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Use of Membrane Transport in an Absorption Thermal Transfer Cycle

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A thermal transfer cycle utilizing membrane osmotic transport of water against a pressure rise is investigated from the viewpoint of the operation of a conventional absorption heat pump using an aqueous solution as the working fluid. Physical sorption, similar or equivalent to condensation, of water vapor in the membrane material is considered to be an essential step in the overall process of water transport. The thermal nature of this step, during which the heat of sorption similar in amount to the heat of condensation for water vapor must evolve at the evaporator temperature or lower, disqualifies the system in performance as a heat pump. Simple flow relations for the aqueous sugar solution are derived under simplifying assumptions. A set of numerical calculations is given to show that the inferred steps are well within the limits of thermodynamics.

Nomenclature

A = flow area, m^2
D = hydraulic diameter, m
f = friction factor
g = 980 cm/s^2
h = enthalpy, kJ/kg
l = total height of standing structure, m
l_1 = module equivalent length for upward flow, m
l_2 = module equivalent length for downward flow, m
l_m = vertical height of membrane module, m
m = mass flow rate, g/s
p = absolute pressure, kPa
Q_e = evaporator heat input, W
Q_g = generator heat input, W
Re = Reynolds number
s = entropy, kJ/kg-K
T = temperature, K
V = flow velocity, m/s
v = specific volume, m^3/kg
μ = viscosity, centipoise
ρ = density, kg/m^3
Φ = availability, kJ/kgmole

Subscripts

a = membrane surface facing evaporator
b = membrane surface facing solution
w = liquid water
1 = upward flow
2 = downward flow

Introduction

A CONVENTIONAL absorption heat pump cycle using a binary aqueous solution as the working fluid with the solvent water performing as the refrigerant is considered. The aqueous solution can be either ionic or nonionic. A description of the working details of this cycle is given in most textbooks on refrigeration and air conditioning.[1] A scheme had been proposed to modify this cycle by making use of the thermodynamic phenomenon of osmotic flow of the solvent that takes place against an adverse pressure difference from the compartment of the pure water (the evaporator), across a semipermeable membrane that allows only water to permeate through, and into the compartment of the aqueous solution (the generator). The motivation behind the attempted modification was to remove the power input required for mechanically pumping the aqueous solution in the conventional cycle, an obviously desirable simplification from the viewpoint of system operation.

The use of osmosis to pump the refrigerant in a heat pump cycle is certainly an innovative idea in its own right. Whether such a cycle could operate to pump heat with a temperature lift, however, remains to be proved in thermodynamic principles. An experimental unit having such a combination has been investigated,[2] and it is of significant interest to explore from the viewpoint of its potential for possible space thermal management applications. The objective of this work is to find out if the osmotically driven thermal transfer cycle obtained by operating the experimental unit could be identified as a heat pump cycle.

Experimental Osmotic Unit

The experimental osmotic thermal transfer unit assembled specifically for the purpose of performing process feasibility studies is shown in Fig. 1. The essential components of the unit are the generator, the condenser, the expansion valve, the evaporator, and the membrane pumping module. The first four components are essentially the same both in nature and in function as those in a standard absorption refrigeration cycle; however, the membrane pumping module takes the place of the refrigerant absorber and the solution circulating pump. The space within the membrane module is divided into two distinct, mutually exclusive compartments separated by

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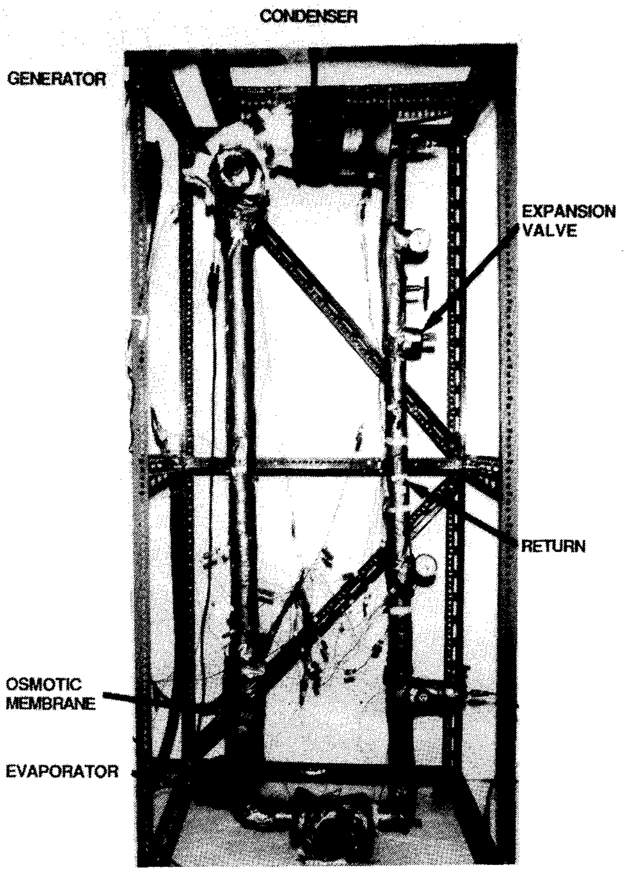

Fig. 1 Physical setup of experimental osmotic thermal transfer unit.

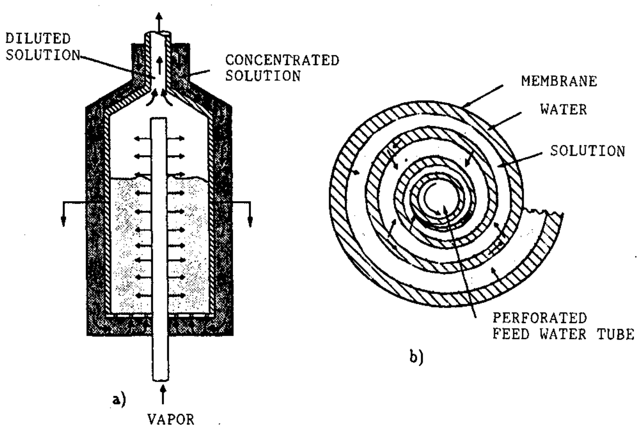

Fig. 2 Schematic illustration: a) osmotic membrane module and b) spirally wrapped cellulose triacetate membranes.

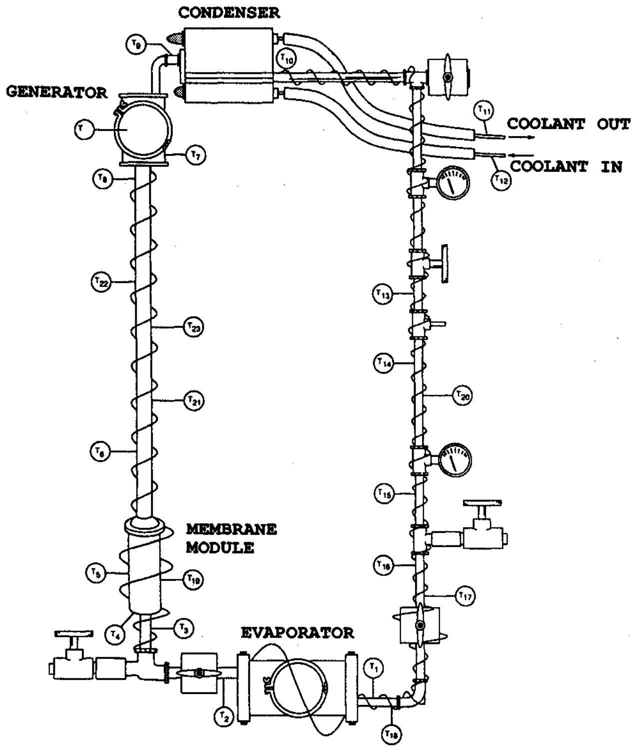

Fig. 3 Schematic diagram of experimental unit showing functional components and locations of thermocouples.

cellulose triacetate membranes through which only permeation of pure water is possible. One compartment is connected to the generator by a standing concentric double-pipe structure for facilitating the circulation by natural convection, which is created by a density difference due to either a change in solute concentration or a change in temperature or both of an aqueous sugar solution with a molality very close to being unity. The other compartment of the membrane module is spatially in open communication with the evaporator of the thermal transfer unit.

Water vapor boils off from the solution in the generator by receiving heat from a high temperature source. It then changes into liquid water in the condenser by heat transfer to the condenser cooling fluid. The condensate throttles across the expansion valve and a liquid-vapor mixture at a lower temperature is formed. In the evaporator the liquid water evaporates readily by heat absorption and the resulting vapor flows into the module where it is pumped by osmosis action across the cellulose triacetate membrane, mixed with the circulating flow of the solution, and carried back into the generator, thus completing the mass transport cycle of the refrigerant water. The aqueous solution serving only as a refrigerant carrier is confined in circulating flow within the space formed by the generator, the standing double pipe, and the solution compartment of the membrane module. Transparent plexiglass windows are mounted on the walls of the generator and evaporator for observing the liquid levels, which are visually estimated by using a linear scale.

A sketch of the membrane pumping module and the flowpath of the aqueous sugar solution is shown in Fig. 2a and the geometric arrangement of the cellulose triacetate membranes wrapped around the perforated plastic tube that feeds water vapor to the module is shown in Fig. 2b. Two separate sheets of the membrane, each combined with a sheet of plastic mesh to provide rigid support, are used in the wrapping structure. The top and bottom edges of these membrane sheets are joined together (see Drolen et al.[2] for details) so the liquid solution can freely flow upward vertically through the membrane container while the water vapor, fed via the perforated tube, which seems to be trapped within the space between neighboring sheets of membrane, can flow across the membranes only by osmotic action.

Figure 3, which is reproduced from the report by Drolen et al.,[2] is a sketch of the experimental setup after the system is stripped of all external insulation. The sketch shows the locations of the thermocouples used to monitor the fluid temperatures. All of the thermocouples have self-adhesive backings and are tape-mounted on the external surface of the metal walls, except at points 7, 11, and 12 where the probes are made directly to sense the fluid temperatures. The generator and the evaporator are equipped with electric heaters for facilitating controlled heat input. Polyglycol is used as the condenser cooling fluid and its temeprature at the condenser inlet is fixed by a constant temperature heat bath. The temperature of the latter can be adjusted and is fixed at a value in order to give the desired water vapor pressure in the condenser. The pressure gauges are of the Bourdon-tube type giving both positive and vacuum readings, and Swagelok on-off service valves and fittings are used throughout the system. A fine metering valve for performing the throttling process, however, had to be employed in order to sustain a proper flow of the refrigerant into the evaporator.[2]

Membrane Transport of Water Vapor

Because the components of the modified unit are the same as those of an absorption refrigeration cycle, except for the membrane pumping module, it is only necessary to ascertain the process characteristics of the latter in order to see if the performance of such a unit would qualify as a heat pump cycle. The transport of water across the membrane from the evaporator side, where water exists in the form of bulk vapor, to the solution side, where liquid water prevails, is believed to occur in the following steps: 1) physical adsorption of water vapor onto the membrane surface; 2) adsorbed water dissolves and diffuses in the membrane material to get transported to the other side; and 3) separation of dissolved water from the membrane to mix with the circulating aqueous solution. Adsorption of vapor on a surface is a spontaneous process and, therefore, is accompanied by a decrease in the free energy of the system. Because the process involves a loss of degrees of freedom (primarily normal to the surface) of the vapor in passing from the free bulk vapor state to the adsorbed state confined on the surface, there is also a decrease in entropy.[3] Therefore, it follows ($\Delta H = \Delta F + T\Delta S$ with $\Delta F < 0$ and $\Delta S < 0$) that heat is evolved in the transformation and, without exception, all physical adsorption processes must always be exothermic. The heat released ($-\Delta H$) is generally referred to as the heat of sorption.

Sorption of water vapor in organic polymers and colloidal fibers has been experimentally investigated by many workers.[4–6] Physical data showed that initially the heat of sorption either increases or decreases, depending on the nature of the forces involved, as the uptake of water goes up in the material. After sufficient water is sorbed, however, the heat of sorption becomes independent of the nature of the solid material but comes close to the heat of condensation for water vapor.[6] This suggests that, at high water uptake in the polymer, sorption is like condensation and, other than the difference in entropy, the behavior of the water adsorbed should resemble that of a liquid. This is true for all the polymer materials used in the experiments reported by these authors, particularly cellulose triacetate.

The cellulose triacetate polymer in the membrane module of the Hughes thermal transfer unit must be at a state of total imbibition in liquid water, perhaps with a significant degree of swelling, because the membrane, which is permeable to water is in direct physical contact with the aqueous solution on one side. Rapid simultaneous heat and mass transfer take place incessantly between the liquid-like water on the surface of the membrane material and the water vapor on the side of the evaporator. It seems inevitable that the vapor must be characterized by thermal equilibrium with the membrane at the interface before sorption can take place. The experimental observation above suggests that the water vapor must first cool to saturation and then condense upon the membrane by releasing the heat of condensation and the adsorbed water, which is like a liquid in physical behavior is transported to mix with the aqueous solution. The process is spontaneous and the vapor is in thermal communication only with the aqueous solution via the membrane. Therefore, to accommodate the attendant heat transfer, such a process is possible if the membrane (and, thus, the solution) is at a temperature lower than that of the approaching vapor. This is borne out by the simple fact that flow of water across the membrane has never been observed to take place under a condition in which the system is in thermal equilibrium; i.e., when the aqueous solution, the membrane, and the water in the evaporator are all at the same temperature. It is also demonstrated by Drolen, et al.[2] who noted from the results of their hot membrane experiments that "as the membrane temperature became closer to the evapoator temperature, the osmotic pumping started slowing down and eventually stopped."

In order to accomplish membrane pumping, the water in the evaporator must be continuously heated or maintained at an elevated temperature. Figure 4 shows a typical temperature-time tracing under the condition of a constant evaporator heat input of 40W and a generator input of zero. The entire system, except the membrane module and the plexigalss viewports, was blanketed with a layer of Rubatex® closed-cell insulation measuring 1.9 cm (¾″) thick. Initially the system was in thermal equilibrium at a room temperature of about 23°C. The onset of membrane pumping, marked by the letter A in the graph, occurred about one full hour after the heater was activated. The vapor in the membrane module must first be heated by the hot vapor in the evaporator by conduction through the initially cold stagnant vapor, essentially a vapor plug, inside the connecting tube in between the two. The long lead time required for priming seems to be the result of the very slow heat conduction through the stagnant vapor because of its small thermal conductivity.[7] As the vapor plug gets heated, free convection gradually sets in and the heat transfer process quickens in tempo because of direct mixing. Once primed, the temperature at the module vapor inlet (T_3) rises rather rapidly as a result of convective flow and correspondingly, the rate of water transport becomes nearly constant at about 0.02 cc/s. The evaporator power was turned off at the time indicated by point B on the graph. It was noted that pumping stopped within minutes thereafter and T_3 fell off at a much faster rate than T_1 and T_2, the latter being the evaporator temperatures at the inlet and outlet, respectively. There should be little or no vapor flow at all from the evaporator after its power has been turned off. It seems that the rapid fall of T_3 is due to the cooling effect rendered by the circulating aqueous solution (T_4, T_5).

It is of interest to note that, at constant evaporator heat input, the system temperatures never reached steady-state conditions, even after extended operation. An analysis of the evaporator temperature (T_2) showed that the massive evaporator wall and the large extent (judged from its small thermal diffusivity) of the attached Rubatex® insulation resembled the time response of the surface of a semi-infinite solid with

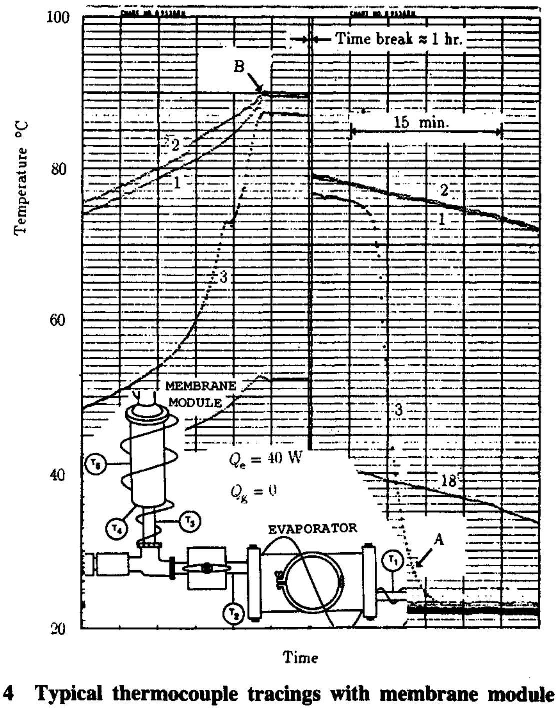

Fig. 4 Typical thermocouple tracings with membrane module uninsulated.

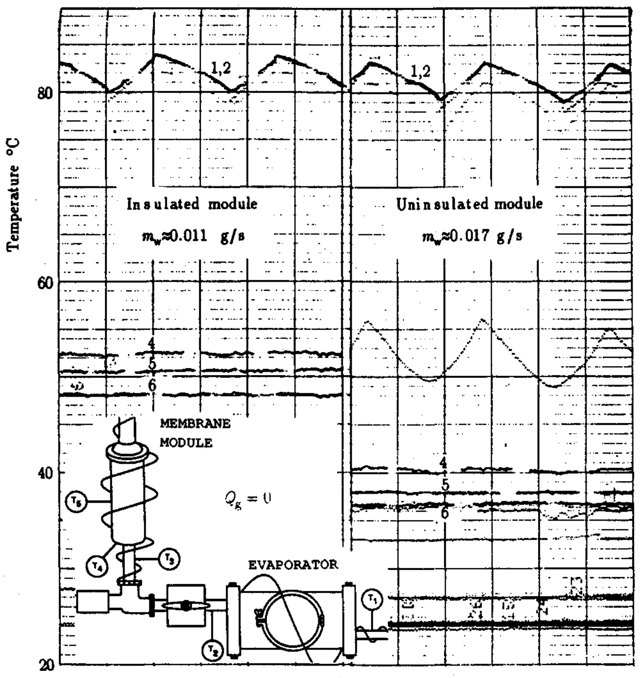

Fig. 5 **Thermocouple tracings obtained by keeping the evaporator at a constant mean temperature.**

the boundary condition of a constant surface heat flux.[8] Two separate runs, corresponding to cases of an insulated and an uninsulated membrane module, respectively, were performed under a condition wherein the evaporator temperature was controlled to within a range of ±2°C, about a median of roughly 82°C, and, again, a generator heat input zero. A typical comparison is shown in Fig. 5. Using the noninsulated case as a standard for comparison, the insulated test yielded a membrane temperature (T_4, T_5) in excess of more than 10°C higher and, correspondingly, a reduction of more than 30% in the water rate-of-transport has been observed. This seems to indicate that the membrane pumping rate is closely related to either the temperature or the enthalpy difference between the vapor bulk and the saturated vapor at the membrane temperature.

Approximate Qualification Analysis

The mechanism described in the preceding section for membrane transport is deduced from the observations made in sorption studies of water vapor; that is, water sorbed in the membrane is liquid-like in character. To see if such a mechanism is well within the limitations set forth by the laws of thermodynamics, thus lending support for its justification, we make the following calculations.

The flow velocities of the circulating aqueous solution can be estimated by applying the steady-state force balance equations to the two streams inside the coaxial standing double-pipe and double-shelled membrane module sketched in Fig. 6. The stream is in the annular space, where the strong (heavy) solution flows downward and the other is in the center pipe where the weak (less heavy) solution flows upward. For the sake of simplicity, the upward and downward flows inside the membrane module are treated in the same manner as their counterparts inside the double pipe with equivalent lengths l_1 and l_2, respectively. The flows are considered to have the same static pressures at the top as well as at the bottom of the vertical structure. The steady-state force balance equations are

$$\frac{m_1V_1 - m_2V_{b1}}{A_1} = p_b - p_t - \rho_1 gl - f_1\frac{\rho_1V_1^2}{2}\frac{l_c + l_1}{D_1} \tag{1}$$

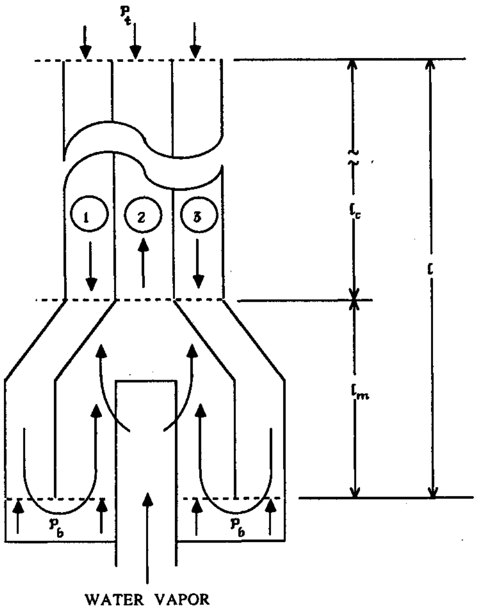

Fig. 6 **Flow configurations of aqueous solution in the vertical structure of the double pipe and the membrane module.**

for the upflow, and

$$0 = p_t - p_b + \rho_2 gl - f_2\frac{\rho_2V_2^2}{2}\frac{l_c + l_2}{D_2} \tag{2}$$

for the downflow. Here A_1 and A_2, each assumed to be constant for simplicity, are the cross-sectional areas of the upward and downward flows, respectively, ρ the density, f the friction factor, V the mean flow velocity, V_{b1} the inlet velocity of the upward pipe flow, and D the hydraulic diameter; $g = 980$ cm/s^2 is the gravitational acceleration, l is the total height of the structure (including the membrane module), l_c the height of the straight double-pipe column, p_t and p_b are the static pressures of the aqueous solutions at the very top and the very bottom of the total flow structure, respectively, and

$$m = \rho AV \tag{3}$$

is the mass flow rate of the solution.

The flow rates are related by mass conservation so that

$$m_1 = m_2 + m_w \tag{4}$$

$$\rho_1A_1V_1 = \rho_2A_2V_2 + m_w \tag{5}$$

where m_w is the mass rate of water transport across the membrane into the pipe stream, which is flowing upward. In Eq. (1) for the force balance, the axial momentum flux associated with the inflow of m_w is equated to zero.

The upward flow at the inlet has the same temperature and pressure as the downward flow at the discharge; it then follows that

$$V_{b1} = V_2A_2/A_1 \tag{6}$$

The flows are laminar and, thus, the friction factors are given by

$$f = \frac{64}{Re} = \frac{64\mu}{\rho VD} \tag{7}$$

where $Re = \rho VD/\mu$ is the Reynolds number with μ being the viscosity of the solution. Combining Eqs. (1) and (2) by making use of Eq. (3) for the mass flow rates, Eq. (6) for the inlet velocity V_{b1}, and Eq. (7) for the friction factors, we obtain

$$\left(1 - \frac{\rho_1}{\rho_2}\right) gl = \frac{\rho_1}{\rho_2} V_1^2 - V_2^2 \left(\frac{A_2}{A_1}\right)^2 + \frac{32\mu}{\rho_2 D_2^2}\left[V_1(l_c + l_1)\frac{D_2^2}{D_1^2} + l_c + l_2\right] \tag{8}$$

where $\mu_1 = \mu_2 = \mu$ has been assumed. Variations in the solution density due to changes in temperature are not considered for the sake of simplicity. For illustration purposes, this work accounts for only the density change of the solution due to changes in solute concentration by resorting to the assumption that the volume of the aqueous solution and that of pure water are additive upon mixing. Thus, we are permitted to write

$$A_1V_1 = A_2V_2 + m_w/\rho_w \tag{9}$$

where ρ_w is the density of pure wter. Combining Eqs. (5) and (9), one has the following expressions for the flow velocities in terms of the unknown density ratio ρ_1/ρ_2

$$V_1 = \frac{m_w}{\rho_w A_1}\frac{1 - \rho_w/\rho_2}{1 - \rho_1/\rho_2} \tag{10}$$

and

$$V_2 = \frac{m_w}{\rho_2 A_2}\left[\frac{\rho_1}{\rho_2}\frac{\rho_2}{\rho_w}\frac{1 - \rho_w/\rho_2}{1 - \rho_1/\rho_2} - 1\right] \tag{11}$$

Elimination of the flow velocities V_1 and V_2 in Eq. (8) by making use of Eqs. (10) and (11) gives, after a brief calculation

$$\frac{\rho_1^2}{\rho_2^2} - 2(1 + \varepsilon_1)\frac{\rho_1}{\rho_2} + 1 + \varepsilon_2 = 0 \tag{12}$$

where

$$\varepsilon_1 = \frac{1}{2gl\rho_w^2A_1^2} m_w^2 + \frac{16\mu}{g\rho_w\rho_2A_2D_2^2}\frac{l_c + l_2}{l} m_w, \tag{13}$$

and

$$\varepsilon_2 = \frac{1}{gl\rho_2^2A_1^2} m_w^2 + \frac{32\mu}{g\rho_2^2A_2D_2^2} \cdot \left[\frac{l_c + l_2}{l} - \frac{D_2^2A_2}{D_1^2A_1}\left(\frac{\rho_2}{\rho_w} - 1\right)\frac{l_c + l_1}{l}\right] m_w \tag{14}$$

With the use of the experimentally determined mass rate of water transport across the membrane m_w, Eqs. (12–14) serve to determine the solution density of the upward flow in the center pipe. The flow velocity V_1 is then calculated from Eq. (8) and the mass flow rates of the streams are given by

$$m_1 = \rho_1A_1V_1 \tag{15}$$

and

$$m_2 = m_1 - m_w \tag{16}$$

From the viewpoint of heat transfer, the membrane module works somewhat like a condenser. Water vapor coming from the evaporator is cooled and condensate is formed on the membrane while the heat released is transferred into the aqueous sugar solution. Ideally, if heat transfer occurs at an infinite rate, then the solution and the condensate will reach the same temperature before mixing by osmotic flow takes place. Under this ideal condition, an energy balance then gives

$$T_a = \frac{4.18m_2T_4 + m_w(1.87T_3 + h_{fg})}{4.18m_2 + 1.87m_w} \tag{17}$$

where T_a is the condensing temperature, h_{fg} is the enthalpy of condensation for water vapor, and T_3 and T_4 are the membrane module inlet temperatures of the water vapor and the aqueous sugar solution, respectively. The specific heat of the aqueous solution, because of lack of data, has been arbitrarily assumed to be the same as that of water.

Table 1 Calculated results of Run 1[a]

m_w g/s	m_2 g/s	m_2/m_w	T_a°C	p_a kPa	Re_1
0.020	3.03	152	57.52	17.74	178
0.055	5.01	91	60.06	19.97	295
0.091	6.42	70	61.90	21.73	379

The hydraulic diameters are $D_1 = 1.26$ cm, $D_2 = 1.40$ cm and the flow areas are $A_1 = 1.24$ cm², $A_2 = 5.04$ cm². These dimensions are obtained from the line drawings appeared in an interim progress report.[9] The total height $l \approx 1.5$ m, the standing column has a length of $l_c \approx 1.25$ m, and the vertical height of the membrane module $l_m \approx 0.25$ m. For order of magnitude estimate, the equivalent lengths $l_1 = l_2 = l_m$ are to be assumed.

For illustration, we use the temperature data of Run 1 from the report of Drolen et al.[2] recorded at 3:00 p.m. on May 18: $T_3 = 81.8$°C, $T_4 = 53.7$°C, and a static pressure at the membrane on the solution side $p_b \approx 16$ psia + 59″ solution column ≈ 18 psia = 124 kPa. The strong aqueous sugar solution flowing downward in the annular space has a nominal molality of unity. Its properties evaluated at 50°C ($\approx T_4$) are:[10] $\rho_2/\rho_w = 1.13$, $\mu_2/\mu_w = 3.19$ with $\rho_w = 0.988$ g/cm³, $\mu_w = 0.547$ centipoise and $h_{fg} \approx 2380$ J/g; giving $\rho_2 = 1.11644$ g/cm³ and $\mu = \mu_2 = 1.74$ cp.

The calculated results, employing Eqs. (12–17), are shown in Table 1, where p_a is the condensing pressure at the temperature T_a, for three different values of the mass rate of water transport m_w.

In the above table $Re_1 = \rho_1V_1D_1/\mu$ is the Reynolds number of the upward pipe flow. The table shows that the condensing conditions of the water vapor are quite similar because of the large m_2/m_w ratios and that the flows are laminar, as assumed, because of the small Reynolds numbers ($Re_2 < Re_1$). The 1 M aqueous sugar solution has a mole fraction of water $x_w = 0.9823$. Using an average of $T_a = 333$ K with $p_a = 20$ kPa and a liquid water volume of $v_f = 0.01832$ m³/kgmole we find the system to have an availability

$$\Phi = -RT_a \ln x_w = 49.3 \text{ kJ/kgmole of water}$$

The work required for pumping liquid water is $v_f(p_b - p_a) = 1.9$ kJ/kgmole, which is more than an order of magnitude smaller than the availability Φ and therefore the process is thermodynamically possible. On the other hand, if the water were to transport across the membrane by vapor compression, then the minimum work needed would be by compression along the saturation line. For this case, use $T_a = 333$ K as before with $h_a = 2610$ kJ/kg and $s_a = 7.9096$ kJ/kg·K, and for saturated vapor at $p_b = 124$ kPa, one has $T_b = 379$ K, $h_b = 2679$ kJ/kg, and $s_b = 7.2706$ kJ/kg; and the minimum work for vapor compression is equal to

$$h_b - h_a - \int_a^b T\,\mathrm{d}s \approx h_b - h_a - 0.5(T_a + T_b)(s_b - s_a)$$

$$\doteq 296 \text{ kJ/kg} \doteq 5340 \text{ kJ/kgmole}$$

where the path of integration is along the saturated vapor line. Because, at equilibrium, the free energy of the vapor is identical to that of the liquid, the system's availability remains the same as before. The above figure shows that the work required for vapor compression is more than 100 times higher than what is thermodynamically available. It must, therefore, be concluded that for this case osmotic transport of water by vapor compression is not possible.

Conclusions

The use of the proposed membrane module in an absorption refrigeration system, where water is employed as the refrigerant, to take the place of the refrigerant absorber and the liquid circulation pump does provide water flow from the evaporator to the generator against a pressure rise. The thermal transfer cycle obtained from such a modification, however, no longer functions as does that of a heat pump. This is because water vapor, initially in the free state, must first physically become attached to the membrane before transport can take place. Attachment of water vapor to the membrane material is by physical adsorption, which is always accompanied by the release of heat from the vapor. At high moisture uptake by the membrane material, the sorption process is found to resemble that of condensation. Thus, unlike in a heat pump cycle where one might say that the heat from the evaporator is first lifted to and then released at a higher temperature, here nearly the same heat must be released from the refrigerant at a temperature equal to or lower than that of the evaporator before sorption can take place. It must be concluded that the system cannot be made to perform as a heat pump.

Acknowledgment

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