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Experimental and Modeling Improvements to a Co-Fluid Cycle Utilizing Ionic Liquids and Carbon Dioxide

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ABSTRACT

Carbon dioxide is undergoing a renaissance as an alternative to synthetic refrigerants due to its environmental advantages in addition to a high density and excellent transport properties. A weakness of carbon dioxide is having a critical point which occurs at a lower temperature and higher pressure than most other fluids used as refrigerants. This combination leads to high operating pressures, especially on the heat rejection side of the thermodynamic cycle.

Ionic liquids (IL), which are salts which remain in their liquid phase at room temperatures, have been shown to strongly absorb carbon dioxide. Due to recent advances in ionic liquids, the cation and anion groups are able to be formulated to tailor a variety of fluid properties including liquid-vapor equilibrium characteristics. By selecting appropriate ionic liquids, it is possible to reduce the operating pressure of an air-conditioning system utilizing carbon dioxide to be in the range of conventional refrigerants. Not only are ionic liquids able to physically absorb volatile refrigerants as in other co-fluid cycles, but ionic liquids also offer the possibility of chemical absorption thereby giving the opportunity for greater enthalpy changes.

Conceptually, the ionic co-fluid cycle is similar to a traditional vapor compression cycle. In the high pressure heat exchanger, heat is rejected to lower the enthalpy and to absorb carbon dioxide into the ionic liquid. The enthalpy is further reduced in an internal heat exchanger before the high pressure liquid is passed through a valve to decrease the pressure which causes the fluid mixture to cool. Heat is absorbed by the mixture from the environment, thus boiling additional carbon dioxide. After passing through an internal heat exchanger, the fluid is mechanically compressed and the cycle is repeated.

System modeling work was utilized to identify important thermodynamic characteristics for achieving good performance. These characteristics included heats of mixing, solubility, entropy of mixing, and viscosity. Using experimentally and numerically determined IL-CO₂ mixture properties, system models were able to predicatively select anion and cation pairs for optimizing performance.

The ionic liquids selected from the modeling exercises were subsequently synthesized for demonstration in a laboratory. An air conditioning system was built from components designed for use with conventional refrigerants. The system was installed in a facility which was instrumented to measure air and refrigerant pressures and temperatures. Air flow rate and temperature information allowed the cooling capacity to be measured. The power consumption of the pump and compressor used to circulate the working fluids was measured so that COP could be determined.

Modeling results were validated with experimental findings. The emphasis of modeling and experiments was to determine the effect of operational parameters on system performance. The loading of ionic liquid and carbon dioxide, along with valve opening and compressor speed, was found to dramatically alter the operating pressures. The difference and ratio between high and low side pressures directly affected the specific cooling capacity and COP, respectively. While the model had strong agreement with the experimental results, non-idealities to be incorporated in more sophisticated models are identified.

1. INTRODUCTION

Co-fluid cycles are similar to traditional vapor compression cycles, but have some aspects of absorption cycles. Such a cycle was presented by Spauschus, *et al.* (1999) to show a possibility for lowering the operating pressure of carbon dioxide to that of a traditional automotive AC system. To quickly familiarize the concept, a co-fluid cycle involves two fluid components: a volatile refrigerant and a low volatility liquid co-fluid from which the volatile refrigerant is absorbed or released. Because the volatile component goes in and out of solution, the latent heat, heat of mixing, and in some cases reversible chemical reactions, give the potential for greater specific enthalpy changes. Additionally, the co-fluid decreases the operating pressure of the volatile fluid, which may be beneficial for high pressure fluids. A schematic showing the main components of the co-fluid cycle are shown in the Figure 1. The desorber is the heat exchanger which is used to absorb heat or remove humidity from its surroundings; it is analogous to an evaporator in a traditional vapor compression system. In the desorber, volatile refrigerant is vaporized from the co-fluid. An internal heat exchanger further utilizes the fluid to decrease the enthalpy of the fluid which will enter the expansion valve. After the internal heat exchanger, the refrigerant vapor and the co-fluid are compressed to a high temperature, high pressure condition. This process may be accomplished by a compressor alone, or in the case of these experiments, by a compressor in parallel with a pump. The impact of these compression options will be discussed in detail later. After the compression process, heat is rejected from the high pressure fluid mixture which causes the mixture to cool. Cooling the high pressure fluid causes the volatile refrigerant to be absorbed by the co-fluid; this process takes places in the resorber. This process is furthered by use of the previously described internal heat exchanger. After passing through the high pressure side of the internal heat exchanger, the pressure of the mixture is decreased by a valve. The valve causes the volatile mixture to come out of solution thus decreasing the temperature of the fluid mixture. The fluid then returns to the desorber, thus completing the cycle.

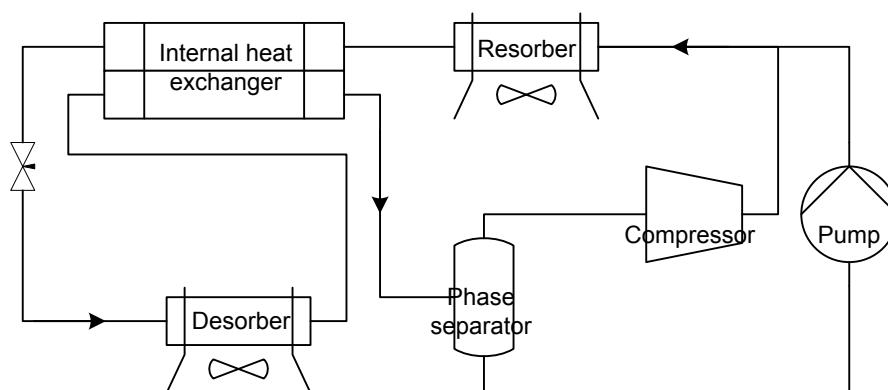


Figure 1. Schematic of co-fluid cycle components

The co-fluid pair utilized in this study was an ionic liquid with carbon dioxide. The advantages of carbon dioxide at this point are well known: it is a naturally occurring fluid with minimal environmental impact in the quantities used for HVAC&R, non-flammable, very low toxicity, inexpensive, and with good transport properties. The major downsides of carbon dioxide are due to its low critical temperature and high critical pressure. The co-fluid cycle continues to take advantage of carbon dioxide's strengths, while avoiding its primary weaknesses. Ionic liquids are a class of salts which are liquids at room temperatures this differentiates them in a key way from one of the main problems of lithium bromide, the salt commonly used in absorption refrigeration. By selecting ionic fluids with certain cation and anion pairs, it is possible to tailor the properties of the ionic liquid to control for key parameters such as heat of mixing and viscosity. From measured mixture properties, it is possible to construct a pressure-enthalpy property diagram for a co-fluid pair, as demonstrated by Greenfield *et al.* (1999). Figure 2 shows the pressure-enthalpy diagram for the ionic liquid hmim Tf2N and CO₂, with a maximum mass concentration of 10% of carbon dioxide (i.e. 1 kg of CO₂ to 9 kg of IL). For this concentration, the uppermost, dashed red, iso-concentration line corresponds to the conditions at which all carbon dioxide is absorbed by the ionic liquid. Above this line the concentration no longer changes, since there is no more vapor carbon dioxide which may be absorbed. The red lines below the fully absorbed line correspond to lower concentrations of carbon dioxide in the liquid phase. To put in terms of refrigerants used for traditional vapor compression, this is effectually a zoomed in view near the liquid saturation line for a strongly zeotropic mixture. An ideal cycle is shown in the sketch for illustrative purposes; in this illustration the heat is rejected to a 35°C ambient from a 24°C indoor environment.

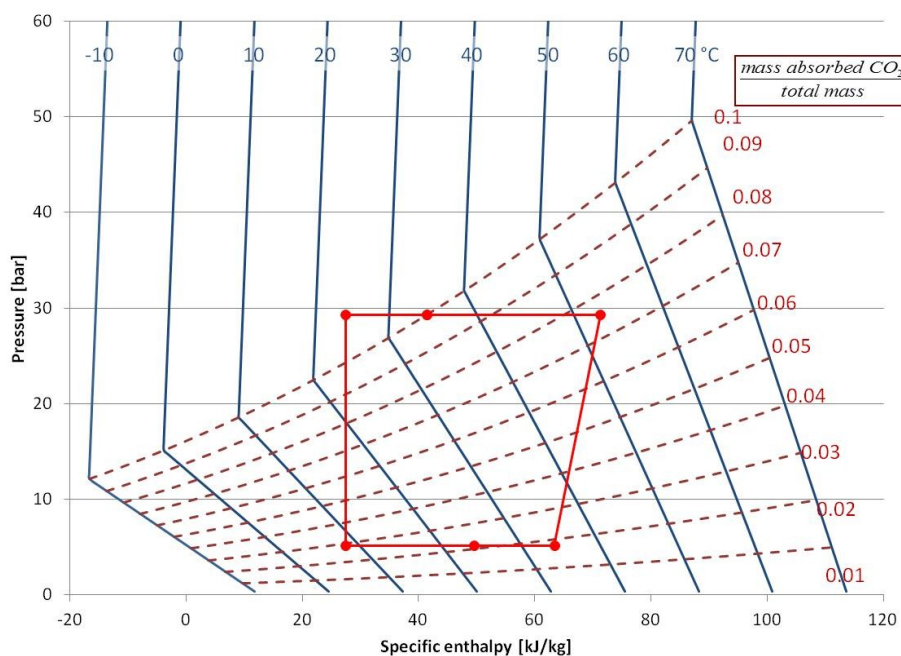


Figure 2. Pressure-enthalpy diagram for hmim Tf₂N and CO₂

The fluid property diagram can be utilized to sketch a cycle with comparable COPs to conventional air conditioning system. Some thought about the operating region for the entire cycle will quickly bring to mind other practical considerations. The first is that, the mechanism used to compress the fluid must handle much higher mass flows of liquid than is typical for vapor compression. Secondly, the shape of the isotherms means that heat exchanger design must be carefully considered. Thirdly, there will be a strong impact on system performance caused by the selection of concentrations and operating pressures.

2. MODELING FOR IONIC FLUID SELECTION

Performance is characterized by coefficient of performance (COP, the ratio of net cooling power to net rate of input work) and cooling capacity (ratio of net cooling power to net rate of circulation of working fluid). These quantities vary with properties of the working fluid, including enthalpy and entropy of CO₂ absorption, molecular weight,

density, and heat capacity. Performance also varies with discharge pressure, suction pressure, and ratio of total CO₂ to total IL in the apparatus.

Performance for the hardware pictured in Figure 1 was estimated using a “cycle model” in which equations represented the essential physical processes associated with each hardware component. The cycle model is similar to that described in Mozurkewich et al. (2002). Each of the three heat exchangers was modeled using two equations, one expressing conservation of energy and the other expressing heat-transfer rate as parameterized by effectiveness. The expansion valve was modeled as a constant enthalpy device, and compression was assumed to be isentropic. Two different compression schemes were considered. For the case of “wet compression,” in which liquid and vapor are compressed in a single device, the mixture was assumed to maintain equilibrium throughout the compression process. For the case of separate compression of the two phases, as shown in the figure, each phase was individually treated isentropically, and then the two phases were recombined isenthalpically at fixed pump-and-compressor outlet pressure. This second approach pays a theoretical penalty due to the non-equilibrium mixing process, but it has the great practical advantage of allowing use of standard pumps and compressors.

This modeling approach involves unrealistic assumptions. The most serious assumption is that equilibrium is attained at the exit of each hardware component. The requirement for CO₂ to absorb into or desorb out of the ionic liquid makes the attainment of equilibrium unlikely. A second assumption is neglect of viscosity, which causes underestimation of required work input. Thus this cycle model is unsuitable for a full engineering analysis. Its primary use was in identifying suitable thermodynamic characteristics for the working fluid.

Initial experiments used [hmim][TF₂N] as the IL because it is readily available from commercial vendors and its properties are well documented in the literature. Absorption of CO₂ into this IL arises solely from physical processes and is fairly weak, with an enthalpy change roughly -12 kJ/mol. While this value is comparable to the latent heat of pure CO₂, the IL has much larger molecular weight and heat capacity than CO₂. Consequently, the temperature change due to a given enthalpy change is much smaller, and much more mass must be pumped around the circuit to attain equal cooling capacity.

Chemical absorption of CO₂ allows larger enthalpy changes with corresponding improvements in predicted performance. Explorations using the cycle model indicated that a doubling of enthalpy change compared to [hmim][TF₂N] would substantially improve performance. As enthalpy change becomes more exothermic, operating pressures also decrease, vapor density increases, and practical problems arise with plumbing diameter, compressor displacement, etc. The model indicated that a tripling of enthalpy change with respect to [hmim][TF₂N] would probably be excessive. Using this guidance from the cycle model, new ILs were synthesized with refrigeration applications in mind and were screened for thermodynamic properties in the target range and for relatively low viscosity. On this basis a chemically absorbing ionic liquid was selected as the second-generation IL for these experiments.

3. EXPERIMENTAL FACILITIES FOR AIR CONDITIONING SYSTEMS TESTING

To validate the models, a facility was constructed to measure the capacity and efficiency of the co-fluid cycle. A simplified schematic, which depicts the main components and fluid measurement locations, is shown in Figure 3. The desorber utilized in this study was a relatively conventional round-tube plate-fin design having cross-counterflow circuitry. Coil circuiting considerations are important due to the temperature glide. The resorber was a single slab microchannel design designed as an automotive condenser. The internal heat exchanger was a counter flow microchannel sandwich.

Cooling capacity of the desorber was determined by an air-side energy balance. This energy balance is calculated using the difference in temperatures before and after the desorber and the air mass flow rate. Air mass flow is determined from pressure measurements using a calibrated air flow rate nozzle as prescribed by ASHRAE Standard 41.2. Air temperature is measured with a grid of T-type thermocouples before and after the desorber, as well as by thermocouples located in the discharge of the air flow rate nozzle. Air flow through the nozzle is controlled by varying blower speed, while air inlet temperature is controlled with an electric heater. For reference, similar measurements and controls are utilized for the resorber duct, with the exception that a chilled water coil is utilized for absorbing heat that is rejected from the cycle.

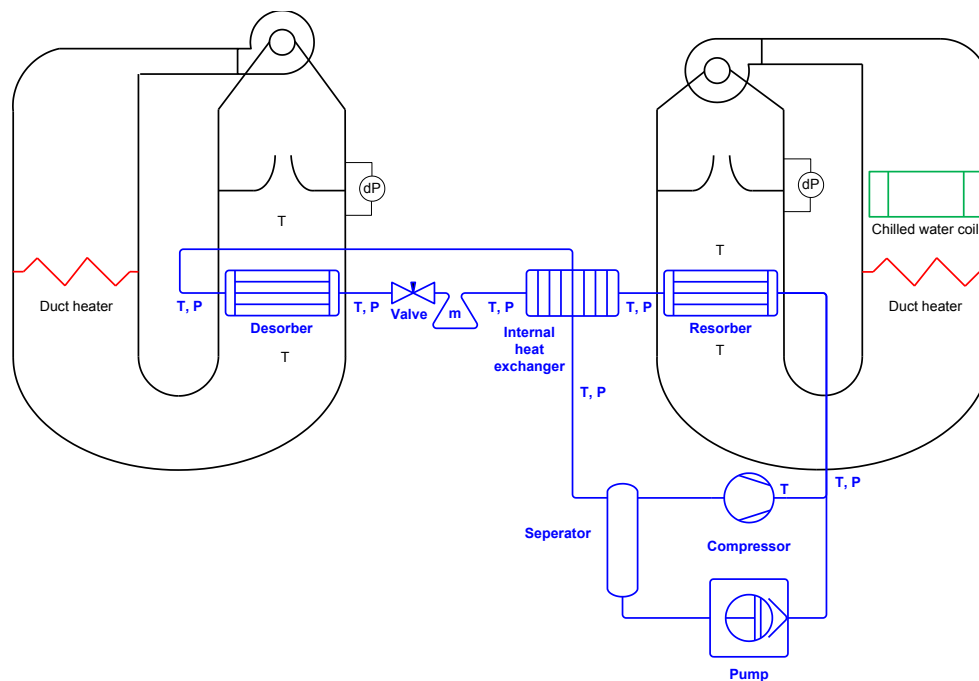


Figure 3. Schematic of experimental facility for systems testing

For reasons related to prototype reliability primarily concerning the possibility for motor torque from liquid slugging, pulley-driven open shaft automotive compressor and open shaft pump were utilized. The automotive compressor was a scroll design which was found to be relatively robust to liquid slugging. The pump was a reciprocating design. For determining system efficiency, the pump and compressor power measured in two different ways. The first method for determining power consumption is by measuring electrical power consumption. However, due to the relatively large size of compressors and pumps available, the motors used to drive the pump and compressor were run at very low speeds. Low speed operation caused low efficiencies in the variable frequency drives used to reduce speed and in the motors themselves. To have a more direct measurement of power consumption, the speed and torque of the drive shafts were measured. The large deviation between electrical and shaft power (roughly 50%), could have been eliminated by having more appropriately sized equipment.

Both the relative concentration of carbon dioxide to ionic liquid, and the total charge of ionic liquid were expected to be important parameters effecting performance. Gravimetrically determined quantities of ionic liquid were drawn into the system via a vacuum. After ionic liquid was charged, carbon dioxide was introduced. Carbon dioxide was charged until the desired equilibrium saturation pressure was reached. Periodically during the carbon dioxide charging process, the system was agitated by the pump to hasten equilibrium.

Flow sights were installed at several key locations to determine whether liquid, vapor, or two phase flow were present. The flow sights were installed before the pump, the compressor, and the expansion valve. By use of these sightglasses, it was possible to confirm proper operation of the separator. While complete absorption of carbon dioxide prior to the expansion valve is desirable, generally some vapor was present in this sightglass. The presence of vapor, caused the coriolis type flow meter which was installed to often give false readings, thus while a mass flow meter was installed it rarely give reliable readings. Therefore all capacity measurements were taken from air side energy balances.

4. TRENDS IN EXPERIMENTAL DATA

The effect of several variables was systematically explored in the experimental facility to determine their effect on system efficiency. The primary values studied were ionic liquid charge, carbon dioxide charge, expansion valve opening, and speeds of the compressor and pump. To make the experimental parameter space more manageable, not

every combination of parameters was measured. For this reason normalized capacity and efficiency are shown to focus on the trends. The data point used for the purpose of normalization had a capacity of 566 W and a COP of 37%. Such a low COP value is due to not having proper equipment for performing wet compression. Tests were conducted at resorber and desorber air inlet temperatures of 25°C and sufficiently low dew point temperatures so as to not have condensation on the desorber coil.

The first parameter studied was ionic liquid charge. Due to the expense involved in producing prototype chemicals, ionic liquid charge was kept as small as possible. Because of the generally small charge of ionic liquid in the system, the effect of changing ionic liquid mass may have been magnified. Because liquid holdup in heat exchangers and connecting tubing is primarily determined by flow rates and heat flux, the main place where ionic liquid is able to accumulate is in the phase separator. The buildup of ionic liquid from the pump suction, through the connecting tubing, and into the phase separator had two major effects. The first was that the net positive suction head on the pump was increased. This decreased the cavitations due to out gassing from the ionic liquid. The second was that the flow rate of ionic liquid through the system was far steadier. Therefore, increased ionic liquid charges allowed for faster pump speeds. The steadiness was increased due to additional liquid because there were always sufficient quantities of ionic liquid to feed the pump. When the ionic liquid charge is too small, the pump is able to drain the fluid separator, and then must wait for vapor to drag the liquid back to the separator. Once enough liquid built up to establish head, the liquid was promptly drained by the pump and the cycle repeated. Because the laboratory was designed for steady state measurement, the effect of ionic liquid charge was not directly quantified. Too much ionic liquid was also found to be detrimental because it lead to unnecessarily high pressure drops in the heat exchangers and eventually did not leave sufficient room for carbon dioxide expansion.

Carbon dioxide charge, or more precisely the relative charge of carbon dioxide to ionic liquid, determines the standstill pressure inside the system. As the compressor and pump are run, the cycle grows from the equilibrium point. Higher carbon dioxide charges therefore, have the tendency to increase both the high and low side pressures. Beneficially, higher high-side pressures allow more carbon dioxide to be absorbed by the ionic liquid in the resorber. However, higher carbon dioxide charges also have the tendency to increase pressure on the low side of the system. Studying the pressure-enthalpy diagram, it can be seen that pressures must be kept low in order to achieve the temperatures cold enough to give cooling potential. From Figure 4, it can be seen that standstill carbon dioxide concentrations corresponding to 20 to 25 bars of standstill pressure were optimal for the [hmim][TF2N] for a wide range of compressor speeds. Different ionic fluids, different volume distribution between the high and low pressure sides of the system, and different component efficiencies would yield different optimal carbon dioxide charges.

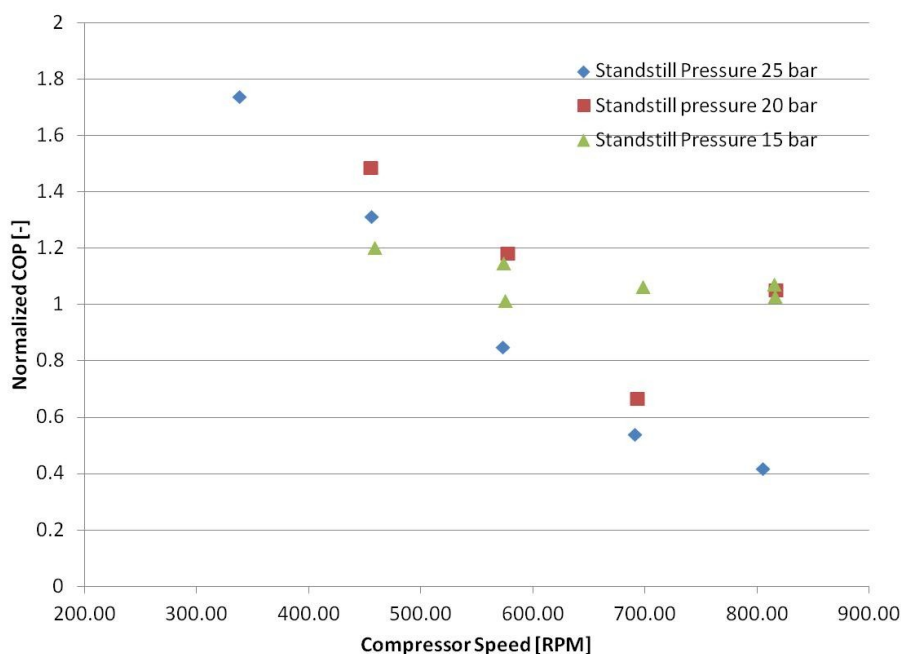


Figure 4. Effect of standstill pressure on system efficiency

The size of the opening in the expansion valve determines the ease at which fluid can move from the high to the low pressure side of the system. As the valve is opened, some combination of smaller pressure drop and larger flow rates is experienced. While the expansion valve is nearly isenthalpic, expansion valve opening affects capacity in two major ways. Firstly, it affects capacity by changing the mass flow of refrigerant. This same effect on capacity is the basis of thermostatic expansion valves in conventional vapor compression systems. The second way in which capacity is affected is by changing the desorber pressure, thus changing the desorber inlet temperature. While expansion valve opening has some effect on saturation pressure in conventional vapor compression systems, the effect is far more pronounced with the ionic liquid co-fluid cycle. The strong effect of pressure on temperature can be seen in the pressure-enthalpy diagram shown in Figure 2. As there is generally a small difference between the desorber inlet temperature and the air inlet temperature, even a slight change in inlet temperature strongly affects the heat transfer rate between the fluid and the air. Taken to the extreme, if there is insufficient expansion, desorber heat transfer can go in the wrong direction. Refrigerant (ionic liquid plus carbon dioxide) flow rate was adjusted by adjusting expansion valve opening while keeping all other parameters constant. The result of this experiment is shown in Figure 5. It can be seen that a more restrictive valve setting, shown by a lower mass flow, resulted in better performance. This is due primarily because the more restrictive valve released more carbon dioxide thus making colder entrance temperatures which utilized the desorber's finite size.

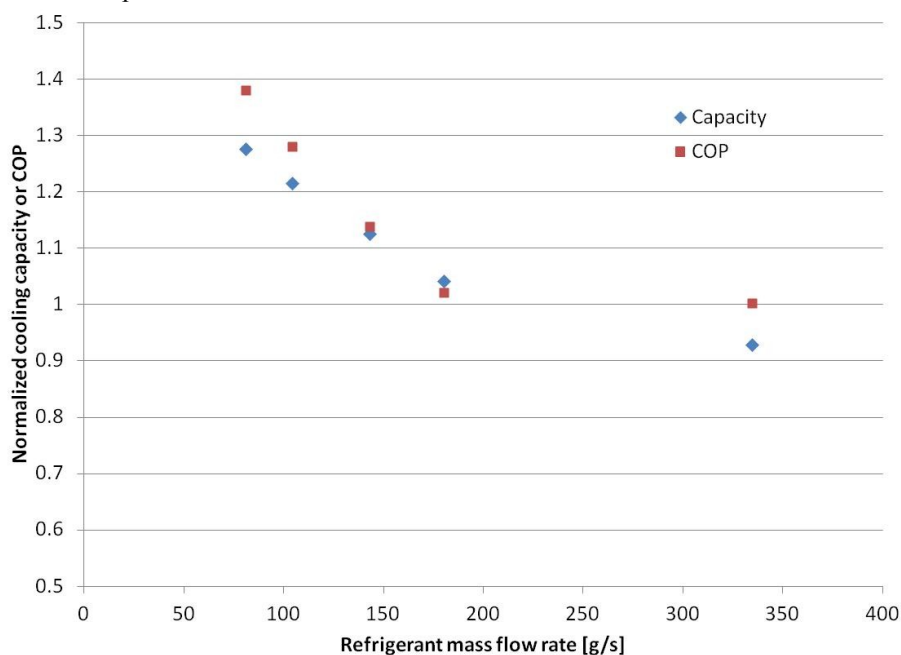


Figure 5. Effect of expansion valve opening on system performance

The effect of compressor and pump speeds was independently explored. While ideally, the co-fluid cycle uses only one machine to move the fluid to the high pressure and temperature state, the experimental facility allowed exploration of the relative flow rates of the two fluids in addition to exploring the relative speed of the pump and compressor relative to the size of the heat exchangers. Generally faster pumps and compressors create greater differences between high and low side pressures, which is similar to closing the expansion valve. In contrast to closing the expansion valve, speeding the compressor and pump simultaneously increases the mass flow rate. The more important part of the study was to explore the relative speed between the pump (liquid flow rate) and compressor (vapor flow rate). The relationship between the pump and compressor speed affects the relative flow rate of ionic liquid to carbon dioxide. A relative increase in carbon dioxide flow rate increases both the high and low side pressures. Higher fraction of carbon dioxide flow also increases the temperature drop for a given pressure drop in the expansion valve. Higher ionic liquid flow rates conversely lower the pressures and decrease temperature drop across the expansion valve. Higher fractions of ionic liquid give greater enthalpy changes in the heat exchangers at the pressure levels being utilized; this is because the ionic liquid causes the carbon dioxide to undergo a phase change instead of remaining as a vapor. The effects of compressor and pump speed can be found in Figures

6 and 7, respectively. In figure 7, the lowest pump speed utilized only the compressor to provide the flow. For both cases, it can be seen that increased speeds resulted in lower efficiencies. This may be symptomatic of heat exchanger being relatively restrictive. An optimal capacity was found for the compressor; this most likely was due to a balance between increasing displacement rates and decreasing gas densities.

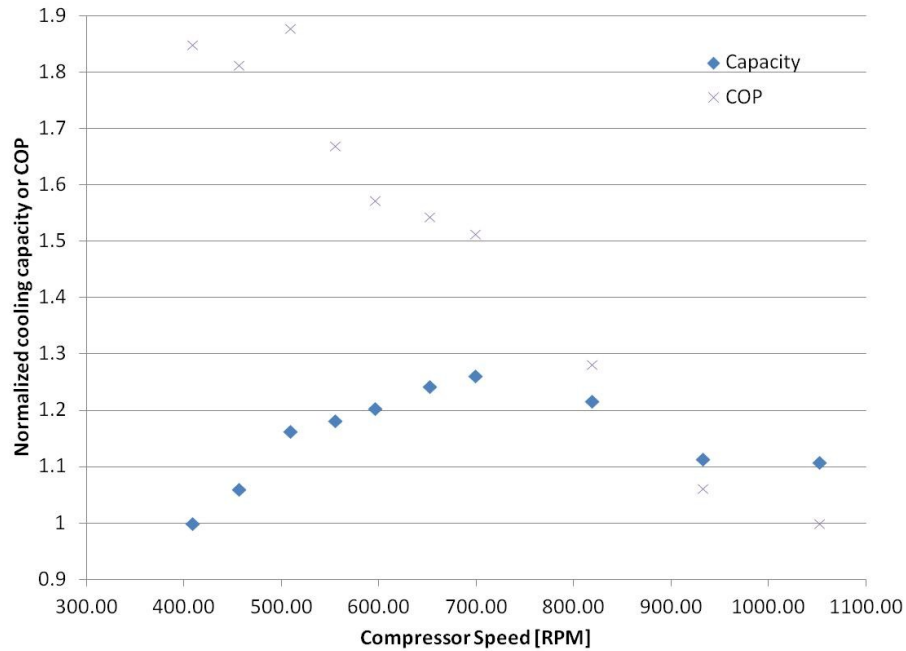


Figure 6. Effect of compressor speed on system performance

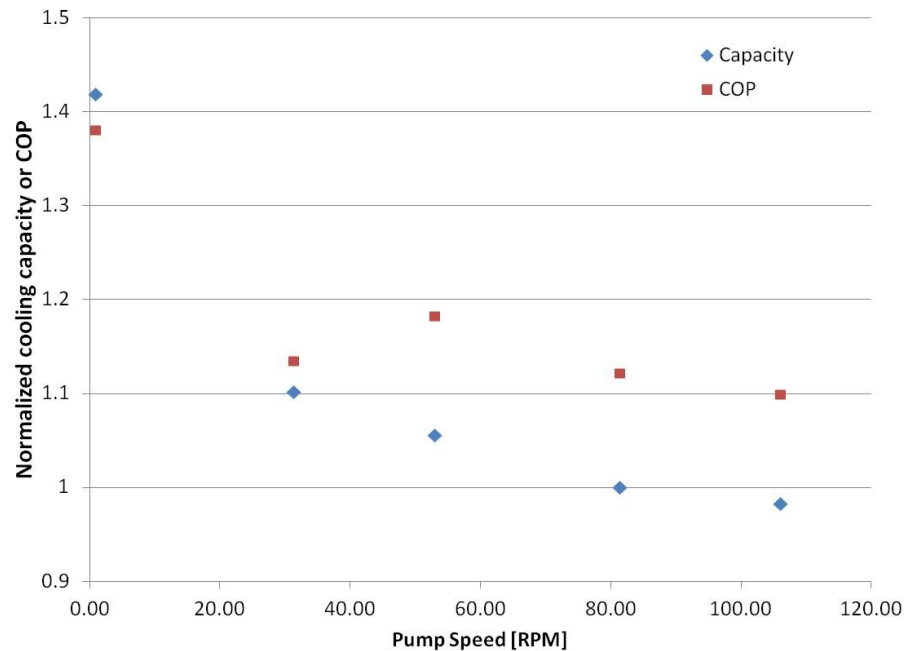


Figure 7. Effect of pump speed on system performance

5. PERFORMANCE NON-IDEALITIES AND AREAS FOR IMPROVEMENT

As mentioned in Section 2, the model incorporated equilibrium thermodynamics and component efficiency or effectiveness assumptions to predict system performance. The modeling results aided in the selection of components used in the laboratory facility. However, assumptions related to non-equilibrium effects and to transport properties such as viscosity were found in the experiments to have a dramatic effect on system performance. Viscosity effects not only pressure drops in pipes, but also refrigerant heat and mass transfer rates. The effect of viscosity will be explored in expansion valve performance and heat exchanger performance.

Due to no work being done and there being minimal heat transfer, the expansion valve is approximately isenthalpic. For conventional systems, where nearly all of the working fluid is volatile, near equilibrium temperatures are reached quickly because mechanical equilibrium between phases happens very quickly. For conventional systems the driving potential is very large, and transport properties do not hinder the evaporation process. In the co-fluid cycle, the majority of the fluid is non-volatile. Therefore, a mass diffusion process must take place for the carbon dioxide to move from within the liquid, to the liquid-vapor interface, before finally being able to be vaporized. Within the liquid phase, the concentration gradient is relatively small, so equilibrium temperatures are not reached initially.

Simplified expansion experiments were performed, in which the mixture was released from high pressure through a needle valve into a flask at room temperature. It was seen that the ionic liquid in the flask continued to outgas for a couple minutes. After a couple minutes of outgassing, the liquid temperature came to the equilibrium temperature. Calculations for equilibrium temperature after the expansion valve were found to differ from the measured temperature drop across the expansion valve in the system by as much as 2K. Because the temperature difference between the desorber air and refrigerant streams was already small, the higher temperatures had a negative impact on performance. To increase the temperature difference before the expansion valve, it would be advantageous to make the flow more turbulent in order to increase the interactions between liquid and vapor and to have a bulk movement of high carbon dioxide regions to the interface.

The relatively high viscosity of the ionic liquid, which are generally between 100 and 1000 cP, and its high concentration relative to the volatile refrigerant, create much higher pressure drops than are experienced in traditional refrigeration systems. As with traditional vapor compression systems, pressure drop on the low pressure side of the system is particularly detrimental from a performance standpoint. The original desorber, an off-the-shelf round tube design selected primarily for its air side heat transfer area and counterflow design, was found to have a pressure drop of as high as 8 bars. For similar capacities in traditional vapor compressions this design had a pressure drop of less than 1 bar. To improve desorber performance, an additional desorber was mounted in parallel (with respect to both the air and refrigerant streams). The parallel desorber decreased pressure drop to approximately 2 bars. The decreased pressure drop, plus some operational improvements, was able to increase system efficiency by more than 55%. It is expected that utilizing heat exchangers optimized for the desorber and the internal heat exchanger could yield further performance improvements.

6. CONCLUSIONS

The performance trends of a co-fluid cycle utilizing ionic liquids and carbon dioxide was explored both computationally and empirically. It was found that the unique properties of ionic liquids could be utilized to create a cycle which utilized phase changes of carbon dioxide at pressures far low than those in a transcritical cycle. While it was found that chemically reacting ionic liquids have advantages of higher enthalpy changes and even lower operating pressures, preliminary experiments were conducted with physically reacting ionic liquids. The effect of several operating conditions was explored experimentally. It was found that sufficient liquid quantities were necessary for the system to run smoothly. An optimal carbon dioxide concentration was found to improve efficiency over a wide range of compressor speeds. More restrictive valve settings gave increased capacity due to allowing a larger temperature drop. Lower pump and compressor speeds lead to better efficiencies. Viscosity, which was not included in the model, was shown empirically to alter the expected temperature at the outlet of the expansion valve and to lead to high pressure drops in the desorber.

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