12 g/s water vapor removal rate, the humidity ratio of the exiting air ($w_{air,out}$) will be reduced to 9.69 g/kg. The equilibrium humidity ratio of an ILD solution ($w_{sol,in}$) at 68% concentration and 26°C is 6.53 g/kg. Based on these values, the latent effectiveness of the full-size MLDAD is calculated as 0.3.

Table 5. Dimensions of a MILDAD with 0.3 kW latent cooling capacity

Overall permeance	0.000027	g/(m ² -s-Pa)
Vapor pressure difference	700	Pa
Water removal rate	0.0189	g/(m ² -s)
Total membrane surface area	6.5	m ²
Tube diameter	0.003	m
Tube length	0.457	m
Surface area per tube	0.005	m ²
Total number of tubes	1371	[-]
Tube length	0.457	m
Tube spacing (2× dia.)	0.007	m
MILDAD width	0.762	m
MILDAD height	0.076	m

6. Conclusions

Small- and medium-scale prototypes of MILDAD using hollow tubes made with nonporous membranes and an ILD was successfully built by Xergy Inc. The MILDAD prototypes use two unique technologies. One is nonporous membranes, which offer some advantages over porous membranes commonly used in previous studies, including higher selectivity for water vapor (eliminating penetration of liquid desiccant through the membrane). The other is the ionic liquid desiccant (ILD), which is a synthesized salt in the liquid phase. The ILD used in this study is [EMIM][OAc], which has a high dehumidification capacity, is noncorrosive to metals, and does not crystallize even at very low temperatures.

The small-scale prototype was evaluated at ORNL for both dehumidification and humidification (regeneration) performance under various operating conditions. The test results indicated that the small-scale prototype MILDAD can effectively humidify and dehumidify air. The specific vapor transportation rate ranged from 53 to 105 g/(h-m²) for humidification, and it was between 32 to 47 g/(h-m²) for dehumidification, depending on operating conditions. The measured permeance of the prototype ranged from 0.4 to 0.6 E-8 kg/(m²-s-Pa) in dehumidification mode and from 0.4 to 2.9 E-8 kg/(m²-s-Pa) in humidification mode.

The specific vapor transportation rate of the medium-scale prototype, 144 g/(h-m²), which is about 3 times higher than that of the small-scale prototype. It is thought to be due to higher flow rate and the longer tube length, which increase the contacting time for more heat and mass exchange between ILD and air.

Based on the measured permeance, it was calculated that a full-size MILDAD unit, which has 1,371 membrane tubes, could meet the latent cooling load of a 5,000 Btu (1.5 kW) air-conditioner with a 0.3 latent effectiveness.

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