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ABSTRACT

The main objective of the project was to develop a CO_2 capture process based on sub-ambient temperature operation of a hollow fiber membrane. The program aims to reach the eventual DOE program goal of > 90% CO_2 capture from existing PC fired power plants with < 35% increase in the cost of electricity. The project involves closed-loop testing of commercial fiber bundles under simulated process conditions to test the mechanical integrity and operability of membrane module structural component under sub ambient temperature.

A commercial MEDAL 12" bundle exhibited excellent mechanical integrity for 2 months. However, selectivity was ~25% lower than expected at sub-ambient conditions. This could be attributed to a small feed to permeate leak or bundle non-ideality. To investigate further, and due to compressor flow limitations, the 12" bundle was replaced with a 6" bundle to conduct tests with lower permeate/feed ratios, as originally planned.

The commercial 6" bundle was used for both parametric testing as well as long-term stability testing at sub-ambient conditions. Parametric studies were carried out both near the start and end of the long-term test. The parametric studies characterized membrane performance over a broad range of feed conditions: temperature (-25°C to -45°C), pressure (160 psig to 200 psig), and CO_2 feed concentration (18% to 12%). Performance of the membrane bundle was markedly better at lower temperature (-45ºC), higher pressure (200 psig) and higher CO_2 feed concentration (18%). The long-term test was conducted at these experimentally determined "optimum" feed conditions. Membrane performance was stable over 8 months at sub-ambient temperature operation. The experimentally measured high performance of the membrane bundle at sub-ambient operating conditions provides justification for interest in sub-ambient membrane processing of flue gas.

In a parallel activity, the impact of contaminants (100 ppm SOx and NOx) on membrane performance was tested in the laboratory with membrane minipermeators. NO permeance is intermediate between CO_2 and N_2; while both SO_2 and NO_2 are more permeable than CO_2 at cold condition. This implies that SO_2 and NO_2 will be efficiently removed with CO_2 into the membrane permeate in the proposed cold membrane process.

Calculations were performed by Air Liquide Engineering (ALE) to estimate capture costs based on the proposed sub-ambient temperature membrane process for 90% CO_2 capture from an air-fired coal power plant delivering 550 MW net electricity. Membrane performance in the process simulation was defined by the final parametric test results. This analysis involved refining the process simulation model, obtaining relevant capital cost estimates and using these to estimate a 20-year levelized cost of electricity (LCOE). A sensitivity analysis shows CO_2 capture specific energy requirements of 216-242 kwh/T CO_2 captured. The LCOE estimating methodology followed DOE/NETL study 2010/1397. This analysis indicates increases in LCOE between 48% and 53%. For most equipment, the budgetary capital cost estimates are expected to be valid within ± 20%. The most significant capital costs are due to the (i) feed compression and associated gas pretreatment and (ii) membrane system. For both items, there is a realistic chance for cost reductions in the immediate future (0-5 years) as well as long term reductions. The process continues to hold promise with anticipated cost reductions in compression and membrane operations. In particular, membrane costs could be reduced significantly by

increased production volume (economy of scale) as well as optimization of bundle size and configuration for this application.

PFD definition for a potential field test has been completed through (i) simulation work at DRTC, (ii) discussions with compressor manufacturers and (iii) a field visit to the NCCC, Wilsonville, AL. The PC4 facility at the NCCC is a suitable site for a 0.1 MW scale test.

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1. Executive Summary

The main objective of the project was to develop a CO_2 capture process based on sub-ambient temperature operation of a hollow fiber membrane. The program includes closed-loop testing of commercial fiber bundles under simulated process conditions. The goal was to demonstrate operability and to validate mechanical integrity of the membrane module's structural components under sub-ambient temperature operating conditions. All project objectives and milestones were accomplished in the two year project period.

A bench-scale system was built to measure the ability of commercial modules to operate at the high separation efficiency previously measured using minipermeators in the laboratory. Testing was performed with commercial MEDAL 12" and 6" bundles in a simulated clean CO_2/N_2 flue gas at -20 to -45°C / 1 to 1.5 MPa (10 to 15 bar). CO_2/N_2 (as simulated clean flue gas) was compressed and water-cooled before chilling in the high efficiency finned multi-stream heat exchanger. Cooling in the heat exchanger was provided by the return of retentate gas cooled by Joule-Thomson expansion across a valve, and the return of permeate expanded across the membrane. The system was operated in recycle mode with membrane permeate and expanded residue streams recycled to the compressor suction. This mode of operation decreased the operating cost of the test while conducting a long-term validation of the module operability at cold temperatures.

Testing was first carried out to confirm mechanical integrity of a 12" membrane bundle at cold conditions. No mechanical degradation was observed in the tube-sheet and sealing components over a two months test period with a 12" bundle under cold operating conditions (July through August 2011). The bundle was exposed to pressures as high as 15 bar, with CO_2 concentrations in the 15-30% range and temperatures down to -40ºC routinely (with excursions down to -60ºC). The membrane also experienced several stops / re-starts with complete system de-pressurization. The membrane separation performance was stable over this period.

The 12" membrane bundle exhibited a similar temperature response as observed for laboratory minipermeators. As the operating temperature decreased, CO_2 permeance decreased by approximately 15% compared to the ambient temperature value while N_2 permeance decreased approximately 300%. Though the 12" bundle mechanical integrity was validated, the selectivity at the coldest temperatures was ~ 25% lower than expected. After several diagnostic tests, the possible causes were identified as either a leak in a small section of fiber in this particular bundle or bundle non-ideality. To further study these possible causes, testing was initiated, as planned, with a 6" bundle with lower stage cuts than was possible with the 12" bundle due to compressor limitations.

The 6" bundle was used to develop design data as well as to conduct a long-term stability test. Parametric studies were carried out near the start and end of the 6" bundle long term test. The 6" bundle performance map characterizes membrane performance over a broad range of feed conditions: temperature (-25°C to -45°C), pressure (160 psig to 200 psig), and CO_2 feed concentration (18% to 12%). Results from this study were used to validate the 12" bundle data. The improved performance of the 6" bundle relative to the 12" bundle is attributable to the higher degree of ideality in the 6" bundle.

For a given feed pressure and feed composition, the 6" bundle performance was markedly better when operated at -45°C compared to -40°C. The majority of the long-term test was conducted at the experimentally determined "optimum" feed conditions (-45°C, 200 psi, and 18% CO_2). At the "optimum" feed conditions of the long-term test, membrane performance of the 6" bundle exceeds the performance estimate (based on data with laboratory minipermeators at -40°C), used for the initial LCOE estimation in the project proposal.

The long-term test confirmed stability of membrane performance over approximately 8 months operation and established a conditioned membrane baseline at sub-ambient temperature prior to the final parametric study. The experimentally measured high performance of the membrane bundle at sub-ambient operating conditions provides justification for interest in sub-ambient membrane processing of flue gas

In a parallel activity, laboratory measurements of membrane minipermeators performance exposed to CO_2 /N_2 feed mixtures containing either ~ 100 ppm of SO_2, 100 ppm of NO or 100 ppm of NO_2 were completed. The SO_2 permeance at 15°C was measured to be similar to CO_2, while the value at -40°C is approximately 4 times higher than the CO_2 permeance. This implies that SO_2 in the feed flue gas will be efficiently removed from the flue gas into the membrane permeate in our proposed process where > 90% of the CO_2 is permeated through the membrane and be captured in the CO_2 liquefier. NO permeance is intermediate between CO_2 and N_2; hence, NO concentration is expected to be unchanged by the membrane unit. However, actual NO_x distribution will be more complicated due to NO_2 equilibrium and kinetic effects in the NO-O_2-NO_2 system.

Calculations were performed by Air Liquide Engineering (ALE) to estimate capture costs based on the proposed sub-ambient temperature membrane process for 90% CO_2 capture from an air-fired coal power plant delivering 550 MW net electricity. Membrane performance in the process simulation was defined by the final parametric test results. This analysis involved refining the process simulation model, obtaining relevant capital cost estimates and using these to estimate a 20-year levelized cost of electricity (LCOE). The energy capture estimate was coupled with capital cost estimates to calculate the levelized cost of electricity (LCOE) for 90% CO_2 capture from an air-fired 550 MW net coal power plant.

A sensitivity analysis shows CO_2 capture specific energy requirements of 216-242 kwh/T CO_2 captured. The LCOE estimating methodology followed DOE/NETL study 2010/1397. This analysis indicates increases in LCOE between 48% and 53%. For most equipment, the budgetary capital cost estimates are expected to be valid within ± 20%. The most significant capital costs are due to the (i) feed compression and associated gas pretreatment and (ii) membrane system. For both items, there is a realistic chance for cost reductions in the immediate future (0-5 years) as well as long term reductions. The process continues to hold promise with anticipated cost reductions in compression and membrane operations. In particular, membrane costs could be reduced significantly by increased production volume (economy of scale) as well as optimization of bundle size and configuration for this application.

A process flow diagram (PFD) definition for a potential field test was completed through (i) simulation work at DRTC, (ii) discussions with compressor manufacturers and (iii) a field visit to the NCCC, Wilsonville, AL. The PC4 facility at the NCCC is a suitable site for a 0.1 MW scale test.

2. Introduction

2.1 Process concept

Air Liquide is developing a cost effective hybrid CO_2 capture process based on sub-ambient temperature operation of a hollow fiber membrane in combination with cryogenic distillation. The development program utilizes several key Air Liquide strengths: an existing program for coal oxy-combustion with CO_2 recovery [1,2], cryogenic processing expertise, and membrane manufacturing through MEDAL™, an Air Liquide subsidiary. The cold membrane development work [3] is supported through an U.S. DOE / NETL program aimed at CO_2 recovery by retrofitting existing pulverized coal fired power plants.

For most membrane materials, permeability decreases and selectivity increases with a decrease in operating temperature. However, measurements of commercially available Air Liquide membranes operated at temperatures below -20°C show two to four times increase in CO_2/N_2 selectivity with minimal CO_2 permeance loss compared to ambient temperature values. Operation of these commercial Air Liquide membranes at low temperatures provides an unprecedented combination of CO_2 permeability and selectivity.

Both high membrane module productivity and high selectivity are critical for cost-efficient CO_2 capture [4-8]. High selectivity reduces the energy cost of CO_2 capture while high module productivity reduces the capital cost of the membrane system. The proposed hybrid CO_2 capture process concept couples the unique high performance membrane with cryogenic processing technology to efficiently capture at least 90% of the CO_2 in the flue gas from an air fired power plant. The ultimate target is to achieve this degree of CO_2 capture with increase in the levelized cost of electricity of less than 35%. The process concept is illustrated in the simplified process block flow diagram as shown in Figure 1.

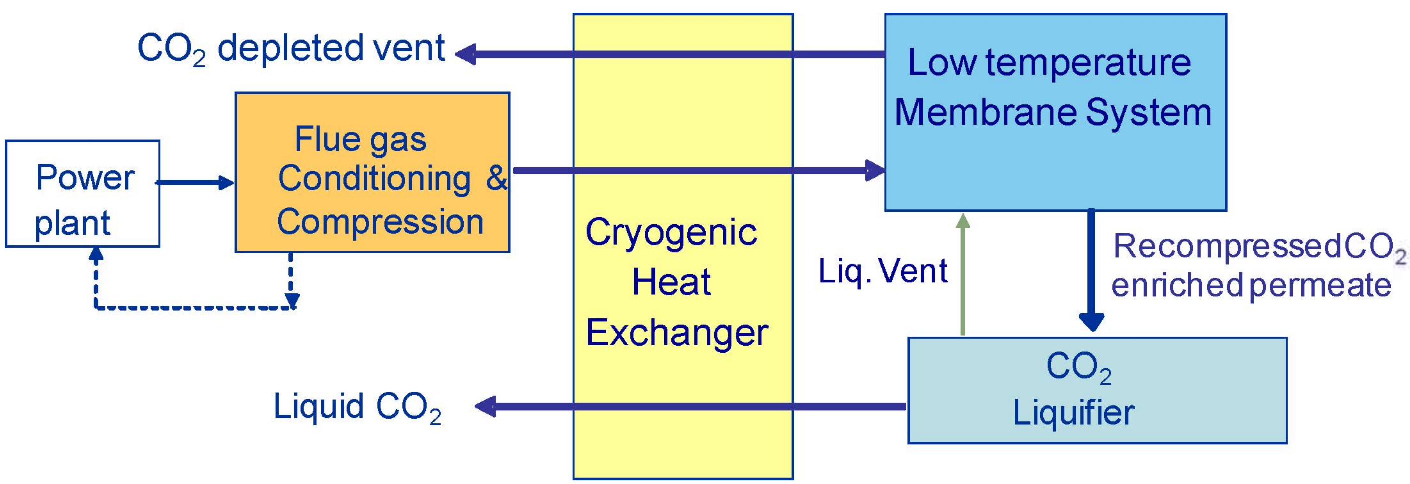


Figure 1. Sub-Ambient Membrane System for CO_2 separation

As in some previous literature [9, 10], the membrane serves as a CO_2 pre-concentrator sending a small CO_2 -rich stream to the cryogenic purification unit (CPU). However, in contrast to previous membrane schemes, the membrane is now operated at cold temperatures. The process is feasible because of the exceptional permeance-selectivity characteristics of the commercial Air Liquide (AL) polyimide membrane when operated at sub-ambient temperatures. Simulations and preliminary cost analyses show that an integrated carbon capture process scheme can take advantage of these membrane properties.

2.2 Membrane performance at cold temperatures

For most commercial membrane gas separations (CO_2/CH_4, He/N2, O_2/N_2), gas diffusion through the polymer rather than gas solubility in the polymer is the controlling phenomena determining the overall gas permeability. Solubility depends on the penetrant activity and affinity for the polymer matrix. Diffusivity depends on molecular mobility, i.e. the molecular size of the penetrant and free-volume morphology of the polymer. The permeation activation energy, ΔE_P, can be expressed in terms of the activation energy for diffusion ΔE_P and the enthalpy ΔH_S of solution [$\Delta E_P = \Delta E_D + \Delta H_S$]. Since diffusivity is usually the controlling parameter, the general rule is that overall permeability decreases and selectivity increases with a decrease in operating temperature. It is possible to deviate from this text-book rule for gases such as CO_2 which have high affinity for polyimides. In such a case, the high exothermic heat of solution compensates for the diffusion activation energy leading to lower or even negative values of ΔE_P.

Several commercial AL polyimide membranes show similar phenomena in that CO_2 permeance begins to level off as the temperature decreases. As a result, at temperatures < -20°C; the CO_2 permeance is ~ 2x higher than the value predicted by a simple Arrhenius extrapolation of ambient and higher temperature (20°-70°C) data. CO_2 /N_2 selectivity continues to increase as temperature decreases. The net effect of cold temperature operation is as if a new material had been discovered with unprecedented permeance-selectivity characteristics on the Robeson[11] trade-off plot, as illustrated in Figure 2.

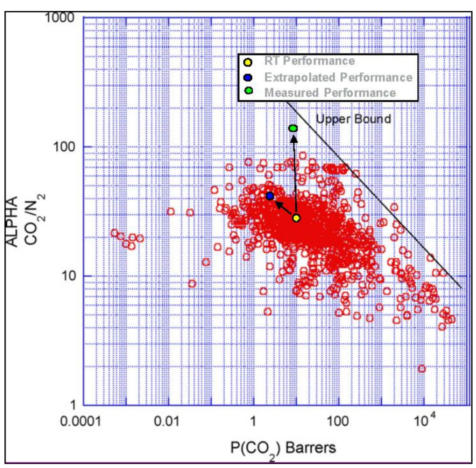


Figure 2. Plot of back-calculated equivalent permeability and selectivity at ambient temperature (yellow) and at -30°C (green) on a Robeson [11] plot for CO_2/N_2. The cold temperature estimate was made by estimating effective skin thickness of the hollow fiber from ambient temperature data for the fiber and dense film. The blue point shows the performance at -30°C predicted by extrapolation of data from 20-50°C while the green point is the actual performance at -30°C.

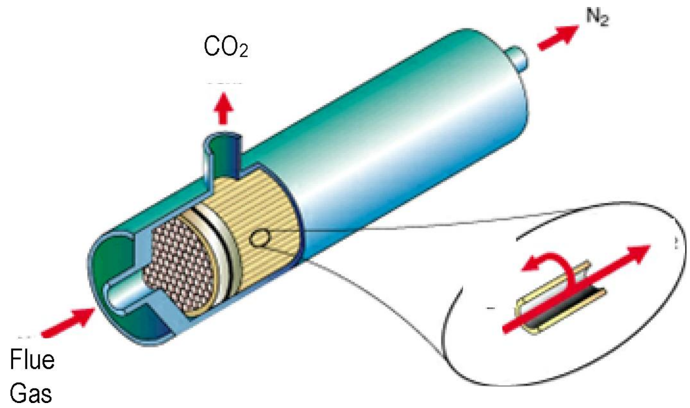


Figure 3. Air Liquide hollow-fiber membrane module for gas separation

The hollow fiber membrane module configuration (Figure 3), used by AL, is the most economic configuration in terms of cost/membrane area. This is particularly important for flue gas treatment. Due to the small hollow fiber size and the module construction method, commercial AL hollow fiber modules have an order of magnitude advantage in packing density (membrane area /module volume) over competing spiral wound configurations and an even greater advantage over plate and frame membranes. Typical AL hollow fiber modules contain as much as 10x more active membrane area compared to a typical multi-leaf spiral wound module.

Low membrane installed costs are particularly important because of the sheer volumes associated with flue gas processing. Commercial large-scale CO_2 separation membrane systems based on hollow fiber modules are operated at capacities approaching the flue gas volumes expected from power plants. For example, at a commercial Air Liquide facility in Grissik, Indonesia operating since 2000, Air Liquide membranes remove CO_2 from ~ 12,000 tons/day of natural gas [12]. The small footprint and operational simplicity of such compact membrane units is very advantageous in retrofitting existing power plants with space limitations, such as those in cities or other populated areas.

2.3 Approach

The project work is directed towards developing a CO_2 capture process based on sub-ambient temperature operation of a hollow fiber membrane. For most membrane materials, permeability decreases and selectivity increases with a decrease in operating temperature. However, laboratory measurements of the AL membranes operated at temperatures below -20°C show two to four times higher CO_2/ N_2 selectivity with minimal loss of CO_2 permeance compared to ambient temperature values. This remarkable membrane performance is the basis of a process concept shown in Figure 4.

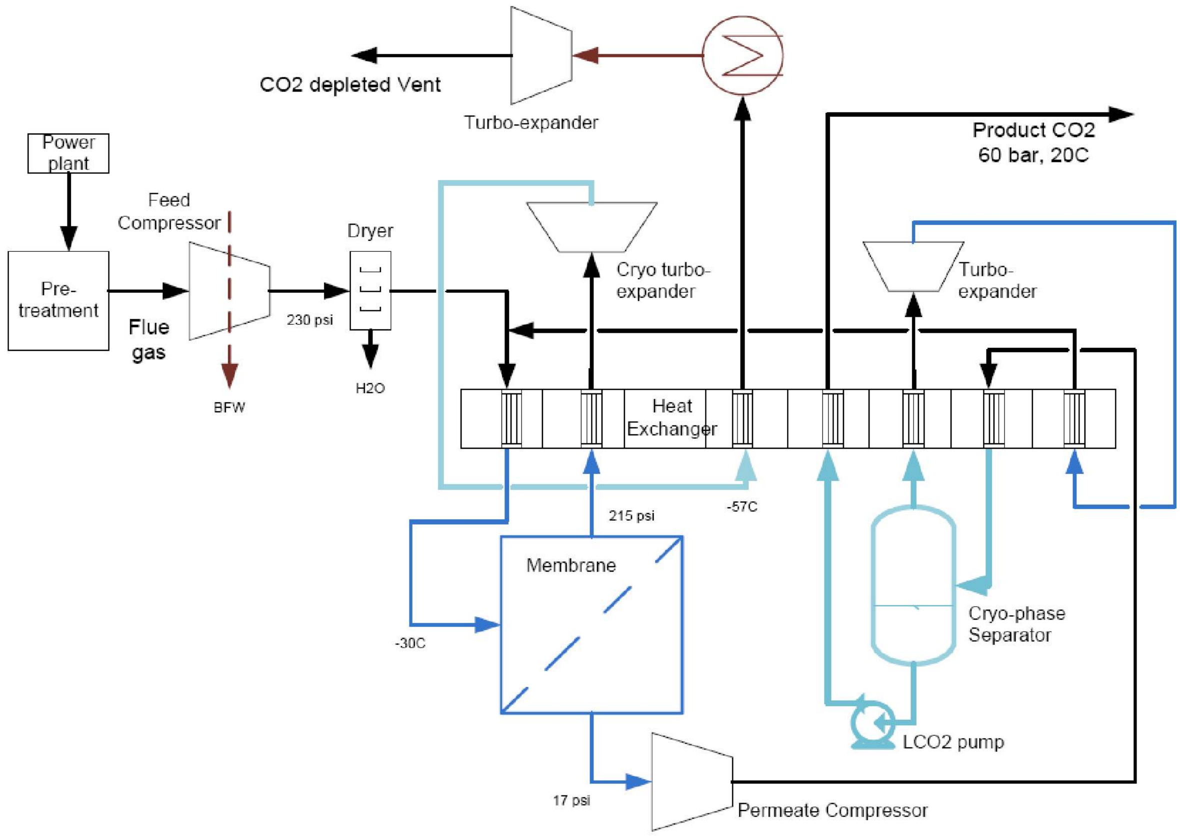


Figure 4: CO_2 Capture Process Schematic

The main areas of uncertainty are the (i) required flue gas pre-treatment before efficient compression of the flue gas, (ii) the mechanical integrity of the membrane bundle operated at -20ºC to -45ºC and (iii) the ability of commercial modules to operate at the high separation efficiency measured using mini-permeators in the laboratory. Flue gas pretreatment prior to compression are extensively addressed by AL initiatives within AL's coal oxy-combustion program. This project addresses commercial membrane operability for CO_2 capture at cold operating conditions.

The main objective of this project was to validate structural and mechanical integrity of a housed commercial membrane assembly operated at sub-ambient temperatures. The bench scale testing involves long-term, closed-loop testing of commercial fiber bundles under simulated process conditions. A bench-scale system tested commercial 6" and 12" bundles in a synthetic (clean) CO_2 / N_2 flue gas at -20° to -50°C / 1.0 to 1.7 MPa. The system operated with full gas recirculation. Membrane permeate and expanded residue streams were recycled to the compressor suction. This mode of operation decreases the operating cost of the test while validating the module operability at cold temperatures and full design flow rates.

In a parallel activity, AL measured laboratory performance of membrane mini-permeators exposed to CO_2/N_2 feed mixtures containing approximately 100 ppm of SO_2 or approximately 100 ppm of NO_x. Measurements were performed at the cold conditions similar to the commercial bundle measurements.

2.4 Organization of project Activities

Work in 4Q 2010 - 3Q 2012 was directed towards the following *Project Management Program tasks*:

Task 2.1 Design and fabrication of a closed loop sub-ambient test system for CO_2/N_2
Task 2.2: Test mechanical integrity of bundle/housing assembly at sub-ambient temperature
Task 2.3: Map bundle performance as a function of temperature, pressure and composition
Task 2.4: Demonstrate enhanced membrane performance over long term at cold temperature
Task 3.1: Modify lab cryo-test bench for low temperature SOx and NOx testing
Task 3.2: Measure SOx and NOx permeances with minipermeators
Task 4.1: Design and cost estimate for LCOE re-evaluation of conceptual process
Task 4.2: Design PFD of a slip-stream demonstration unit with NETL input

These tasks were directed towards meeting four milestones as listed below:

Milestone 1 - Complete closed loop sub-ambient temperature test system
Milestone 2 - Complete mini-permeator contaminant testing.
Milestone 3 - Complete mechanical integrity testing of the sub-ambient membrane assembly:
Milestone 4 - Complete design and budget evaluation of slipstream test system, and simulation and economic evaluation of commercial facility

The project tasks and milestones schedule are shown in Table 1. All project activities and deliverables were completed with the two year project period.

Table 1: Project Task / Milestone schedule

Task / Subtask	Task description	Project Year 1				Project Year 2				Planned start Date	Planned End Date	Actual start Date	Actual End Date
		Q1	Q2	Q3	Q4	Q5	Q6	Q7	Q8				
1	Kickoff meeting												
	PHASE 1: Experimental work												
2	Demonstrate commercial scale bundle operation at sub-ambient temperature									10/1/2010	6/30/2012	10/1/2010	6/30/2012
2.1	Design and fabrication of a closed loop sub-ambient test system for CO2/N2			M 1						10/1/2010	6/30/2011	10/1/2010	7/5/2011
2.2	Test mechanical integrity of bundle/housing assembly at sub-ambient temperature					M 3				6/30/2011	12/30/2011	7/5/2011	9/30/2011
2.3	Map bundle performance as a function of temperature, pressure and composition									1/2/2012	2/1/2012	10/14/2011	11/2/2011
2.4	Demonstrate enhanced membrane performance over long term at cold temperature									2/1/2012	6/30/2012	9/15/2011	6/30/2012
3	Laboratory Scale Flue Gas Contaminant Testing									10/1/2010	9/30/2011	10/1/2010	6/30/2012
3.1	Modify lab cryo-test bench for low temperature SOx and NOx testing									10/1/2010	4/1/2011	10/1/2010	4/1/2011
3.2	Measure SOx and NOx membrane performance on mini-permeator				M 2					4/1/2011	9/30/2011	4/1/2011	6/30/2012
	PHASE 2: Design work												
4	Sub-ambient Membrane/Cryogenic System Design									4/15/2012	9/30/2012	4/15/2012	9/30/2012
4.1	Use Phase I data to design commercial facility and estimate LCOE increase for CO2 capture.									4/15/2012	6/30/2012	3/7/2012	7/20/2012
4.2	With NETL input, Use Phase I data to design PFD of a slip-stream demonstration unit.								M 4	6/30/2012	9/30/2012	7/23/2012	9/30/2012
	In progress												
	Finished												
	Projected												

Milestones (M1-M4) are indicated in the respective quarters. Task activities are shaded as per status (in progress, finished, or projected in future).

3. Experimental Method

3.1 Design and fabrication of a closed loop sub-ambient test system for CO_2/N_2 (Task 2.1)

Design and fabrication of a closed loop sub-ambient bench scale system was completed in June 2011. The final safety review of the DRTC Experimental Risk Analysis procedure was completed and the skid was energized in July 2011. The PID of the bench scale test skid is shown in Figure 5.

The cold box contains the heat-exchanger, membrane and the J-T expansion valve. The unit was designed to operate in full recycle mode with make-up from pre-mixed feed gas cylinders in order to save operating cost. CO_2 concentrations of all three streams (feed, retentate and permeate) were continuously measured by an on-line IR analyzer skid. This analysis skid was designed so that analyzed gas is also recycled back without loss.

Skid operation was controlled by 3 parameters: feed pressure, retentate flow rate and membrane feed temperature. These were monitored by sensors connected to a PLC and adjusted by modulating the three flow control valves shown in Figure 5. Feed gas at the desired composition was metered into the recirculation loop near the compressor suction in order to compensate for small losses through leaks etc.

Tests were conducted with commercial 12” and 6” Medal commercial membrane bundles. For the 12” bundle, the feed and residue flow rates were measured by Coriolis flow meters and all three streams (feed, residue and permeate) were analyzed by IR for CO_2 content. For the 6” bundle, all three streams were measured in terms of both flow rate and composition. The mass balance error was typically less than 1%. Various data reconciliation schemes were evaluated with the assistance of the DRTC Applied Mathematics Group. The most robust calculation scheme used the 3 concentrations and the averaged feed rate to back-calculate the membrane permeances.

Though the membrane is located in a cold box, the energy for cooling the feed stream mainly comes from Joule-Thomson expansion of the pressurized residue gas. This ‘self-refrigeration” scheme with expansion of the residue stream was found effective, even using relatively inefficient J-T cooling across the residue expansion valve. As shown in Figure 6a, the rate of system cooling is fast. Figure 6b shows the approach temperature in the heat exchanger as a function of the membrane feed temperature.

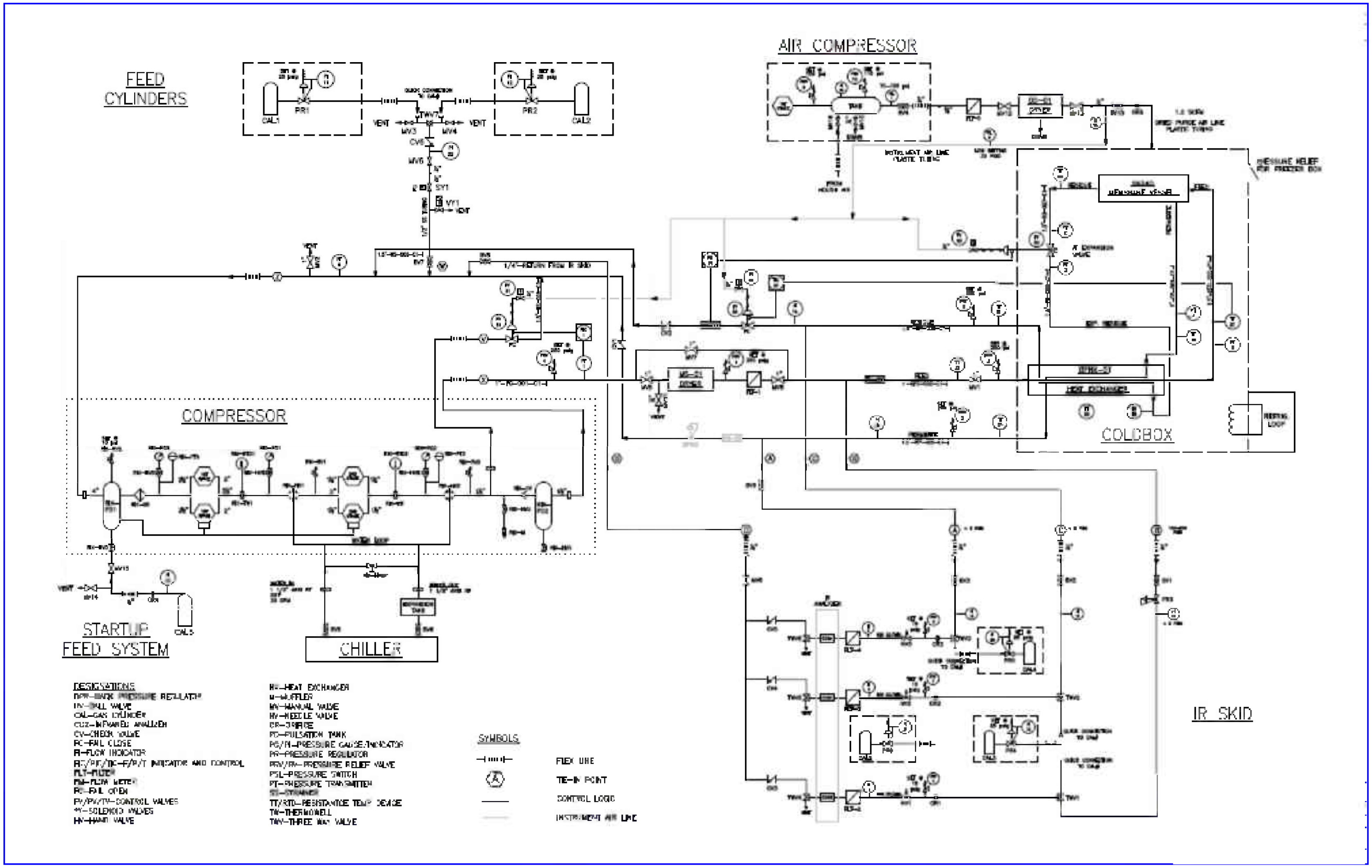
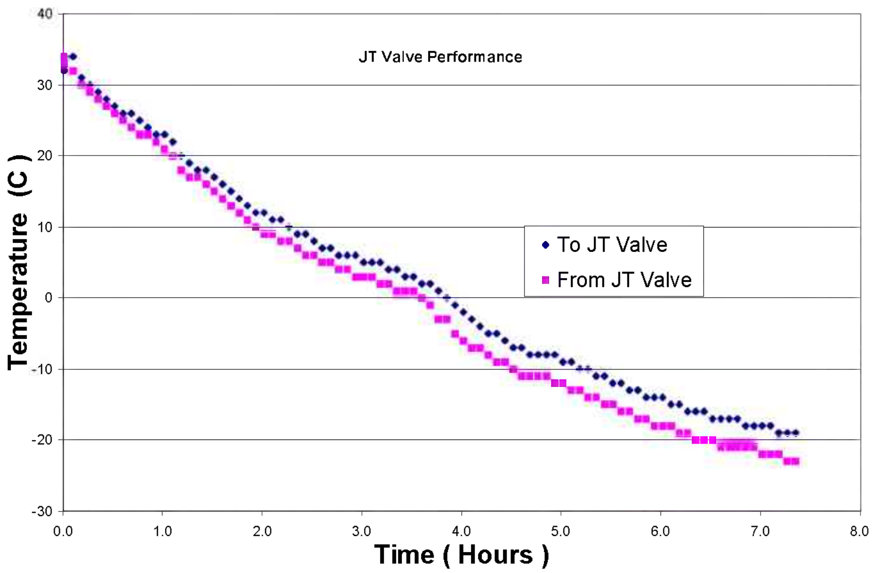


Figure 5. PID of integrated bench scale test skid



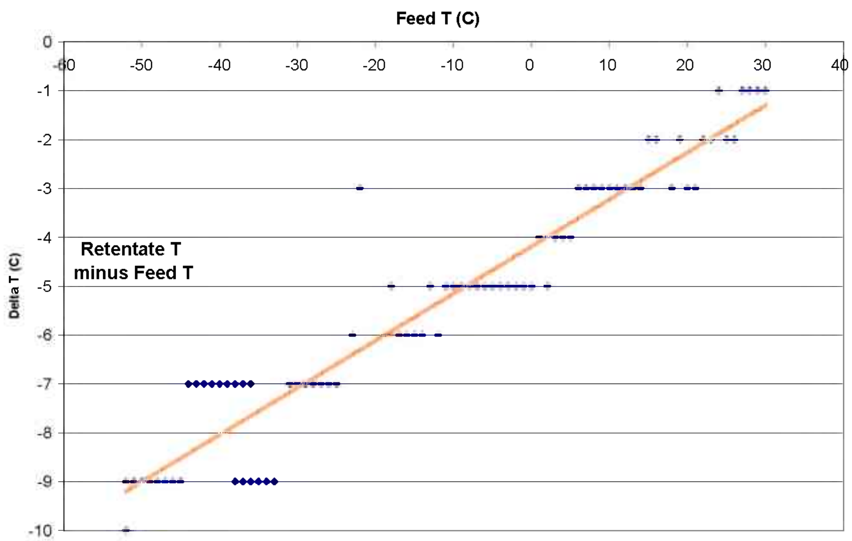


Figure 6. (6a) Feed gas temperature as a function of time on initial cool-own.
(6b) Heat exchanger approach temperature as a function of feed gas temperature.

3.2 Laboratory scale flue gas contaminant testing (task 3.1)

The existing laboratory cold test facility at DRTC was modified for safe testing of feeds containing toxic components. The schematic for cold membrane test system for feed gases with toxic components SOx and NOx is shown in Figure 7.

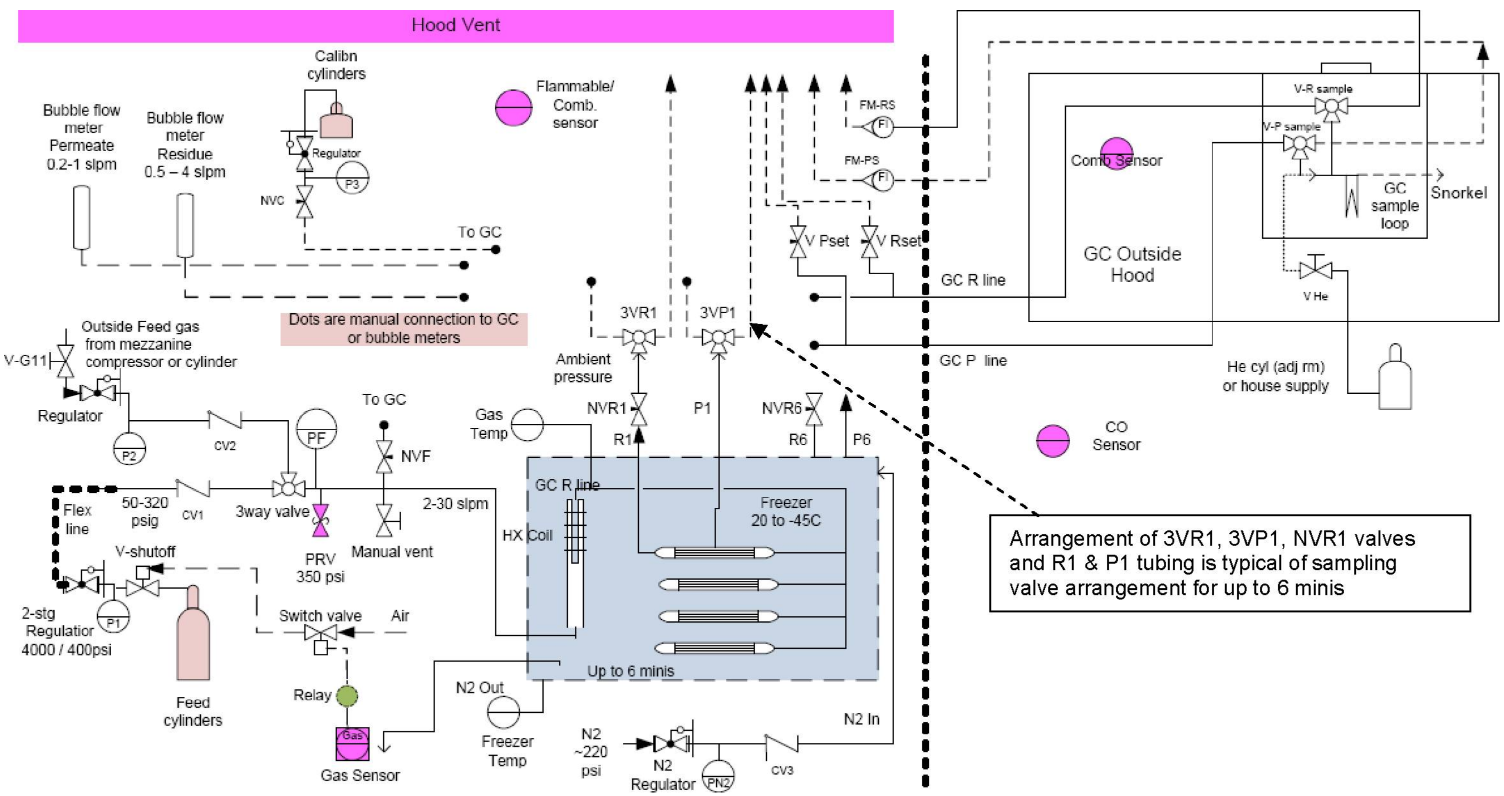


Figure 7. Schematic of cold membrane test system for feed gases with toxic components (SO_2, NO)

4. Results and Discussion

4.1 Mechanical integrity test of bundle/housing assembly at sub-ambient temperature (task 2.2)

A commercial 12" MEDAL bundle was purchased. The standard quality control air test confirmed a productivity / recovery separation performance typical of this MEDAL product line. The O-rings for this bundle were selected for cold temperature use.

Differential rates of thermal contraction upon cooling could potentially create leakage paths in the membrane and /or vessel assembly. The membrane assembly consists of many material types with varying thermal expansion coefficients: metal (housing and center tube), thermosets (epoxy, O-rings), thermoplastics (O-rings, membrane fiber) etc. Hence,the goal of the mechanical integrity test was to verify (and if necessary resolve) the proper functioning of the assembly components at cold conditions. This testing was done with a 12" bundle as this geometry would put the highest stress on these components.

CO_2/N_2 testing at sub-ambient condition was carried out between 14-July-2011 to 01-September-2011. Justification for passing the mechanical mechanical integrity test is based on the following observations:

A. **Constant performance over time**

Over the test period, the bundle was exposed to pressures as high as 15 bar and temperatures down to -40ºC routinely (with excursions down to -60ºC). The system also experienced several shutdowns / re-starts especially in the early phases of testing. Each shutdown involves a rapid de-pressurization of the system, though the bundle temperature remains relatively stable. As shown in Figures 8-10, the calculated bundle performance and especially the selectivity was the same between the first testing day (7/14) and 4 weeks later (8/10)

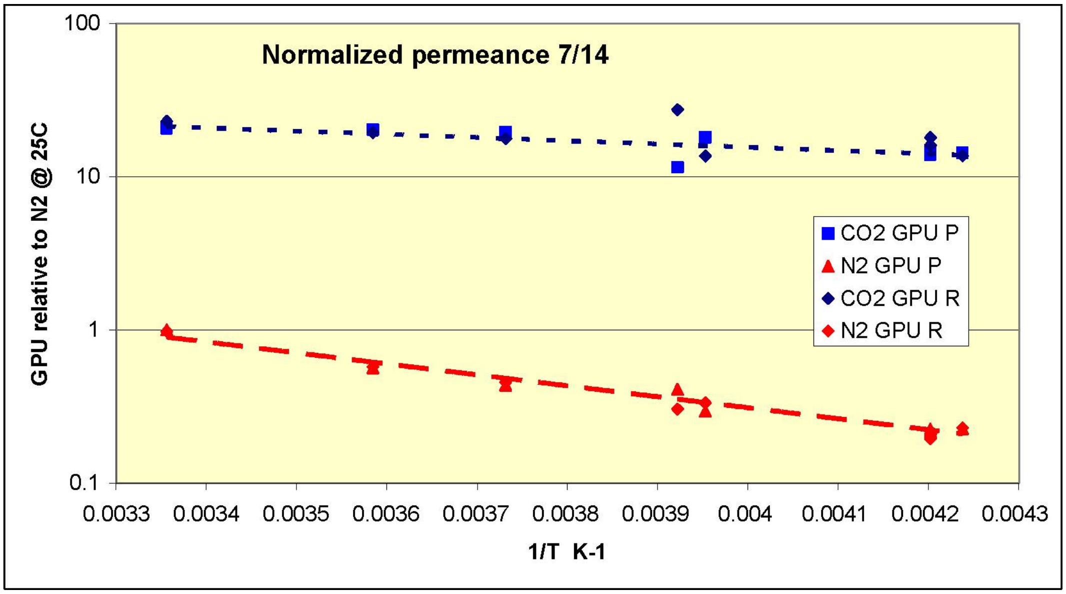


Figure 8. Normalized CO_2 and N_2 permeance at 200 psi, 18% CO_2/N_2 as a function of feed temperature on 14-July-2011. The permeance values are calculated from either permeate (P) or residue (R) compositions and then normalized as per initial ambient temperature N_2 permeance.

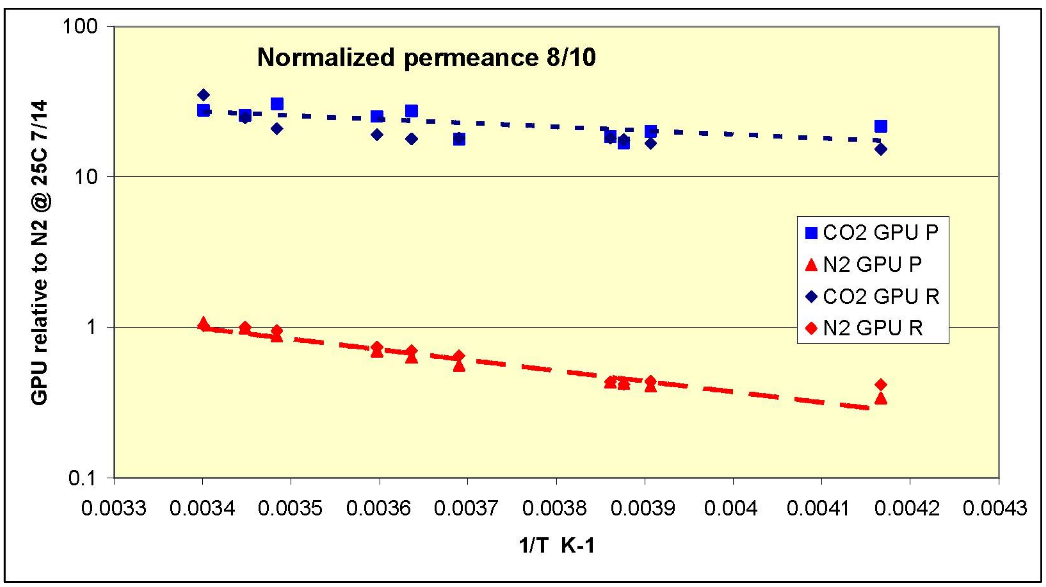


Figure 9. Normalized CO_2 and N_2 permeance at 200 psi, 18% CO_2/N_2 as a function of feed temperature on 10-August-2011. The permeance values are calculated from either permeate (P) or residue (R) compositions and then normalized as per initial ambient temperature N_2 permeance on 14-July-2011.

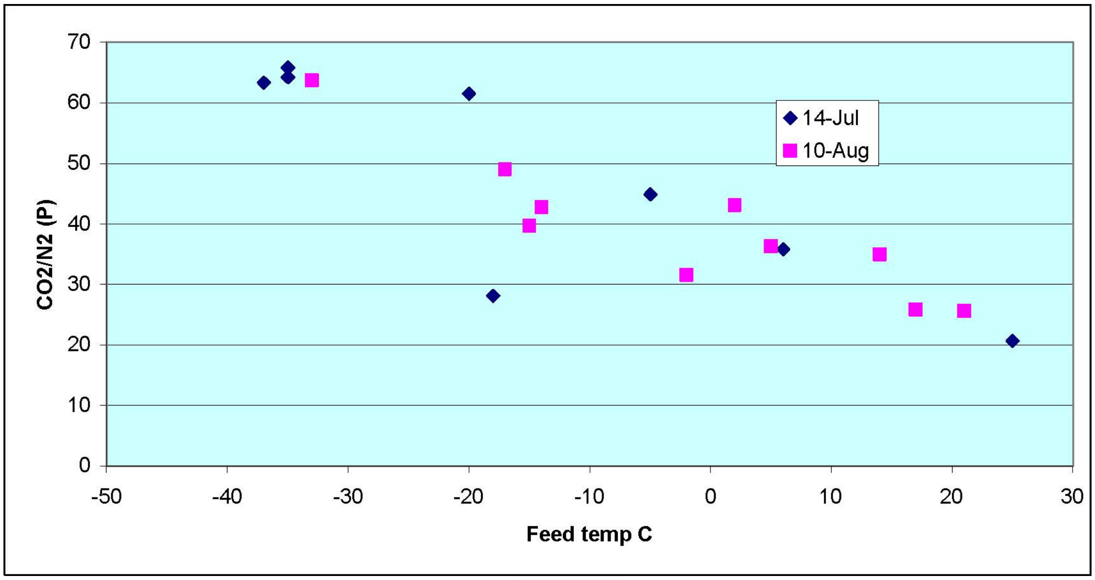


Figure 10. CO_2 / N_2 selectivity at 200 psi, 18% CO_2/N_2 as a function of feed temperature; comparison of data taken on 14-July -2011 and 10-August-2011.

B. Constant performance with O-ring change

In order to examine the possibility that the O-rings may have been compromised in use, the O-rings sealing the permeate from the pressurized sides were replaced on 16-August-2011. This replacement made minimal change in the membrane performance. In addition, diagnostic measurements showed no change:

- SF_6 "permeance" before and after change remained higher than expectations
- N_2 permeance measured with pure gas (< 0.3% CO_2) at ~ 17°C / 175 psi was within experimental error before and after the O-ring change

C. Constant permeance of tracer molecule (SF_6)

Low concentrations (~ 0.05 to 0.2%) of SF_6 tracer were fed into the recirculating gas. Residue and permeate samples were collected in sample bombs and analyzed by the DRTC Analytical Group using an FTIR technique. These measurements were used to calculate the "SF_6 permeance" through the bundle. Since SF_6 is a large molecule, its intrinsic permeance is very low. Thus a measurable "SF_6 permeance" indicates a "leak" transport pathway by a mechanism other than solution-diffusion though a defect-free bundle.

These samples were taken over the second month of 12" bundle testing. There was no trend in the "SF_6 permeance" values over this time. However, the permeance was about 10x higher than would be expected based on the theoretical value. This indicates the presence of defects in a commercial bundle that are not present in laboratory scale mini-permeators. The leakage through these defects is too small to be apparent at the lower selectivities ($\alpha(CO_2/N_2)$ =25) at ambient temperature, but could explain the lower than expected CO_2/N_2 selectivity at -40°C ($\alpha(CO_2/N_2)$ =65) .

The SF_6 measurements with the 12" bundle left open the possibility that the SF_6 permeance was in fact low initially before rising to the values measured in the 2nd month. However, similar measurements were made immediately after installation of the 6” bundle. This initial permeance rate with the 6” bundle was similar to that measured with the 12” bundle; thus strongly suggesting that the 12” bundle was not damaged during its test.

D. 12” bundle performance and replacement of 12” bundle with 6” bundle

The 12” membrane bundle exhibited similar temperature response as previously observed for laboratory minipermeators. As the operating temperature was decreased from ambient to -40°C, CO_2 permeance decreased by only 15% compared to the ambient temperature value while N_2 permeance decreased by approximately 300%.

Though the mechanical integrity test was passed, the calculated selectivity through the 12” bundle was less than expected ($\alpha CO_2/N_2$ ~ 65 vs 90). There are several possible causes

- ✔ Possible leak in a small fiber section of the bundle (indicated by low pressure reverse pressurization testing)
- ✔ Bundle non-ideality (deviations from ideal counter-current flow in bundle)
- ✔ CO_2 pinch behavior with high CO_2 recovery

The last two (non-artifact) reasons are potentially important and justified further study. However, the 12” permeation rate was too high for the existing compressor flow to achieve the low stage cut conditions needed for baseline bundle performance and study non-ideality issues. Moreover, at these test conditions, the back-calculation of membrane permeance is more sensitive to small experimental errors and numerical methods.

The 12” bundle was removed and sent to MEDAL for air testing and further leak checking. This final testing matched the initial QC results within experimental error. The 12” bundle was then replaced, as planned with a smaller 6” bundle, which can be operated at lower stage cuts in the pilot system. The 6” bundle testing was used to generate the performance map needed for process design.

4.2 Parametric study to map bundle performance (task 2.3)

As discussed in the previous section, the 12” bundle permeation rate was too high for the existing compressor flow to achieve the low stage cut conditions needed for an accurate measurement of baseline bundle performance and study of non-ideality issues. Moreover, at these operating conditions, the back-calculation of membrane permeance is more sensitive to small experimental errors and choice of numerical methods. The 12” bundle was replaced with a smaller 6” bundle that can be operated at lower stage cuts in the pilot system.

Parametric studies were carried out both towards the start and end of the long term test. After the initial cool-down period, the bundle was operated at -40°C / 200 psi. These operating conditions are identical to the operating conditions for 12” bundle testing. The performance of both bundles was analyzed by AL proprietary software that allows an estimation of bundle non-ideality effects in combination with the intrinsic fiber performance. Diagnostic permeation tests were also run on the 6” bundle, similar to previous tests with the 12” bundle. These analyses suggest that under the same operating conditions, the fiber in both bundles have similar separation performance. The improved performance of the 6" bundle relative to the 12" bundle is attributable to the higher degree of ideality in the 6" bundle.

A. Parametric Test 1 - Task 2.3 requires measurement of bundle performance map where permeance and selectivity of the membrane bundle are measured as a function of temperature, pressure, and CO_2 composition.

The feed conditions were varied over the ranges shown in Table 2 below. The variation of these parameters over time is shown in Figure 11:

Table 2. Feed conditions variation

Inlet Pressure psi	Inlet Temperature (°C)	CO_2 % in Feed Gas
160 and 200	-25, -35 and -45	12 and 18

In addition to these variables, performance scans were performed as a function of feed flow / stage cut to simultaneously determine the importance of bundle non-ideality. Stage cut is the ratio of permeate / feed flows; higher stage cut corresponds to higher CO_2 recovery in permeate. Variation in stage cut also changes the feed-side CO_2 activity profile through the membrane bundle.

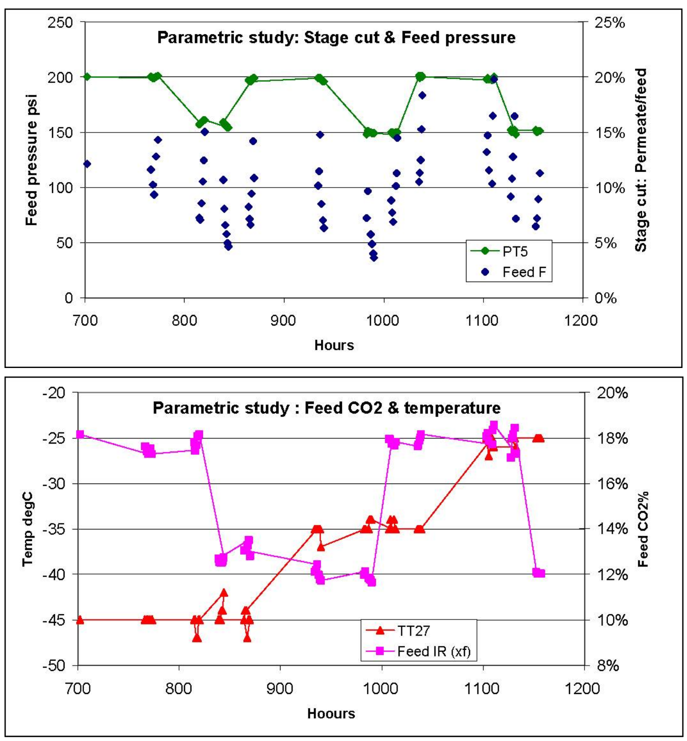


Figure 11: Parametric Variation for Performance Map with 6" Bundle Testing

Feed Temperature: -45C, -35 and -25C

Feed Pressure: 200 and 160 psig

Feed Concentration: 18% and 12% CO_2

Representative plots of the obtained data are shown in:

- Figure 12: CO_2 permeance and CO_2 / N_2 selectivity as a function of stage cut at -45°C, 200 and 160 psi, and 18% and 12% CO_2 concentration. This figure illustrates the effect of the feed CO_2 concentration.
- Figure 13: CO_2 permeance and CO_2 / N_2 selectivity as a function of stage cut at -45°, -35°, and -25°C, 200 psi, and 18% CO_2 concentration. This figure illustrates the beneficial effect of the colder feed temperature

The reported CO_2 permeance is normalized by the initial, ambient temperature data in these figures.

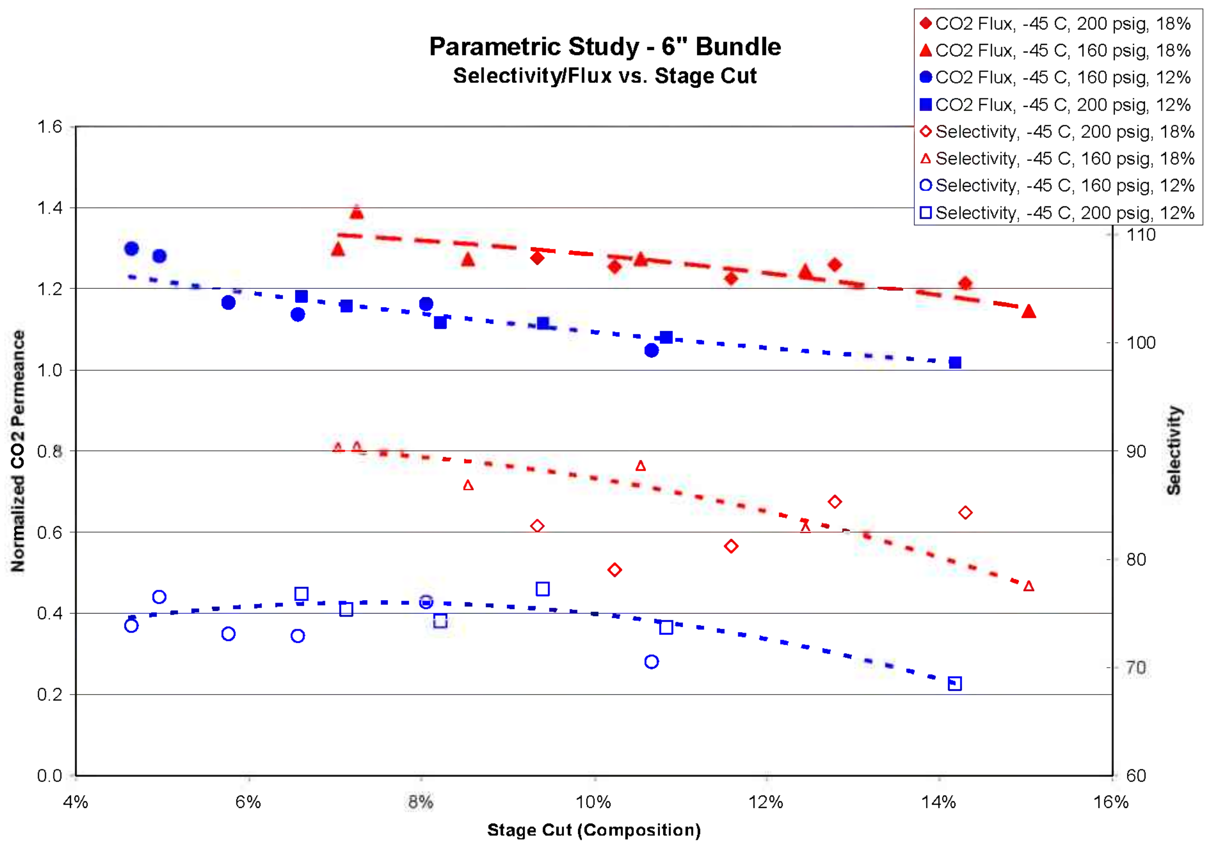


Figure 12: CO_2 Permeance and CO_2 / N_2 Selectivity as a Function of Stage Cut at Feed Temperature of -45°C, Feed Pressures of 200 and 160 psi, and Feed CO_2 Concentrations of 18% and 12%

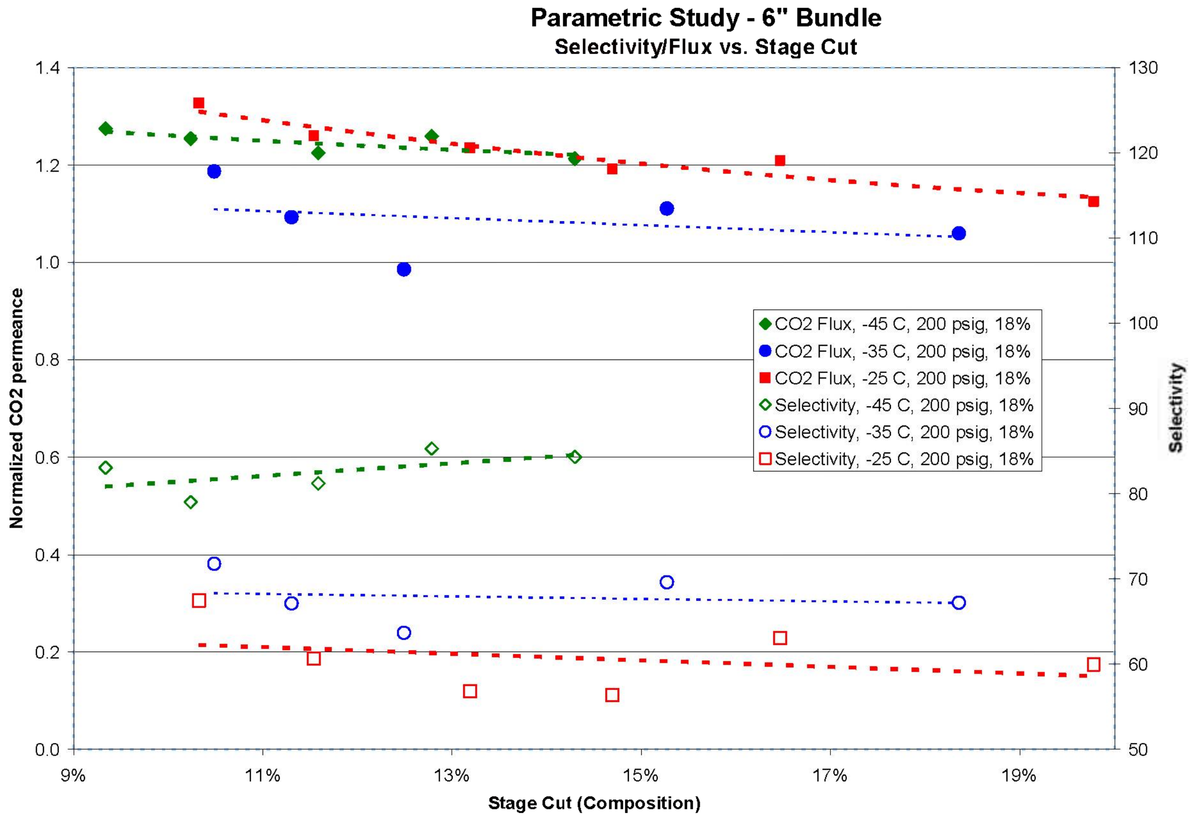


Figure 13: CO_2 Permeance and CO_2 / N_2 Selectivity as a Function of Stage Cut at Feed Temperatures of -45°, -35°, and -25°C at 200 psi Feed Pressure, and 18% CO_2 feed concentration

B. Parametric Test 2 - This mapping study was replicated at the end of the long-term stability test. These data was used in Task 4 for determining optimum process parameters. The effect of CO_2 feed side activity at -45°C is illustrated in Figure 14. This figure shows the calculated bundle CO_2 permeance and selectivity as a function of stage cut for the 18% and 12% CO_2 feed concentration cases. Higher CO_2 activity corresponds to both higher CO_2 permeance and selectivity. Thus, higher feed pressure has three benefits at cold temperature operation; the first two are well known while the third is unique to cold temperature operation:

(i) lower membrane area requirement due to higher CO_2 partial pressure difference across the membrane
(ii) higher CO_2 purity in permeate due to higher pressure ratio across the membrane
(iii) Intrinsically higher CO_2 permeance and selectivity at higher CO_2 activity

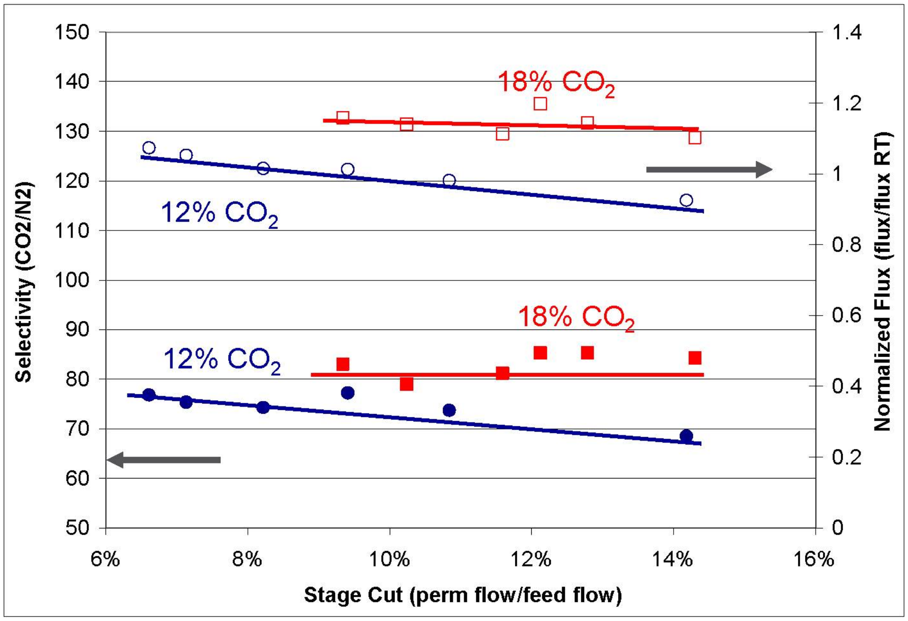


Figure 14: CO_2 Permeance and CO_2/N_2 Selectivity as a function of feed CO_2 concentration (12% or 18%) as a function of stage cut; at -45°C / 200 psi. The right side Y-axis shows normalized permeance (CO_2 permeance at cold conditions /permeance at initial ambient temperature).

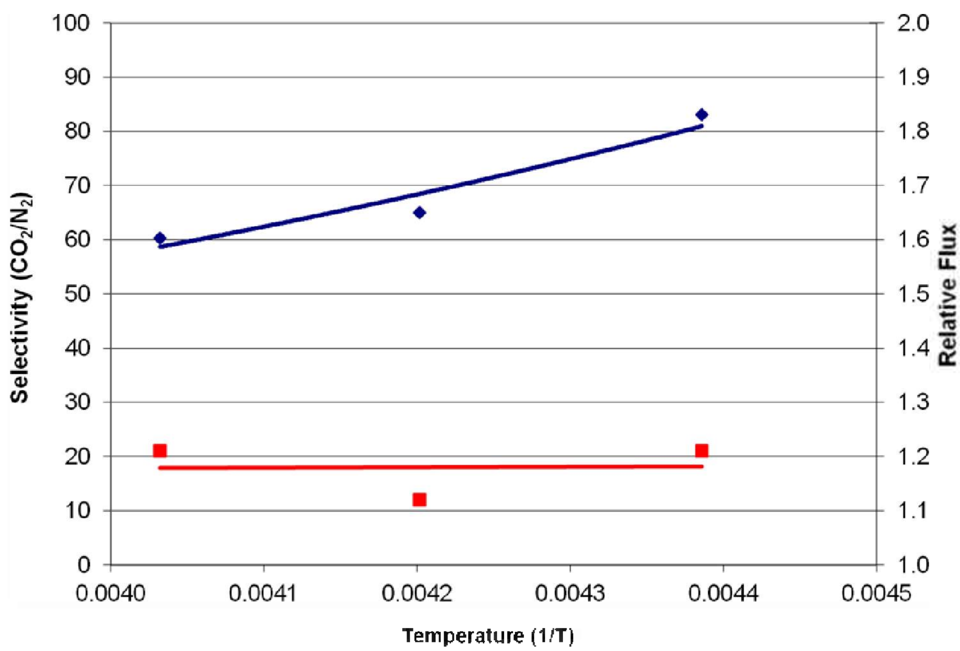


Figure 15 : Final parametric study: temperature dependence of membrane permeance-selectivity at 200 psi, 18%feed CO_2.The right side Y-axis shows normalized permeance (CO_2 permeance at cold conditions /permeance at initial ambient temperature).

The effect of temperature on the membrane performance in the final parametric test is illustrated in Figure 15. As expected CO_2/N_2 selectivity increases with decreasing temperature. However, unique to the cold membrane operation, the CO_2 permeance at the coldest temperature (-45°C) is similar to that at -25°C. It can also be seen that at these cold temperatures (-25° to -45°C), the membrane permeance is in fact higher than that measured initially at ambient temperature (normalized CO_2 permeances > 1).

The effect of temperature (-25° to -45°C) is also illustrated in Figure 16 which shows the achieved CO2 % in permeate as a function of the CO2 recovery. CO2 recovery is related to stage cut; specifically:

CO2 recovery = CO2 in permeate / CO2 in feed

= stage cut x (%CO2 in permeate / %CO2 in feed)

The data plotted in this figure are for a feed concentration of 18% CO2 at 200 psi. The data were obtained over a range of CO2 recoveries from 44% to 85%. At a fixed pressure / temperature condition, the CO2 recovery from the feed gas can be increased as required by decreasing the feed flow rate. As discussed previously, calculations of the intrinsic membrane performance at high CO2 recovery operation are less accurate. While there is no problem to operate at CO2 recoveries of 90% or even higher, the accuracy of the membrane performance estimates falls off as the CO2 driving pressure across the membrane decreases.
Superimposed on these 6" bundle performance curves are the more limited recovery range data (71 to 85%) with the 12" bundle at -40°C / 18% CO2 / 200 psi. The range of CO2 recovery conditions with the 12" bundle was limited by the compressor capacity.

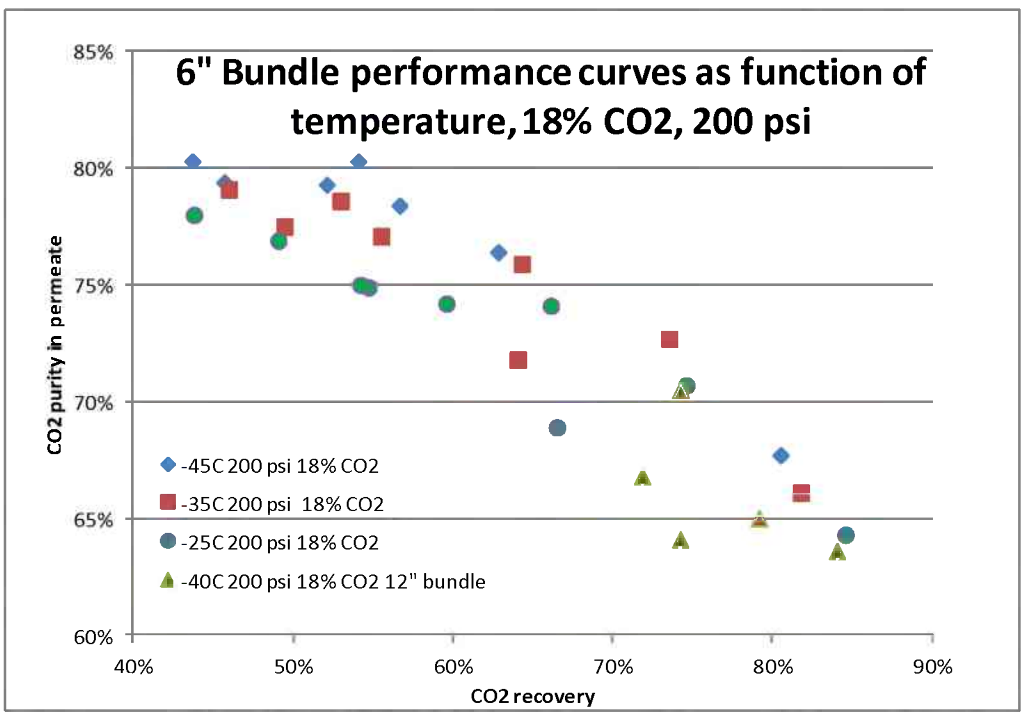


Figure 16 : Temperature dependence of 6" bundle membrane performance shown as plots of % CO2 in permeate as a function of CO2 recovery in the membrane at 200 psi, 18%feed CO_2. Data curves are at -25º, -35º and -45ºC. This plot also includes data for the 12" bundle at -40 ºC, 200 psi, 18%feed CO_2.

Taken all together, these tests indicate that the membrane performance is best at the coldest temperature and highest feed pressure that can be achieved. In this testing, the minimum temperature and highest feed pressure were limited respectively by the membrane vessel rating and by the compressor capability. In terms of the process choice, the optimized variables also depend on the energy costs of achieving these desirable pressure / temperature conditions.

4.3 Long term test with 6" Bundle (Task 2.4)

Following the completion of the performance map with the 6" bundle, operating conditions were returned to the "optimum" feed conditions of -45°C / 200 psi / 18% CO_2 in N_2 to conduct long-term test (Task 2.4) at these conditions. The timing of the two parametric studies is indicated by bars on Figure 17 which shows the entire history of the long term testing.

The test data are shown in the Appendix. The majority of the testing was run at 200 psi, -45°C and 18% CO_2 feed. These pressure and temperature conditions were indicated to be the optimum by the first parametric study. These choices of conditions were validated by the second and final parametric test.

Between these two tests, the membrane underwent a long term stability evaluation. This stability test is the main experimental achievement of the test program. The object of the stability test was to:

- evaluate stability of membrane performance over approximately 6 months operation at sub-ambient temperature
- establish a conditioned membrane baseline prior to the final parametric study.

The data quality from the 6" bundle is more accurate for membrane performance analysis than the 12" bundle primarily because the 6" bundle can be operated at low stage cut. The results showed no degradation of the excellent separation performance at -45°C.

In the "standard" operating mode, the feed concentration of 18% CO_2 is reduced to about 9% in the residue stream. The CO2 recovery during this standard operating mode was ~ 50-55%. Bundle performance may be over-stated due to the high CO_2 activity through the entire bundle length at the standard measurement conditions. Operating at low stage cut has the unintended consequence of a higher CO_2 concentration in the residue stream than high recovery operation. To compensate for this potential problem, measurement conditions were altered to measure bundle performance at both high and low stage cut during the latter part of the test. In the latter part of the exposure, stage cut was increased (by decreasing feed flow) to achieve a residue concentration of about 4% CO_2. The CO2 recovery during this operating mode was ~ 78-84%. As previously discussed, still higher CO2 recoveries are possible by further decreasing the feed flow, but the permeation performance back-calculation is then less accurate.
Periodically, membrane performance was re-measured at standard conditions to track performance over time. The excellent performance stability of the 6" bundle over this 6 month test bears out similar data with mini-permeators tested in the lab for more than 1 year.

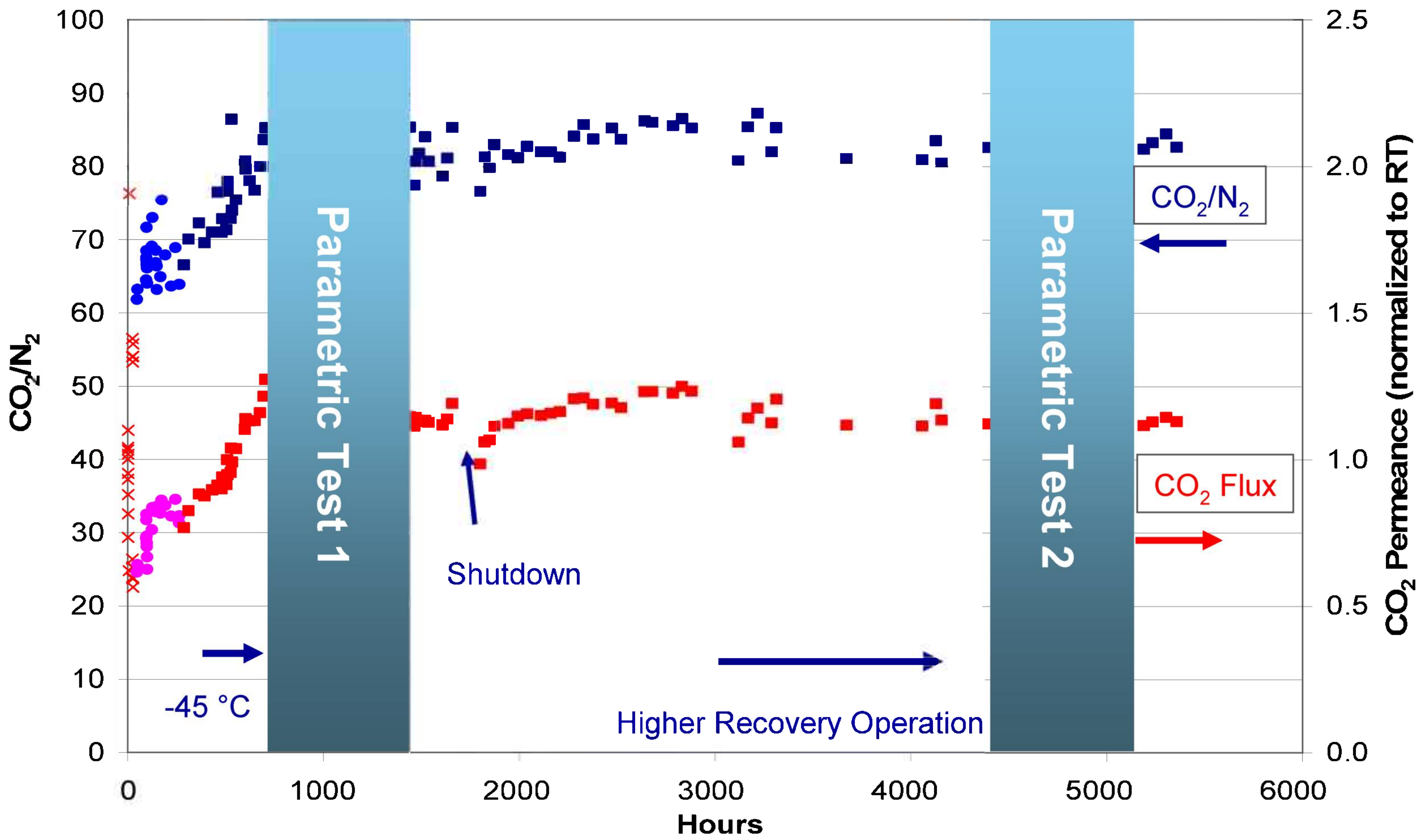


Figure 17: CO_2 Permeance and CO_2/N_2 Selectivity for 6" Bundle Testing. Normal test conditions: -45C, 200 psig, 18% CO_2 in feed. The right side Y-axis shows normalized permeance (CO_2 permeance at cold conditions /permeance at initial ambient temperature). The bundle was operated with an average CO2 recovery of ~ 80% after 3000 hours and ~ 50% at previous times.

4.4 SOx and NOx membrane performance measurement on mini-permeator (Task 3.2)

Attempts were made initially to perform lab permeation measurements with CO_2/N_2 feed mixtures containing 100-1000 ppm SO_2. The initial results were not reliable due to SO_2 analysis inconsistencies. Though permeance could be calculated from these measurements, the mass balance errors were too high for these estimates to be trustworthy. Measurements with higher concentration (2% SO_2) containing mixtures indicated that the problem was not intrinsic to the permeation system but rather arose from inaccuracy of the GC used for the analysis. A new microGC was set up and measurements completed with the originally proposed 100 ppm SO_2 feed mixture. Permeance measurements using laboratory permeators were completed with CO_2/N_2 feed mixtures containing ~ 100 ppm SO_2, or NO_2, or NO.

The SO_2 measurements were made on a laboratory prepared mini-permeator with 0.43 ft^2 area operated with bore feed. Before testing with the SO_2 mixtures, the mini was tested with 18% CO_2 in N_2 at -40°C / 200 psi for 5 days. It was verified that the mini was operating at the expected performance (CO_2/N_2 ~ 130) before starting the SO_2 measurements.

The SO_2 measurements were made at 140 psia using a feed mixture of 20% CO_2 and 100 ppm SO_2 in N_2 over a temperature range of -40° to 15°C. Calibration mixtures with 25 ppm and 1000 ppm SO_2 were used to calibrate the SO_2 concentration readings in residue and permeate streams. The mini-permeator was operated in this mode with a stage cut varying from 10% – 15%. The results are summarized in Figure 18 below.

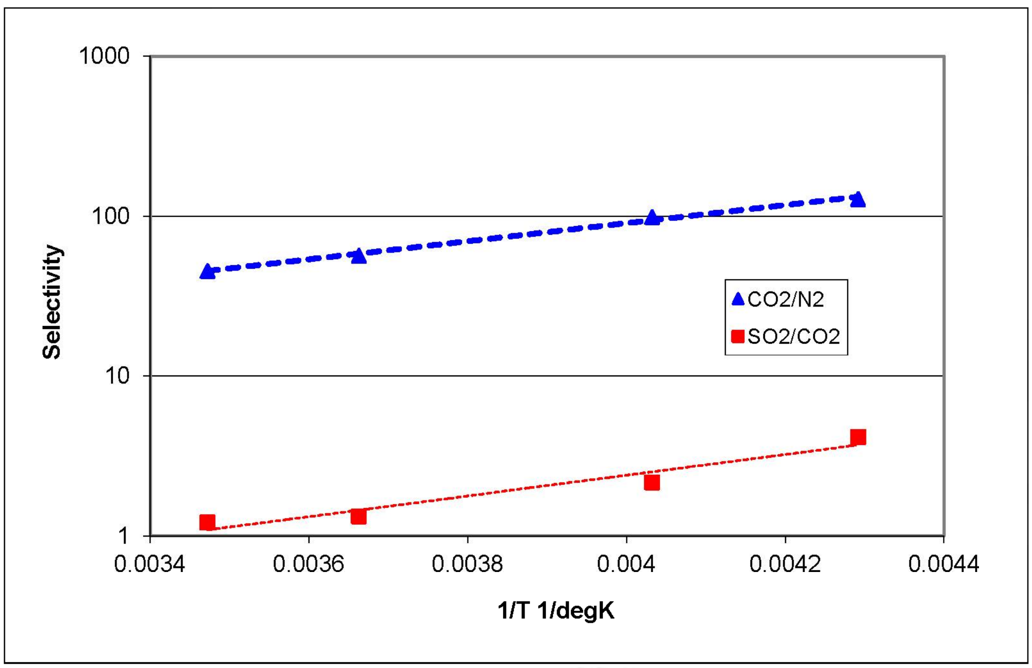


Figure 18: Temperature dependence of CO_2/N_2 and CO_2/SO_2 Selectivity for Laboratory Permeator. 100 ppm SO_2, 20% CO_2 in N_2 at 140 psi

SO_2 permeance does not decrease monotonically in this temperature range. While SO_2 permeance at 15°C is similar to CO_2, the value at -40°C is approximately 4x higher than CO_2 permeance. This implies that SO_2 in the feed flue gas will be efficiently removed into the membrane permeate in our proposed process.

NO_2 measurements were made at 145 psia using a feed mixture of 20% CO_2 and 100 ppm NO_2 in N_2 over a temperature range of -40° to 15°C. Component compositions were analyzed by a microGC with a thermal conductivity (GC/TCD) and He as the carrier gas. Calibration mixtures with 1000 ppm NO_2 were used to calibrate the NO_2 concentration readings in the permeate streams. NO_2 measurements were more difficult than similar SO_2 measurements and the low residue concentrations (< 25 ppm) could not be accurately measured. To overcome this problem, the mini-permeators were operated at low stage cuts and permeance values were calculated from the flow rates, feed composition, and permeate composition.

The NO measurements were made at 140 psia using a feed mixture of 20% CO_2 and 100 ppm NO in N_2 over a temperature range of -40°, -10°, and 21°C. Analysis by GC/TCD was not possible because of the low thermal conductivity difference between NO and available carrier gases. With the assistance of the DRTC Analysis group, compositions were measured by FTIR. Calibration mixtures with 25 ppm and 674 ppm NO were used to calibrate the NO concentration readings in residue and permeate streams.

The overall results are summarized in Figure 19, which shows the measured permeation values as a function of temperature for SO_2, NO_2, NO, and N_2 relative to CO_2. Both SO_2 and NO_2 are more permeable than CO_2. These contaminants will be efficiently removed into the membrane permeate in the proposed process. NO is approximately 7x faster than N_2. NO compositions are not expected to change substantially through the membrane unit. However, the true NO_x distribution behavior will be more complicated due to the NO-O_2-NO_2 equilibrium coupled with kinetic effects from the residence time at high pressure.

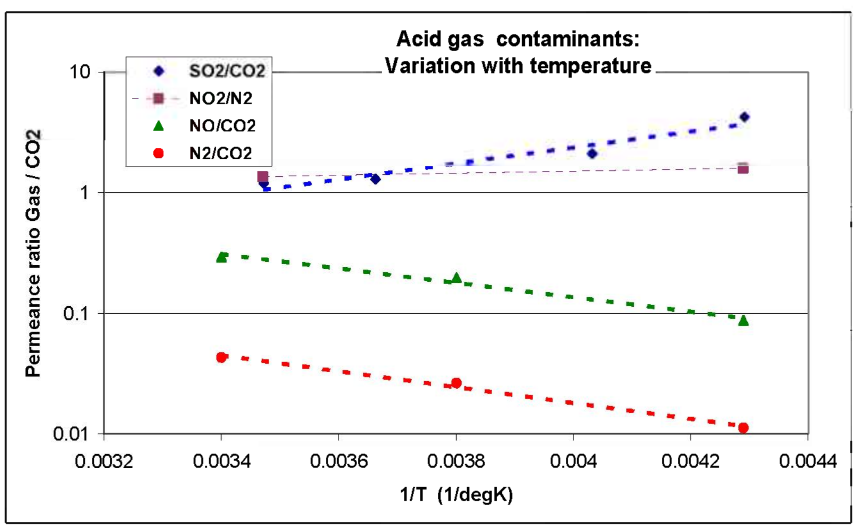


Figure 19: Temperature dependence permeance ratio (Gas permeance / CO_2 for SO_2, NO_2, NO, and N_2. Data based on laboratory mini-permeator studies with 20% CO_2 in N_2 at ~ 140 psia containing 100 ppm of each acid gas

4.5 Commercial facility design and LCOE re-evaluation (Task 4.1)

The process viability to treat flue gas from a net 550 MW power plant was initially estimated at the time of proposal submission in 2010 (Project Narrative). The LCOE was re-evaluated in task 4.1 with more accurate estimates of the plant cost and membrane performance.

The previous LCOE calculation basis was taken from the DOE/NETL study 2007/1291 *Pulverized Coal Oxycombustion Power Plants (Revision 2, August 2008).* Case 1 (supercritical boiler without CO_2 capture) and Case 3 (supercritical boiler with CO_2 capture with the EconamineTM process) were used to develop LCOE costs for the cold membrane process. For the revised LCOE, we used as a basis the more recent DOE/NETL study 2010/1397 *Cost and Performance Baseline for Fossil Energy Plants Volume 1: Bituminous Coal and Natural Gas to Electricity (Revision 2, November 2010*). Cases 11 and 12 of this new study represent the base case without CO_2 capture and the amine case with 90% CO_2 capture, respectively.

The LCOE re-evaluation procedure relied on combining the input from 2 acitivities:
- Engineering validation of the process simulation scheme
- Capital cost estimates for equipment

4.5.1. Process scheme

DRTC executed an Engineering Services Contract with Air Liquide Engineering (ALE, Champigny France) for design and cost estimation of the cold membrane process for CO_2 capture at a 550 MW net air-fired coal power plant. The ALE group has previous experience in CO_2 capture technologies and directs coal oxy-combustion activities for AL. A block diagram of the process is shown in Figure 20.

The engineering assessment included a better estimation of utility requirements (cooling water, steam for dryer regeneration); these are not shown in Figure 19 for reasons of clarity.

. With DRTC support, ALE developed cost estimates for individual process blocks:
- SO_2 scrubbing tower
- Low pressure filtration and de-saturation
- Compressor technology to deliver 16 bar
- BFW heat exchanger(s)
- Dehydration and contaminant removal at 16 bar
- Optimized BAHX arrangement
- Permeate compressor

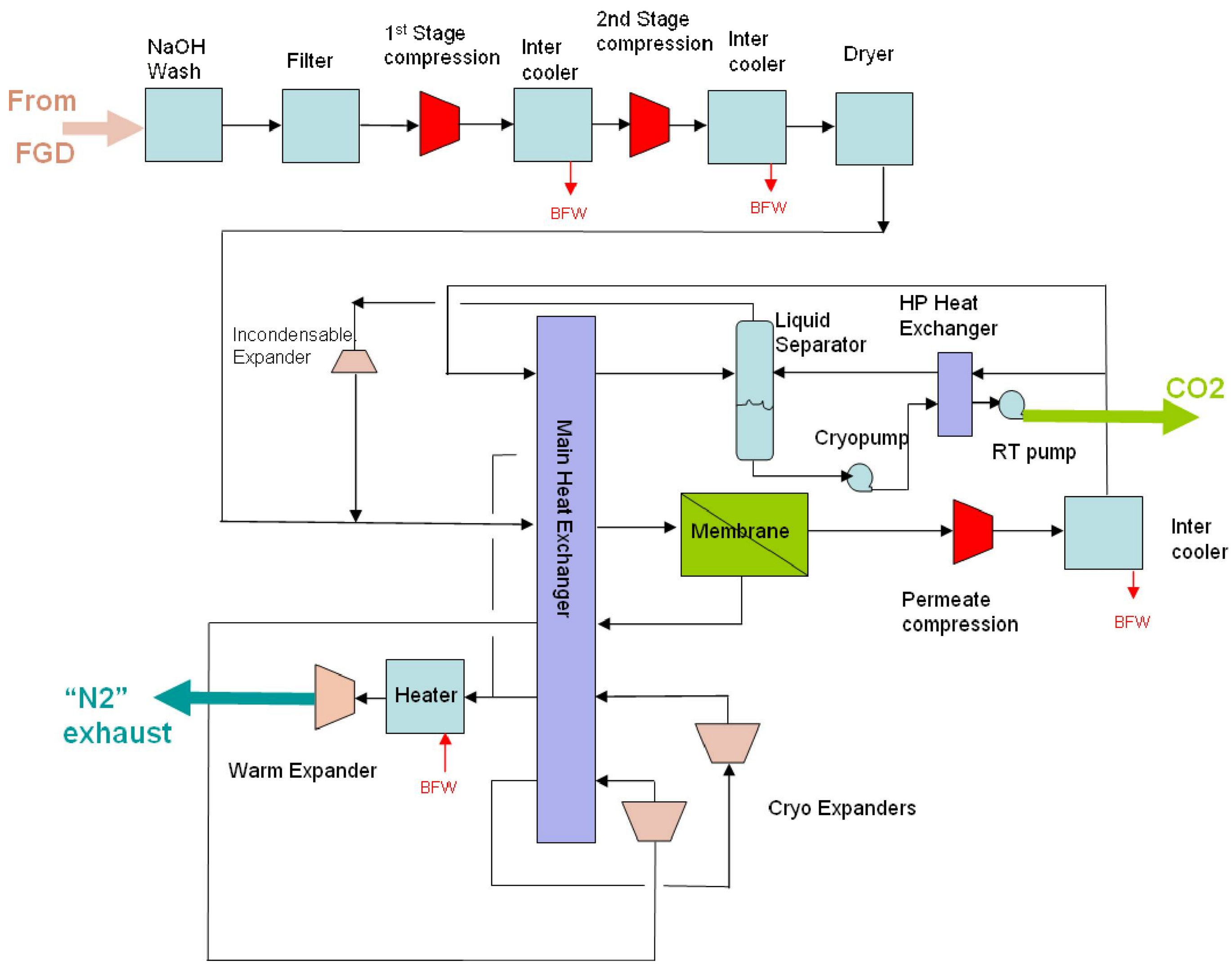


Figure 20: Block diagram showing equipment items in conceptual cold membrane process for CO2 capture from FGD treated power point flue gas

Since the proposed scheme uses high feed compression, it needs an efficient method of energy recovery. The main energy input and recovery steps corresponding to the scheme in Figure 16 are summarized in Table 3. The process viability depends crucially on the compression and expander efficiencies as well as the valorization of the boiler feed water. CO_2 capture energy was estimated through HYSYS simulation of the cold membrane process operating on a FGD and SCR pre-treated flue gas from air-fired coal power plant. The high efficiency of the compression and expander rotating machines simulation was validated by corresponding manufacturers. Boiler feed water valorization (power plant equivalent kwh for BFW at 147ºC) was estimated consistent with previous oxy-combustion studies (DOE/NETL 2007-1291).

Table 3. Main energy usage and recovery elements in process corresponding to Figure 19.

Main Energy input operations	Main energy recovery operations
Feed compression (1.0 to 16.0 bar)	Cold pressurized residue turbo-expansion
Permeate re-compression from 1-2 bar to 17 bar	Final warm residue turbo-expansion
Drier adsorbent regeneration	Boiler feed water (BFW) credit from compression
Liquid CO_2 pump to 150 bar	Recycle stream turbo-expansion

The process viability depends crucially on the compression and expander efficiencies as well as the valorization of the boiler feed water. The high efficiency of the compression and expander rotating machines used in the simulation were validated by the corresponding manufacturers. In order to obtain vendor quotes within the 3-month time-frame, we have had to simplify the expansion and turbo-expansion scheme at the cost of a small reduction in process energy efficiency. We also had to choose a less than optimum compression arrangement in order to get vendor quotes in the time available.

Boiler feed water valorization (power plant equivalent kwh for BFW at 147ºC) was estimated consistent with previous oxy-combustion studies (DOE/NETL 2007-1291) [13]. The summary energy demand and recovery for the main operations listed in Table 3, is shown in Figure 21.

Sensitivity analysis was done assuming variations in the compressor efficiency and BFW valorization. The specific energy for CO_2 capture by this process ranged from 216 – 242 kwh/T of CO_2 captured.

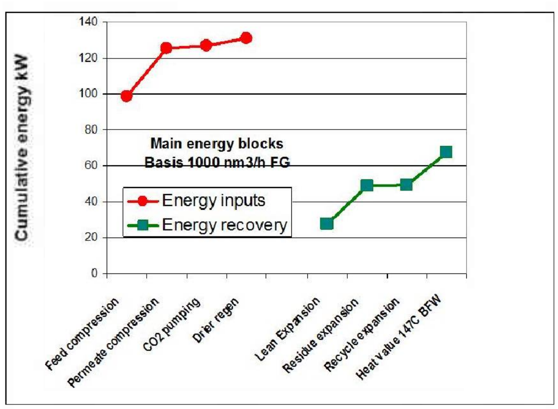


Figure 21. Simulation results showing cumulative energy demand (in red) and recovery (green) from the main energy intensive operations. Basis 1000 nm^3/h flue gas. The gap between final red and green lines is the parasitic energy for CO_2 capture.

4.5.2. Equipment capital cost estimation

Most equipment costs in the original cost estimate (at time of project proposal) were scaled from equipment quotes received previously for an oxy-combustion CO_2 capture project. This involved considerable scale up (5-8x) of equipment costs. For the current exercise, a more accurate estimate of plant capital cost was generated using ALE engineering design work and supplier quotes for more relevant reference equipment.

ALE obtained cost estimates for equipment on a scale directly suitable for CO_2 capture from the 550 MW (net) plant. ALE process equipment costing was based on (i) vendor quotes for the major equipment and (ii) the internal ALE database for other equipment. Equipment sizes / number of trains were based on the willingness of vendors to quote at that scale. As mentioned above, we have had to simplify the expansion and turbo-expansion scheme in order to get vendor quotes. For some equipment, ALE has suitable internal references either from previous CO_2 capture studies or from large-scale cryogenic air separation plants. The sources for costing important equipment are listed in Table 4.

For most equipment, the budgetary cost estimates are expected to be valid within ± 20%. When necessary, euro costs were converted to USD values by a factor of 1.25.

Table 4. Equipment cost source. The equipment scale is indicated by the # of trains needed for CO_2 capture from a 550 MW (net) PC plant.

Vendor quotes (trains)	AL Engineering database / (trains)
SOx scrubber (2)	Inter-coolers (3)
PM filters (1)	Driers (10)
Feed compressors (3)	Liquefier (3)
Permeate compressor (1)	Cold residue turbines (3)
CO_2 pumps (3)	Membrane (1)
Cryo heat exchangers (3)	Warm expander (2)

Figure 22 shows the relative capital costs of the main process equipment blocks. The most significant capital costs are due to the (i) feed compression and associated gas pretreatment and (ii) membrane system. For both items, there is a realistic chance of cost reductions in the immediate future (0-5 years) as well as long term reductions. The immediate cost reductions come from factors such as economy of scale for membrane manufacturing and increased equipment (compression, pre-treatment) optimization.

For the base case, the conservative membrane costing used in the original estimate is retained. However, in the long term and with increased operation scale, these costs could reduce further. Other cost reductions are possible in SO_2 scrubbing tower, compression train and turbo-expansion equipment.

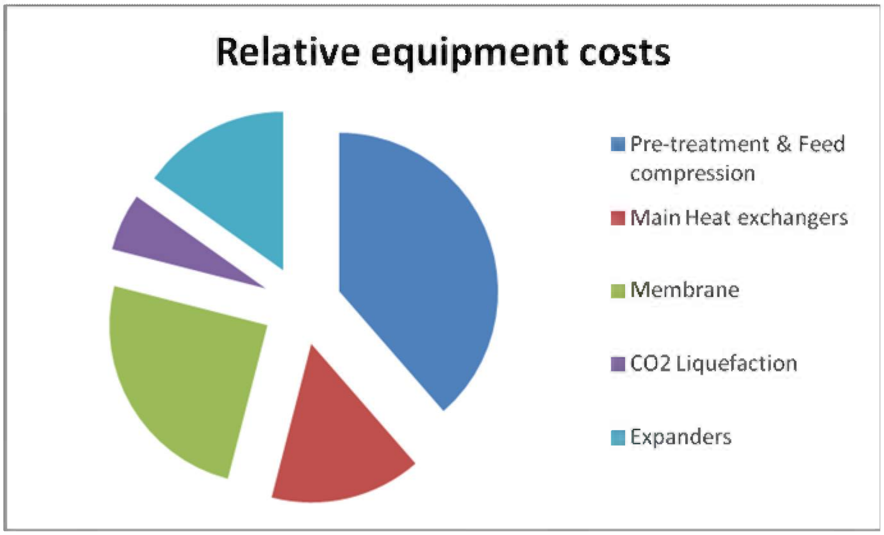


Figure 22. Relative capital cost (non-installed) for major equipment blocks of conceptual hybrid cryogenic + cold membrane process for 90% CO_2 recovery from a 550 MW (net) coal power plant.

4.5.3. LCOE estimation

The energy capture estimate was coupled with capital cost estimates to calculate the levelized cost of electricity (LCOE) for 90% CO_2 capture from an air-fired 550 MW net coal power plant delivering. The costing methodology followed DOE/NETL study 2010/1397 [14]. This analysis indicates increases in LCOE between 48% and 53%.

This equipment estimating approach based on 2012 quotes is a conservative method but is a useful bench-mark for the future. Compressor and expander costs were affected by willingness of vendors to supply quotes for the conceptual exercise and are not optimized for this application. For the base case, the conservative membrane costing used in the original calculation is retained. However, as mentioned above, in the long term and with increased operation scale, these costs could reduce further. Other cost reductions are possible in SO_2 scrubbing tower, compression train and turbo-expansion equipment.

The compression train and membrane system are the primary targets for further reductions in LCOE. In particular, membrane costs could be reduced by:

(i) Decreasing raw material costs and decreased overhead cost with increased manufacturing scale: lower the per membrane bundle cost (economy of scale).
(ii) Improving membrane bundle counter-current efficiency: decrease membrane area required by increasing separation efficiency.
(iii) Optimizing membrane bundle configuration to decrease membrane area required for a nominal increase in the energy penalty
(iv) Reducing skid costs by increasing membrane bundle size from present 12" diameter to a larger (30" - 36") diameter

Improvements in membrane bundle counter-current efficiency and optimizing the membrane configuration design are two essential technical advances for decreasing membrane costs and thereby decreasing LCOE costs.

4.6 PFD for potential field test (Task 4.2)

The costing exercise results were discussed with NETL / DOE on July 23 2012. As a result of this meeting, AL proceeded with definition of a PFD for a possible field test. As per the meeting recommendation, the PFD was targeted for a flue gas flow rate corresponding to 0.1 MW i.e. the same capacity as the current bench scale test. This would allow some of the existing equipment (membrane, cold heat exchanger, cold box) to be re-used for a field test.

In order to make this exercise as meaningful as possible, AL visited the post-combustion CO_2 capture facility (PC4) at the National Carbon Capture Center (NCCC), Wilsonville, AL. Our visit (E. Sanders, S Kulkarni, D Hasse) was hosted by F Morton and J Wheeldon (NCCC). The goal was to understand the test facility capabilities, gas treatment, utilities and space restrictions. Flue gas at PC4 is available after hot ESP and SCR followed by wet FGD. The flue gas can be further caustic scrubbed to reduce SO_2 to 2 ppm.

A schematic of the PFD is shown in Figure 22. Various scenarios were simulated based on the flue gas pre-treatment and by varying a range of feed flow rates. An example stream composition is given in Table 5.

The field test goal is to test the membrane with real flue gas. This PFD does not include a possible subsequent CO_2 liquefaction section. The membrane and liquefaction steps are well integrated in the cold membrane scheme (see Figure 4) resulting in a membrane feed of ~ 18% CO_2 , i.e higher than the ~13% in the dried flue gas.. The expected membrane operation conditions in the conceptual scheme are simulated in this PFD by recycling a fraction of the CO_2 enriched permeate. Depending on the range of feed flow rates expected to be tested, the CO_2 recovery in this configuration would vary between 40 – 90%.

One of the principal uncertainties in designing the field test is in specifying the compression and pre-treatment:
(i) The compression scheme shown in Figure 23 is designed to prevent condensation within the compression stages. Compressor start-up considerations may require small modifications. Discussions with two compressor manufacturers are underway to validate this approach. Electrical requirements at 0.1 MW scale can be handled by NCCC
(ii) Compressor pre-treatment requirements will be further validated with compressor manufacturers. The particulate count in the NCCC treated flue gas is "low". Particle size and distribution is assumed to be consistent with other wet FGD processes". It is possible that the compressor may not need extensive additional pre-filtration. Other issues are presence of SO_3 and heavy metals. The absence of Hg needs to be further confirmed.

PC4 facilities seem satisfactory for other field test requirements at the 0.1 MW scale (cooling water, space availability, analytical requirements etc).

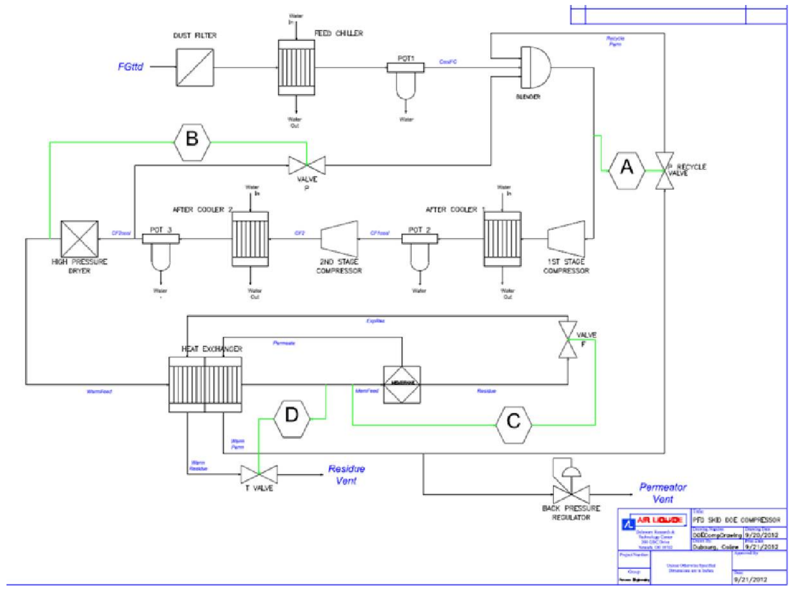


Figure 23. PFD of potential field test (0.1 MW scale) to demonstrate cold membrane operation with flue gas

Table 5. Indicative stream compositions

Name	Treated FG	Cool FG	Comp feed	Warm Feed	Mem Feed	Res	Permeate	ExpRes	Residue vent	Warm Perm	Permeate Vent	Recyle Perm
Temperature [C]	54	25	24	20	-45	-49	-45	-54	17	17	17	17
Pressure [bar]	1.1	1.1	1.1	16.0	15.9	15.5	1.2	2.5	2.4	1.1	1.1	1.1
Molar Flow [Nm3/h(gas)]	500	447	498	484	484	366	118	366	366	118	67	51
Mass Flow [kg/h]	641	598	688	676	676	470	207	470	470	207	117	90
Comp Mole Frac												
H_2O	13.4%	3.1%	2.8%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%	0.0%
CO_2	11.1%	12.4%	18.1%	18.6%	18.6%	2.9%	67.1%	2.9%	2.9%	67.1%	67.1%	67.2%
N_2	68.9%	77.1%	71.3%	73.3%	73.3%	90.2%	21.3%	90.2%	90.2%	21.3%	21.3%	21.2%
O_2	6.6%	7.4%	7.8%	8.1%	8.1%	6.9%	11.6%	6.9%	6.9%	11.6%	11.6%	11.6%
SO_2 ppm	25	28	28	9	9	1	32	1	1	32	32	32
NO_2 ppm	60	66	67	20	20	3	73	3	3	73	73	73

5. Conclusion

The projected tasks and associated milestones have been completed within the DOE budget and on schedule. The milestones and significant project findings are summarized below:

Milestone 1 - Complete closed loop sub-ambient temperature test system: The closed loop sub-ambient temperature test system was critical to the execution of the activities defined in the program. The target date for completion of this milestone was June 2011.

- This project milestone was completed with start-up of the skid (July 2011). Synthetic flue gas (CO_2/N_2) was used for all testing. The gas handling and cold box section of the skid are designed to be transportable for field testing.

- The skid uses an auto-refrigeration mechanism to achieve cold membrane temperatures (-10° to -45°C) through a combination of Joule-Thompson expansion cooling of the residue stream across a control valve and multi-stream heat-exchange for cooling of the incoming feed in an insulated Cold Box. The proposed conceptual process uses a similar cooling method (based on commercial Air Liquide cryogenic methodology using even more efficient turbo-expansion) to achieve cold feed temperatures. This feed cooling system has worked reliably.

Milestone 2 - Complete mini-permeator contaminant testing. Although no strong interactions between the primary flue gas contaminants (NO_x, SO_x) and the membrane are expected, this was verified in the laboratory at cold temperatures. The target date for completion of this milestone was September 2011.

- Measurements with CO_2/N_2 mixtures containing 100 ppm of either SO_2, NO_2 or NO mixtures were completed October 2011. Two long-term static exposure tests with SO_2.and NO_2 mixtures respectively were completed June 2012 with DOE permission.

Milestone 3 - Complete mechanical integrity testing of the sub-ambient membrane assembly: Tests with the 12" bundle show that the bundle maintains mechanical integrity through the operating range.

Significant findings during Phase I testing are described below:

- CTE (Coefficient of Thermal Expansion) analysis was performed to select the sealing components and clearances used in bundle testing. Integrity testing was performed with the largest currently available commercial MEDAL 12" bundle using a (clean) CO_2/N_2 synthetic flue gas mixture. The bundle was exposed to pressures as high as 15 bar and temperatures down to -40ºC routinely (with excursions down to -60ºC). 12" bundle separation performance was stable over a 3 month test period

- After mechanical integrity validation with the 12" bundle, performance testing was performed with a 6" bundle. To keep program costs and operational costs down, performance testing was designed for a 6" bundle. The 12" bundle requires 3.5 times the gas flow of a 6" bundle. Due to the higher permeate flow of the 12" bundle, it was not possible to access the entire parametric test space with available flows/compositions. The limited data indicates that 12” bundle performance was decreased by small leaks in a section of fiber and by deviation from ideal counter-current behavior

- Qualitatively, both the 6” and 12” membrane bundles exhibited the same temperature behavior as laboratory minipermeators. As operating temperature is decreased, CO_2 permeance decreases by only ~ 15% compared to the ambient temperature value while N_2 permeance decreases ~ 3x. Increased separation performance with minimal membrane productivity loss at cold temperature was observed with 6” bundles, 12” bundles as well as minipermeators. However, the back-calculated performance of fiber in the bundle has a lower selectivity (CO_2/N_2 ~ 60-80) than the extremely high selectivity (CO_2/N_2 > 90) measured for minipermeators. This short-fall appears to be due to various small leak paths in commercial bundles which are significant only for these high selectivity membrane fibers and/or non-ideal flow patterns that prevent the bundle from achieving ideal countercurrent flow performance. To compensate these effects, bundles were operated at -45°C instead of the -30°C to -40°C originally proposed operating range based on mini-permeator data.

- Membrane longevity was confirmed by a total 8-month long exposure of the 6” bundle to sub ambient temperature operating conditions. The majority of the testing was run at 200 psi, -45°C and 18% CO_2 feed. These pressure and temperature conditions were determined to be the optimum by the first parametric study. Under these test conditions, bundles membrane performance was stable. There was no decline in either permeance or selectivity. The measured membrane lifetime is better than expectations based on ambient temperature operations but is an expected consequence of low temperature membrane operation. The longer lifetime of module will reduce annual membrane replacements.for commercial facilities. This improved membrane lifetime provides further justification for pursuing cold membrane technology development in general and field testing in particular.

- Parametric tests were performed at the 1 month and 6 month mark of the long term test with the 6” bundle. Experimental results indicate that membrane performance is best at the coldest temperature and highest feed pressure achievable. In this testing, the minimum temperature and highest feed pressure were limited respectively by the membrane vessel rating and by the compressor capability. In terms of the process choice, the optimized variables will also depend on the energy and captial costs of achieving these desirable pressure/temperature conditions.

- The membrane performance validated in Phase 1 testing is 10% lower CO_2/N_2 selectivity and 15% higher CO_2 permeance than the proposal submission estimate based on mini-permeator laboratory testing. A rough estimate of the impact of this small performance change made by updating the original DRTC-created HYSYS™ process simulation and budget estimate shows that the membrane area required would be 30% less and the net effect on the LCOE increase is beneficial (~ 30% increase in LCOE compared to 35% in original simulation).

Milestone 4 - Complete design and budget evaluation of slipstream test system, and simulation and economic evaluation of commercial facility: Updated engineering and cost analyses were presented to NETL July 2012. Based on positive feedback, AL proceeded with the development of a process flow diagram (PFD) for a field test. PFD definition for potential field test was completed through (i) simulation work at DRTC, (ii) discussions with compressor manufacturers and (iii) a field visit to the NCCC, Wilsonville, AL. The PC4 facility at the NCCC is a suitable site for a 0.1 MW scale test. This milestone was completed in September 2012.

Significant findings during Milestone 4 (Phase 2) work are described below:

- After completion of experimental work (Phase 1), engineering design and LCOE evaluation (Phase 2) commenced with assistance from Air Liquide Engineering-Champigny, France. The design is based on FGD and SCR treated flue gas. The proposed process concept uses relatively high flue gas compression to 15 bar; this energy is used to provide the process cooling and later expanded for partial energy recovery. Heat of compression is used partially as credit in form of boiler feed water. Flue gas heat is assumed to reheat the CO_2-depleted vent gas before final expansion for energy recovery.

- In the revised process simulation, membrane performance is based on the 6" membrane bundle test data . The original DRTC-created process simulation was modified to decrease the process complexity and incorporate a more rigorous simulation of:
    - Decreased process complexity of expansion turbine layout
    - Separated the main cryo heat exchanger into two separate units.
    - Improved estimate of pre-treatment pressure drops.
    - Increased approach temperature in heat exchangers
    - Increased regeneration temperatures in drier.
    - Improved estimates of feed booster efficiencies based on vendor supplied axial and centrifugal compressor performance.

  The net effect of these changes increases the expected specific energy of CO_2 capture by 20% over the proposal submission estimate. 7% of this specific energy increase is due to the decrease in selectivity of the membrane bundle compared to mini-permeator data. The remaining 13% is attributable to the process changes detailed above. Improvement in compressor efficiency by the use of existing axial-radial machines or by using less conservative estimates for the energy value of pre-heated boiler feed water would reduce the specific energy penalty. A sensitivity analysis shows the specific energy for CO_2 capture to lie within the range of 216-242 kwh/T CO_2 captured.

- Cost calculations are based on 90% CO_2 capture from an air-fired coal power plant delivering net 550 MW, using DOE/NETL study 2010/1397. Process equipment cost estimates were based on (i) vendor quotes as available and (ii) internal ALE database estimates for other equipment. For most equipment, the budgetary cost estimates are valid within ± 20%. Equipment sizes/number of trains are based on the willingness of vendors to quote at scale available today. This analysis indicates increases in LCOE between 48% and 53%. The most significant capital cost items are (i) feed compression and associated gas pretreatment and (ii) membrane system. For both items, there is a realistic chance of cost reductions in the immediate future as well as in the long term. Improvements in membrane bundle counter-current efficiency and configuration are feasible technical advances for decreasing membrane costs and further decreasing LCOE costs.

6. Technology Transfer / Project publications

- S. S. Kulkarni, D. J. Hasse, P. Shanbhag, E. Sanders, J-P. Tranier, M. Bennett, "CO_2 capture by sub-ambient membrane operation", NETL CO_2 capture technology meeting, Pittsburgh, Sept 2010

- S. S. Kulkarni, D. J. Hasse, E. Sanders, J-P. Tranier, P. Shanbhag, "CO_2 capture by sub-ambient membrane operation", North American Membrane Society, June 2011

- E. Sanders, D. Hasse, E. Corson, D. Kratzer, S. Kulkarni, "CO_2 capture by sub-ambient membrane operation" NETL CO_2 capture technology meeting, Pittsburgh, August 2011

- D. J. Hasse, S. S. Kulkarni, E. Corson, E. Sanders, J-P. Tranier, "CO_2 capture by sub-ambient membrane operation", North American Membrane Society, New Orleans, June 2012

- D. Hasse, S. Kulkarni, E. Sanders, E. Corson, J-P. Tranier, "CO_2 capture by sub-ambient membrane operation" International conference on Green House Gas Technologies, Kyoto, November 2012. (to be published in Energy Procedia)

- S. Kulkarni, D. Hasse, E. Sanders, E. Corson, J-P. Tranier, "CO_2 capture by sub-ambient membrane operation" NETL CO_2 capture technology meeting, Pittsburgh, July 2012
    - The detailed results of the LCOE re-evaluation (Task 4.1) were presented to NETL on July 23, 2012 at Pittsburgh.

- A project summary and potential field testing were discussed during an AL DRTC visit to the National Carbon Capture Center (F Morton and J Wheeldon), September 26, 2012.

- Air Liquide is sponsoring a Ph.D. candidate with Professor Koros' group at Georgia Institute of Technology. This work will investigate the fundamental basis of the unusual cold temperature CO_2 permeability-selectivity.

7. Table of Figures

8. References

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9. List of acronyms

BFW	Boiler feed water
CPU	Cryogenic purification unit
E_P	Activation energy for permeation
FGD	Flue gas desulphurization
GPU	Gas permeance units $cm^3(STP)/cm^2\text{-s-cm(Hg)} \times 10^{-6}$
HX	Heat exchanger
LCOE	Levelized cost of electricity
PFD	Process flow diagram
SCR	Selective catalytic reduction

10. Acknowledgments

We thank our NETL Project Manager Andy O'Palko for his patience and guidance throughout the course of this work. We would also like to thank the many helpful comments from Jose Figueroa, L. Brickett, J. Ciferno and S Vora.

Appendix I

6” Bundle performance data

See Figure 5 for sensor location in Skid. Sensor legend is given below.

TT-21	Warm Permeate Temperature
TT-22	Warm Residue Temperature
TT-23	Dry Feed Gas Temperature
TT-24	Residue from Membrane Temperature
TT-25	Permeate from Membrane Temperature
TT-26	Expanded Residue Temperature
TT-27	Cold Feed Gas Temperature
TT-28	Cold Box Temperature

FM-31	Residue Flow
FM-32	Feed Flow
FM-33	Permeate Flow

PT-1	Compressed Feed Inlet Pressure
PT-2	Residue from Membrane Pressure
PT-3	Expanded Residue Pressure
PT-4	Permeate from Membrane Pressure
PT-5	Cold Feed Gas Pressure
PT-6	Suction Return Pressure

Cumulative Hours on Stream	Feed	Permeate	Retentate	Feed IR (xf)	Permeate IR (xp)	Retentate IR (xr)	TT22	TT23	TT24	TT25	TT26	TT27	TT28	PT1	PT2	PT4	PT5	PT6
	flow	flow	flow	% CO_2	% CO_2	% CO_2	°C	°C	°C	°C	°C	°C	°C	psig	psig	psig	psig	psig
0.0	384.6	65.1	319.5	17.9%	68.3%	10.2%	19	18	16	20	15	17	28	203	176	15.5	201	11
0.2	509.6	63.7	445.9	17.6%	71.1%	11.6%	19	18	15	19	15	17	28	201	160	12.6	196	7
0.8	171.8	51.9	119.9	18.3%	63.4%	4.9%	19	18	13	18	13	14	25	201	189	11.2	200	7
1.1	243.0	55.2	187.8	17.8%	66.4%	7.4%	19	18	13	17	12	14	25	199	179	13.3	199	7
1.4	316.2	61.9	254.3	17.7%	70.4%	8.2%	18	19	12	16	11	13	25	205	183	15.0	203	11
6.4	405.8	40.1	365.7	17.0%	80.5%	12.1%	15	18	-23	-19	-24	-22	-23	201	177	11.0	198	9
22.7	405.8	42.0	363.8	17.7%	77.2%	11.9%	14	17	-24	-22	-25	-22	-20	201	177	10.0	198	8
23.9	440.7	42.0	398.7	16.9%	77.1%	12.2%	14	17	-24	-22	-24	-22	-22	200	174	10.6	197	8
24.5	494.1	43.5	450.6	17.5%	77.6%	12.4%	14	17	-24	-22	-25	-22	-20	200	176	11.0	197	9
25.2	340.6	41.0	299.6	17.5%	76.7%	10.8%	14	17	-24	-22	-26	-23	-22	202	183	10.0	200	8
26.7	289.5	38.0	251.5	17.5%	76.6%	10.2%	15	18	-26	-24	-28	-25	-25	198	177	10.0	196	8
27.2	438.5	40.5	398.0	17.2%	76.9%	12.0%	14	18	-26	-24	-30	-23	-20	202	176	10.5	199	14
47.1	429.4	31.4	398.0	14.9%	74.8%	10.8%	14	18	-42	-38	-46	-40	-24	202	179	10.1	198	8
49.2	441.2	40.6	400.6	17.4%	79.8%	12.0%	14	18	-42	-38	-46	-40	-22	202	179	11.0	198	8
94.0	444.8	44.2	400.6	17.4%	78.9%	11.7%	14	17	-42	-39	-46	-40	-20	202	179	11.1	199	8
94.5	501.2	51.0	450.2	18.9%	81.4%	12.9%	13	17	-42	-39	-45	-40	-22	199	170	12.0	195	9
94.9	551.2	51.5	499.7	18.8%	81.6%	13.3%	13	17	-42	-38	-46	-40	-21	205	180	11.0	201	8
96.0	399.7	49.1	350.6	18.8%	80.8%	11.7%	14	17	-42	-39	-46	-40	-21	202	182	11.5	199	8
96.5	347.8	48.3	299.5	18.8%	80.2%	10.9%	14	18	-43	-39	-46	-40	-23	202	185	11.0	199	8
97.1	295.2	45.7	249.5	18.9%	79.3%	10.1%	14	18	-43	-39	-46	-40	-23	201	187	11.1	200	8
97.5	242.1	42.6	199.5	19.0%	78.1%	9.1%	15	18	-43	-39	-47	-40	-20	202	191	11.0	201	8
99.0	188.3	38.8	149.5	19.1%	76.3%	7.8%	15	19	-43	-39	-47	-40	-21	202	193	11.0	201	8
99.3	154.3	34.0	120.3	19.2%	74.7%	6.9%	15	19	-43	-39	-46	-40	-24	199	187	10.0	197	7
123.7	403.5	54.0	349.5	17.8%	80.2%	10.9%	15	18	-44	-38	-50	-44	-22	203	187	10.1	202	8
125.7	408.2	58.1	350.1	18.1%	80.7%	10.7%	15	19	-42	-38	-45	-40	-25	203	186	10.0	202	8
142.6	408.5	59.0	349.5	17.8%	79.3%	10.5%	14	18	-42	-39	-46	-40	-21	203	186	10.1	201	8
143.5	377.5	57.5	320.0	17.8%	79.1%	10.1%	15	18	-43	-39	-46	-40	-20	203	188	10.0	202	8
147.0	374.5	56.0	318.5	17.7%	78.4%	9.9%	15	19	-42	-38	-45	-40	-24	203	189	10.0	202	8

148.2	375.1	56.1	319.0	17.2%	76.9%	9.6%	15	19	-42	-38	-45	-40	-20	204	189	10.0	203	8
166.7	371.2	51.7	319.5	15.7%	75.3%	9.0%	16	18	-42	-38	-46	-40	-21	203	188	8.7	202	8
173.3	374.2	56.2	318.0	18.0%	80.2%	10.2%	16	20	-43	-39	-46	-40	-20	201	186	10.0	200	8
191.3	377.5	57.1	320.4	18.0%	79.1%	10.1%	16	18	-43	-39	-46	-40	-25	202	187	9.7	200	8
222.2	377.5	57.0	320.5	17.7%	78.5%	9.9%	16	20	-42	-38	-45	-40	-23	201	186	9.0	200	7
244.0	376.1	55.6	320.5	17.8%	78.7%	9.9%	16	20	-42	-38	-45	-40	-20	202	187	9.0	201	7
262.1	377.0	55.7	321.3	17.9%	79.0%	10.1%	15	19	-43	-39	-46	-40	-25	201	186	9.4	200	7
263.7	441.7	67.0	374.7	17.8%	79.6%	9.8%	15	18	-43	-39	-46	-40	-25	225	209	10.0	224	7
286.2	426.1	51.6	374.5	16.5%	78.3%	10.3%	15	19	-47	-43	-52	-45	-24	204	187	9.4	202	7
310.6	357.2	57.1	300.1	18.4%	79.8%	10.0%	16	20	-48	-44	-52	-45	-22	202	188	10.1	200	8
363.8	414.8	53.3	361.5	17.2%	78.5%	10.5%	15	19	-47	-43	-52	-45	-25	202	188	10.5	200	8
392.3	357.1	57.6	299.5	18.4%	79.4%	9.7%	15	19	-47	-43	-51	-45	-25	202	188	10.4	201	8
430.7	357.7	58.2	299.5	18.2%	79.2%	9.6%	14	18	-48	-44	-52	-45	-24	202	188	10.2	200	8
459.1	395.9	57.4	338.5	17.5%	79.7%	10.0%	15	18	-47	-43	-51	-45	-24	202	186	10.1	201	8
478.7	396.4	57.1	339.3	17.5%	78.8%	10.0%	14	18	-47	-43	-52	-45	-23	202	187	10.2	200	8
483.1	397.3	56.8	340.5	17.5%	78.9%	9.8%	15	19	-47	-43	-51	-45	-25	203	188	10.0	200	8
503.2	396.4	57.1	339.3	17.6%	78.9%	9.9%	14	18	-47	-43	-52	-45	-22	202	187	10.0	201	8
507.5	397.8	58.3	339.5	18.1%	79.5%	10.0%	15	19	-48	-44	-52	-45	-24	202	187	10.6	200	8
510.0	378.9	59.4	319.5	18.2%	80.3%	9.8%	15	19	-47	-44	-51	-45	-22	203	187	10.3	200	8
526.2	378.0	57.6	320.4	17.5%	78.3%	9.4%	14	18	-48	-44	-52	-45	-22	202	187	10.0	200	8
532.2	385.5	65.5	320.0	19.0%	80.4%	9.8%	15	19	-48	-44	-52	-45	-20	202	187	10.3	200	9
552.8	383.6	64.1	319.5	18.7%	79.8%	9.5%	15	19	-47	-44	-51	-45	-25	202	187	10.4	200	8
598.2	382.7	63.2	319.5	17.8%	78.9%	9.2%	15	18	-48	-44	-45	-45	-26	202	187	10.7	200	8
601.7	440.5	65.0	375.5	17.9%	79.5%	9.9%	15	19	-48	-44	-52	-45	-20	202	183	10.8	199	8
622.2	440.7	67.0	373.7	17.8%	79.3%	9.8%	15	18	-48	-44	-45	-45	-22	202	184	10.5	199	8
647.1	439.3	67.0	372.3	17.7%	79.0%	9.6%	14	18	-48	-44	-52	-45	-21	202	183	10.2	199	8
676.2	438.4	64.7	373.7	17.4%	78.6%	9.7%	15	19	-48	-44	-52	-45	21	202	183	10.9	199	8
694.7	440.3	64.2	376.1	17.4%	79.0%	9.5%	15	19	-47	-44	-51	-45	-25	203	184	10.7	200	8
702.2	446.4	69.4	377.0	18.2%	80.0%	9.6%	15	19	-47	-44	-51	-45	-24	203	184	11.0	200	8
769.2	571.0	71.3	499.7	17.3%	80.3%	10.8%	14	18	-47	-44	-52	-45	-22	207	185	10.7	199	7
768.2	518.3	68.6	449.7	17.4%	79.4%	10.3%	14	18	-47	-43	-51	-45	-25	203	179	10.7	199	8
766.2	442.1	67.6	374.5	17.6%	79.3%	9.6%	15	18	-48	-44	-52	-45	-23	203	185	10.5	200	8
771.2	378.9	63.2	315.7	17.5%	78.4%	8.6%	15	19	-47	-44	-51	-45	-22	202	187	10.5	200	8

773.3	307.2	57.1	250.1	17.3%	76.4%	7.4%	16	20	-48	-44	-52	-45	-22	203	191	10.3	201	8
815.7	495.5	45.5	450.0	17.5%	78.4%	12.8%	14	18	-47	-45	-51	-45	-38	162	134	10.6	158	9
814.7	549.0	49.5	499.5	17.8%	80.0%	13.0%	14	18	-47	-44	-50	-45	-38	163	130	9.3	157	7.5
817.0	420.2	45.7	374.5	17.7%	78.0%	12.1%	14	19	-48	-45	-52	-47	-39	162	139	10.5	159	9
818.2	363.3	48.1	315.2	18.1%	79.2%	10.9%	15	19	-48	-46	-51	-47	-39	162	144	8.8	160	9
819.1	296.8	47.2	249.6	18.1%	77.0%	9.7%	15	19	-47	-45	-49	-45	-37	162	148	9.2	161	7
820.1	215.2	41.1	174.1	18.2%	73.8%	8.3%	15	20	-47	-45	-51	-45	-36	162	152	10.0	161	9
844.0	527.6	29.8	497.8	12.7%	64.3%	10.2%	14	18	-43	-42	-46	-42	-36	159	124	9.8	154	8
843.0	477.5	29.2	448.3	12.6%	64.3%	9.9%	14	19	-44	-43	-48	-44	-35	159	129	9.7	154	8
842.0	402.5	28.8	373.7	12.5%	64.0%	9.4%	15	19	-45	-44	-49	-44	-34	159	135	9.4	156	8
840.8	342.1	27.4	314.7	12.5%	63.0%	9.0%	15	19	-46	-44	-50	-45	-36	159	140	10.0	157	8
839.8	277.0	27.8	249.2	12.6%	62.8%	8.2%	15	20	-46	-44	-50	-45	-35	158	143	9.7	157	8
838.7	201.5	26.9	174.6	12.7%	61.2%	6.9%	15	20	-47	-45	-51	-45	-37	159	150	9.5	159	8
868.5	212.8	38.2	174.6	13.5%	64.6%	5.1%	15	20	-47	-43	-52	-45	-24	200	192	9.2	199	8
869.7	289.3	40.1	249.2	12.8%	66.8%	6.3%	15	19	-47	-43	-52	-45	-21	200	188	9.0	199	7
867.3	357.7	43.4	314.3	13.3%	70.0%	7.4%	15	19	-47	-43	-52	-47	-23	201	185	9.2	198	7
864.1	418.6	44.4	374.2	13.1%	69.9%	8.0%	15	18	-47	-43	-52	-45	-24	200	181	9.3	197	8
865.2	495.0	45.8	449.2	13.0%	70.6%	8.6%	14	18	-46	-43	-51	-44	-20	200	176	9.5	196	8
866.2	546.0	46.3	499.7	13.0%	71.4%	8.9%	14	18	-46	-43	-51	-44	-24	200	173	9.3	196	8
936.0	213.3	38.7	174.6	12.4%	58.7%	4.4%	15	19	-37	-33	-41	-35	-15	200	191	9.5	199	8
935.0	290.7	41.5	249.2	12.2%	61.7%	5.8%	15	18	-37	-33	-40	-35	-11	200	187	9.2	199	7
934.0	342.6	43.9	298.7	12.1%	63.0%	6.4%	18	18	-36	-33	-40	-35	-14	200	185	8.9	199	7
937.4	418.3	45.0	373.3	12.0%	64.2%	7.1%	15	19	-35	-32	-39	-35	-15	200	180	9.5	198	8
938.7	492.2	43.9	448.3	11.8%	65.9%	7.7%	15	18	-37	-33	-41	-35	-14	200	175	9.1	196	8
939.8	542.2	43.9	498.3	11.7%	66.6%	8.0%	14	18	-37	-33	-41	-37	-12	200	171	9.3	196	8
983.7	199.1	24.5	174.6	12.1%	57.4%	7.3%	14	19	-37	-35	-39	-35	-34	152	141	9.9	151	8
982.6	274.6	24.5	250.1	12.0%	58.9%	8.3%	15	18	-35	-34	-39	-35	-32	150	133	9.3	148	8
986.7	337.9	23.6	314.3	11.8%	59.4%	8.9%	15	19	-37	-35	-39	-35	-33	152	131	10.1	150	9
988.0	398.3	25.0	373.3	11.8%	60.1%	9.3%	15	19	-36	-35	-38	-34	-33	153	126	9.9	149	8
989.3	472.3	22.6	449.7	11.7%	59.6%	9.7%	15	19	-35	-34	-38	-34	-30	152	116	10.3	149	9
989.8	521.0	22.2	498.8	11.7%	60.0%	9.8%	15	19	-35	-34	-38	-34	-32	154	115	9.8	149	8
1009.2	545.5	46.0	499.5	17.7%	78.5%	13.3%	14	18	-37	-35	-40	-35	-31	154	116	8.3	148	7
1008.2	496.9	46.6	450.3	17.9%	77.6%	13.0%	14	18	-35	-35	-37	-34	-29	154	121	9.1	150	7

1007.3	421.4	46.9	374.5	18.0%	77.6%	12.2%	15	19	-37	-35	-40	-35	-34	154	128	8.7	150	7
1011.9	359.5	44.8	314.7	17.8%	76.4%	11.2%	15	19	-36	-35	-37	-34	-32	152	130	8.3	150	7
1012.7	290.7	41.1	249.6	17.7%	74.4%	10.5%	15	20	-36	-35	-39	-35	-33	154	141	9.2	150	7
1013.4	213.3	38.2	175.1	17.8%	72.4%	8.6%	16	20	-37	-35	-40	-35	-34	152	141	8.8	150	7
1035.2	521.0	70.4	450.6	17.6%	79.1%	10.4%	14	17	-37	-34	-40	-35	-14	205	180	10.4	201	7
1036.0	440.6	65.2	375.4	17.7%	77.5%	10.1%	14	17	-37	-34	-40	-35	-15	202	182	11.0	199	8
1037.1	379.0	63.8	315.2	17.9%	77.1%	9.4%	14	17	-38	-34	-41	-35	-19	202	186	10.2	200	7
1037.7	311.4	61.8	249.6	18.0%	75.9%	7.6%	14	17	-38	-34	-40	-35	-14	202	189	9.8	201	7
1038.5	228.4	53.8	174.6	18.2%	72.7%	5.9%	14	18	-37	-34	-39	-35	-16	202	188	9.7	200	8
1108.5	574.7	75.0	499.7	17.7%	78.0%	10.8%	14	17	-28	-25	-29	-25	-17	201	170	10.8	197	7
1105.2	526.1	78.3	447.8	17.9%	76.9%	10.2%	14	17	-28	-25	-30	-27	-19	202	175	10.2	198	7
1103.1	449.7	76.0	373.7	18.1%	75.0%	9.4%	14	17	-27	-25	-28	-25	-14	201	179	10.9	198	7
1104.0	387.4	73.1	314.3	18.2%	74.2%	8.6%	14	17	-28	-25	-29	-25	-19	201	183	10.8	199	8
1109.3	317.6	67.0	250.6	18.4%	74.1%	7.4%	15	18	-29	-26	-30	-26	-16	201	188	10.4	199	7
1110.4	233.1	58.0	175.1	18.6%	70.7%	5.7%	15	18	-28	-26	-28	-26	-18	157	137	10.8	200	8
1132.2	492.2	43.0	449.2	17.3%	72.7%	13.0%	15	18	-27	-24	-29	-26	-18	153	117	11.4	148	10
1127.3	421.9	47.4	374.5	17.1%	72.6%	11.5%	15	18	-27	-25	-29	-25	-14	155	127	9.6	152	7
1128.5	362.9	49.1	313.8	18.0%	73.4%	11.3%	15	18	-27	-25	-29	-25	-14	153	130	9.8	150	8
1129.3	296.8	47.2	249.6	18.2%	72.6%	10.2%	15	18	-27	-25	-29	-25	-13	155	139	9.7	151	7
1130.9	218.0	44.8	173.2	18.5%	70.6%	8.2%	16	19	-27	-25	-29	-25	-13	154	142	9.4	152	8
1152.2	404.9	30.7	374.2	12.1%	59.5%	8.8%	15	18	-26	-24	-28	-25	-17	155	127	8.5	152	7
1153.4	344.9	29.7	315.2	12.0%	58.9%	8.4%	15	18	-26	-24	-29	-25	-15	152	128	8.5	150	8
1155.0	278.4	31.9	249.6	12.0%	57.7%	7.6%	16	19	-26	-24	-29	-25	-15	153	135	8.6	151	7
1156.6	202.0	26.9	175.1	12.0%	55.6%	6.5%	16	19	-27	-24	-29	-25	-16	152	140	9.3	151	8
1174.6	495.9	45.3	450.6	11.9%	62.1%	7.8%	14	17	-27	-25	-29	-25	-14	204	177	10.4	200	9
1176.2	421.4	46.7	374.7	12.1%	62.0%	7.1%	15	18	-26	-24	-28	-25	-16	204	183	10.2	202	8
1177.2	358.6	43.4	315.2	12.1%	61.3%	6.6%	15	18	-27	-25	-29	-26	-19	203	189	10.1	200	8
1179.2	293.5	43.9	249.6	12.1%	59.5%	5.5%	16	19	-26	-24	-28	-25	-18	203	189	10.0	201	8
1180.5	213.8	39.2	174.6	12.1%	56.3%	4.2%	16	19	-27	-25	-29	-25	-17	201	192	9.8	201	8
1199.0	420.9	46.2	374.7	14.5%	74.2%	8.8%	14	18	-47	-43	-53	-45	-14	203	185	9.1	201	7
1200.8	442.1	68.4	373.7	18.0%	81.8%	9.8%	14	17	-47	-42	-52	-44	-6	203	184	8.0	200	5
1270.7	438.4	63.7	374.7	17.5%	80.1%	9.6%	13	17	-48	-43	-53	-45	-6	203	185	9.1	200	6
1293.6	425.7	50.6	375.1	14.9%	74.5%	8.7%	14	17	-46	-41	-51	-44	-8	203	185	9.1	201	7

1366.0	421.9	47.2	374.7	16.1%	76.7%	10.1%	16	19	-38	-33	-43	-37	-6	204	184	9.4	201	7
1438.6	445.5	69.4	376.1	19.1%	82.0%	10.4%	15	19	-48	-44	-52	-45	-25	204	186	11.1	201	8
1463.2	446.9	74.1	372.8	18.9%	81.0%	10.1%	16	19	-48	-43	-52	-45	-24	205	186	10.7	202	8
1469.4	447.3	73.6	373.7	18.9%	81.3%	10.0%	17	21	-47	-43	-51	-45	-24	204	186	10.6	202	8
1486.7	444.0	70.7	373.3	18.8%	81.3%	10.2%	17	21	-47	-43	-52	-45	-20	202	183	10.5	199	8
1517.2	444.5	69.4	375.1	18.6%	81.5%	10.1%	15	19	-48	-44	-52	-45	-22	202	183	10.3	199	8
1535.9	441.7	67.0	374.7	17.9%	80.2%	9.8%	15	18	-48	-44	-52	-45	-25	202	184	9.9	199	7
1607.2	443.6	71.8	371.8	18.4%	80.5%	10.0%	15	19	-48	-44	-52	-45	-20	202	183	10.2	199	7
1631.6	443.1	68.4	374.7	18.3%	80.7%	10.0%	16	20	-48	-44	-52	-45	-24	202	183	10.2	199	7
1655.0	444.5	70.0	374.5	18.5%	81.1%	9.9%	15	19	-48	-44	-52	-45	-24	203	184	10.5	200	7
1798.4	428.5	49.1	379.4	15.1%	76.1%	9.0%	16	20	-47	-44	-52	-45	-28	202	183	8.7	199	7
1822.2	440.3	60.4	379.9	17.1%	79.5%	9.8%	15	19	-47	-44	-52	-45	-29	202	184	9.9	200	7
1846.9	445.9	65.6	380.3	18.1%	80.7%	10.2%	16	20	-48	-44	-52	-45	-24	202	183	10.2	199	8
1870.3	446.4	66.5	379.9	18.1%	80.9%	10.0%	15	19	-48	-44	-52	-45	-24	202	183	9.9	199	7
1943.5	449.7	69.4	380.3	18.3%	81.0%	9.9%	15	19	-48	-44	-52	-45	-24	203	184	9.9	200	7
1990.5	447.8	67.0	380.8	18.1%	80.3%	10.0%	16	20	-47	-44	-51	-45	-25	202	183	10.4	199	8
2038.8	448.3	67.5	380.8	18.1%	80.6%	9.9%	15	19	-48	-44	-52	-45	-25	202	183	10.2	199	7
2110.5	450.6	71.2	379.4	18.5%	81.2%	9.9%	15	19	-48	-44	-52	-45	-29	202	183	9.8	200	7
2158.6	449.7	68.9	380.8	18.3%	80.9%	9.9%	15	19	-48	-44	-51	-45	-27	202	183	10.0	199	7
2206.2	449.2	67.9	381.3	18.4%	80.7%	10.0%	16	19	-48	-44	-52	-45	-27	202	183	10.4	199	7
2279.3	453.0	73.6	379.4	18.8%	81.5%	10.0%	15	19	-48	-44	-52	-45	-25	203	183	10.3	199	7
2326.6	453.0	73.6	379.4	18.7%	81.5%	10.0%	16	20	-48	-44	-51	-45	-25	202	183	10.5	199	7
2374.9	450.6	70.0	380.6	18.3%	80.7%	9.9%	16	19	-48	-44	-52	-45	-29	202	183	10.5	200	7
2471.6	452.1	71.3	380.8	18.3%	80.9%	10.0%	15	19	-48	-44	-52	-45	-27	202	183	10.5	199	7
2518.7	451.1	73.1	378.0	18.4%	81.0%	9.9%	15	18	-48	-44	-52	-45	-25	202	183	10.3	199	7
2639.3	455.4	75.5	379.9	19.0%	81.9%	10.1%	15	19	-48	-44	-51	-45	-29	201	182	10.4	198	7
2677.7	453.9	74.5	379.4	18.8%	81.5%	9.9%	15	19	-48	-44	-51	-45	-27	202	183	10.4	199	7
2782.6	456.8	77.4	379.4	18.7%	81.6%	9.8%	15	19	-48	-45	-52	-45	-26	202	183	10.0	199	6
2830.2	456.8	78.4	378.4	18.5%	81.6%	9.5%	15	19	-48	-44	-52	-45	-24	203	183	9.3	199	5
2878.5	450.2	72.7	377.5	17.7%	80.2%	9.4%	15	19	-48	-44	-51	-45	-29	203	184	9.8	200	6
2854.7	180.3	50.5	129.8	18.6%	71.0%	4.1%	17	22	-48	-44	-52	-45	-28	202	196	8.4	201	6
2975.5	165.2	39.2	126.0	16.7%	69.9%	4.7%	16	21	-47	-43	-52	-44	-29	200	194	7.6	200	6
2998.7	168.0	42.0	126.0	17.8%	71.7%	4.7%	17	22	-49	-45	-52	-45	-35	200	195	7.7	200	6

3047.3	169.4	43.9	125.5	18.5%	72.9%	4.8%	16	21	-49	-45	-52	-45	-36	200	194	7.4	200	5
3119.3	445.0	68.9	376.1	18.1%	81.4%	10.0%	15	19	-48	-45	-51	-45	-35	199	180	8.7	196	5
3167.7	447.3	68.4	378.9	18.0%	81.2%	9.7%	15	19	-48	-45	-51	-45	-35	203	184	9.4	200	6
3215.3	451.1	74.5	376.6	18.6%	82.0%	9.9%	15	19	-48	-45	-51	-45	-30	202	183	9.7	199	6
3287.7	448.3	70.3	378.0	18.1%	81.1%	9.9%	15	19	-48	-45	-51	-45	-33	201	182	9.3	198	6
3311.4	450.6	72.6	378.0	18.2%	81.3%	9.5%	15	19	-48	-45	-51	-45	-34	202	183	9.1	199	5
3358.9	177.9	47.7	130.2	18.1%	71.3%	4.3%	18	23	-49	-45	-52	-45	-30	202	196	7.8	201	5
3503.0	176.0	45.8	130.2	18.0%	70.9%	4.4%	16	22	-49	-45	-52	-45	-33	201	196	7.9	201	6
3527.5	175.5	45.7	129.8	18.0%	71.1%	4.3%	16	22	-49	-45	-52	-45	-34	201	195	7.6	200	5
3623.8	177.4	47.2	130.2	18.3%	71.8%	4.6%	16	21	-49	-45	-52	-45	-30	201	195	8.4	201	6
3670.9	443.1	64.2	378.9	16.9%	79.2%	9.3%	15	20	-48	-45	-52	-45	-34	202	183	9.0	199	6
3890.4	175.5	46.2	129.3	17.7%	70.4%	4.4%	18	23	-49	-45	-53	-45	-35	201	195	8.3	200	6
4032.8	175.5	45.7	129.8	17.7%	70.6%	4.3%	18	23	49	-45	-52	-45	-27	202	196	8.1	201	6
4056.6	446.9	66.1	380.8	17.3%	80.0%	9.4%	15	19	-48	-45	-51	-45	-28	202	183	8.7	199	5
4127.7	449.2	67.9	381.3	17.8%	80.3%	9.6%	16	19	-47	-45	-51	-45	-25	202	183	9.6	199	6
4156.6	448.8	69.4	379.4	17.8%	80.3%	9.7%	16	19	-48	-45	-51	-45	-29	201	182	9.4	198	6
4396.2	437.9	59.9	378.0	16.0%	77.8%	9.0%	15	19	-48	-45	-51	-45	-36	202	183	9.0	199	6
4463.7	450.2	71.8	378.4	17.9%	80.3%	9.6%	16	20	-48	-45	-51	-45	-30	202	183	9.3	199	5
4465.4	180.7	50.5	130.2	18.7%	70.8%	4.2%	18	23	-49	-45	-52	-45	-30	202	196	8.2	201	6
4469.3	177.9	48.1	129.8	18.1%	67.7%	4.2%	16	21	-49	-45	-52	-45	-34	202	196	8.1	201	6
4486.7	425.6	45.7	379.9	16.3%	75.5%	10.9%	16	20	-45	-42	-49	-44	-34	160	137	8.1	157	6
4488.3	167.0	37.2	129.8	17.0%	67.2%	6.0%	17	22	-46	-43	-50	-44	-30	160	153	7.3	159	5
4491.8	166.1	36.8	129.3	16.9%	67.3%	6.0%	18	23	-45	-42	-49	-44	-35	160	153	7.7	159	6
4511.2	413.8	26.4	387.4	10.4%	57.6%	7.7%	16	20	-39	-37	-42	-39	-34	160	135	6.7	157	5
4512.9	155.2	25.0	130.2	10.9%	51.9%	4.4%	16	22	-40	-38	-44	-39	-32	160	152	6.9	159	6
4516.2	154.8	24.6	130.2	10.8%	52.9%	4.5%	17	23	-41	-38	-44	-39	-32	160	152	6.7	159	5
4535.3	428.0	44.4	383.6	12.2%	68.8%	7.5%	16	20	-47	-44	-51	-45	-34	201	182	7.7	199	6
4536.7	165.6	34.9	130.7	12.7%	57.3%	3.5%	16	21	-47	-44	-51	-45	-34	201	195	7.1	201	6
4540.1	163.7	33.5	130.2	12.7%	57.7%	3.6%	16	22	-48	-44	-52	-45	-33	200	194	7.1	200	6
4631.2	456.8	73.6	383.2	18.2%	78.6%	10.0%	15	19	-38	-35	-40	-35	-20	198	177	9.8	195	5
4636.2	181.2	51.9	129.3	18.6%	66.1%	4.1%	15	19	-39	-35	-41	-35	-21	200	193	8.2	199	5
4638.0	311.9	63.2	248.7	17.7%	71.8%	7.3%	15	19	-37	-34	-38	-35	-20	199	186	9.4	198	6
4654.6	443.1	56.6	386.5	18.4%	78.9%	11.8%	16	20	-37	-34	-40	-35	-21	162	137	8.8	159	6

4657.2	300.1	50.5	249.6	18.4%	74.1%	9.8%	16	20	-37	-34	-40	-35	-23	161	145	8.5	159	6
4659.6	170.8	41.0	129.8	18.1%	68.0%	6.2%	17	21	-37	-34	-40	-35	-22	161	153	8.1	160	6
4679.6	418.0	31.5	386.5	12.0%	63.6%	8.6%	15	19	-35	-33	-38	-35	-23	161	134	6.9	157	5
4682.2	279.4	29.8	249.6	12.0%	60.9%	7.2%	17	20	-36	-34	-40	-35	-24	161	145	7.5	159	6
4684.6	158.6	28.8	129.8	12.0%	55.5%	4.6%	17	22	-35	-33	-38	-35	-20	161	153	7.1	160	6
4702.7	430.8	45.6	385.2	11.9%	65.1%	7.2%	17	20	-36	-34	-39	-35	-23	200	179	8.4	197	6
4705.1	293.0	42.4	250.6	12.3%	61.4%	5.7%	16	20	-37	-34	-40	-35	-20	202	189	8.3	200	6
4708.5	165.6	35.8	129.8	12.3%	53.9%	3.1%	17	21	-37	-34	-41	-35	-20	201	195	7.8	201	6
4799.1	464.8	79.7	385.1	18.2%	74.9%	9.5%	16	19	-27	-24	-29	-25	-12	200	177	10.7	197	6
4802.2	321.8	71.9	251.0	18.4%	68.9%	7.0%	16	19	-28	-25	-29	-25	-13	201	187	9.9	200	5
4805.0	185.0	55.2	129.8	18.3%	64.3%	3.5%	17	21	-29	-25	-30	-25	-11	201	194	9.3	200	6
4822.2	446.4	60.9	385.5	18.4%	76.1%	11.6%	17	20	-27	-25	-29	-25	-12	161	133	8.9	157	6
4824.8	303.9	55.2	248.7	18.3%	71.1%	9.3%	17	20	-27	-24	-29	-25	-14	161	144	9.0	159	6
4827.6	175.5	45.3	130.2	18.3%	66.6%	5.5%	18	22	-28	-25	-30	-25	-15	160	151	8.0	159	6
4845.8	420.0	34.5	385.5	11.9%	60.4%	8.5%	17	20	-27	-24	-29	-25	-11	159	131	7.7	156	6
4848.8	287.8	37.7	250.1	12.2%	57.1%	6.8%	17	20	-26	-24	-29	-25	-15	160	143	7.2	158	5
4853.3	163.7	33.5	130.2	12.4%	53.4%	4.0%	18	22	-25	-23	-28	-25	-13	160	151	7.7	460	6
4870.7	432.7	49.5	383.2	11.8%	61.8%	6.9%	16	19	-27	-24	-29	-25	-17	200	178	8.4	197	6
4872.9	297.3	47.7	249.6	12.1%	56.8%	5.3%	16	19	-27	-24	-29	-25	-11	201	187	8.3	199	6
4875.8	171.3	41.1	130.2	12.2%	50.0%	2.5%	17	20	-27	-25	-30	-25	-14	202	195	7.6	501	6
4894.2	420.0	35.7	384.3	12.2%	61.4%	8.6%	17	19	-26	-24	-29	-25	-10	160	132	7.4	157	6
4897.2	163.3	33.5	129.8	12.4%	50.2%	4.3%	16	19	-26	-24	-29	-25	-12	161	152	7.2	160	5
4899.8	285.5	35.9	249.6	12.3%	60.2%	6.8%	16	20	-26	-24	-29	-25	-12	161	144	7.2	159	5
4967.3	453.9	70.3	383.6	18.4%	81.2%	10.2%	16	20	-48	-45	-51	-45	-45	200	181	9.7	197	6
4968.2	556.8	62.7	494.1	18.0%	80.5%	12.1%	16	20	-47	-44	-50	-45	-37	188	139	10.3	183	7
4968.8	518.6	70.8	447.8	18.1%	80.8%	11.1%						-45		201	178	10.6	198	7
4971.8	378.0	63.3	314.7	18.0%	79.3%	9.0%						-45				10.1	199	
4972.4	309.1	59.5	249.6	18.2%	78.0%	8.0%						-45				9.4	199	
4972.9	228.4	54.3	174.1	18.6%	74.4%	6.2%						-45				9.0	200	
4973.4	178.4	48.2	130.2	18.9%	71.1%	4.8%						-45				8.3	200	
4974.0	448.3	64.1	384.2	17.7%	77.9%	10.2%						-45				11.1	198	
4993.1	450.2	64.7	385.5	18.0%	80.2%	10.3%	15	19	-48	-45	-51	-45	-33	201	182	10.4	198	6
4994.5	309.6	59.0	250.6	18.2%	77.1%	7.9%	16	20	-48	-45	-51	-45	-34	201	189	9.7	199	6

4996.2	177.0	46.3	130.7	18.2%	70.3%	4.4%	16	21	-49	-45	-52	-45	-37	202	196	8.4	201	6
4997.7	310.0	60.8	249.2	17.9%	76.3%	7.9%	16	20	-48	-45	-51	-45	-36	201	189	9.6	199	6
5014.4	436.0	50.9	385.1	18.3%	80.6%	12.2%	15	20	-47	-45	-51	-45	-37	162	139	8.6	159	6
5016.8	296.3	48.1	248.2	18.3%	75.4%	10.2%	15	20	-47	-44	-50	-45	-36	161	146	8.4	159	6
5018.9	169.4	39.2	130.2	18.4%	69.6%	6.6%	15	20	-47	-44	-51	-45	-36	161	154	8.2	161	6
5021.4	295.4	46.2	249.2	18.2%	73.9%	10.2%	14	18	-48	-45	-51	-45	-36	161	147	8.5	159	6
5038.7	415.3	31.7	383.6	12.5%	66.2%	9.1%	16	20	-43	-41	-46	-42	-36	160	136	7.3	157	6
5042.0	280.3	29.7	250.6	12.2%	61.6%	7.6%	15	20	-44	-42	-48	-44	-29	162	147	7.2	160	6
5044.6	157.1	26.9	130.2	12.3%	56.4%	5.1%	15	20	-44	-43	-48	-44	-37	161	154	7.0	161	6
5062.2	426.1	41.7	384.4	12.2%	69.5%	7.5%	16	21	-47	-45	-51	-45	-37	201	182	7.7	198	5
5064.4	287.4	38.3	249.1	12.4%	63.4%	6.2%						-45				8.0	199	
5053.7	162.8	33.0	129.8	12.9%	60.5%	3.8%						-45				7.2	199	
5189.2	455.8	71.2	384.6	18.8%	81.7%	10.6%	14	18	-48	-45	-51	-45	-36	199	179	10.4	196	6
5233.5	454.9	70.3	384.6	18.4%	81.4%	10.3%	15	19	-48	-45	-51	-45	-30	200	180	10.0	197	5
5302.6	454.4	69.3	385.1	18.4%	81.4%	10.3%	15	19	-48	-45	-51	-45	-36	199	179	10.2	196	5
5355.8	453.5	67.5	386.0	17.8%	80.4%	10.0%	15	19	-48	-45	-51	-45	-37	202	182	10.1	199	6
5475.2	449.2	66.0	383.2	17.7%	80.0%	10.1%	14	18	-47	-45	-50	-45	-36	199	180	10.2	196	6
5497.3	420.4	36.8	383.6	9.9%	61.2%	6.3%	16	20	-46	-44	-50	-45	-34	200	181	7.2	198	5
5498.4	419.0	34.9	384.1	9.9%	60.8%	6.3%	16	20	-46	-44	-50	-45	-33	199	180	7.8	196	6
5499.4	419.5	34.9	384.6	9.9%	61.2%	6.3%	16	21	-47	-44	-51	-45	-35	199	180	7.2	196	5
5500.6	418.1	34.5	383.6	9.8%	61.1%	6.4%	16	20	-47	-44	-51	-45	-36	201	181	8.1	198	6
5501.8	417.1	33.5	383.6	9.8%	61.5%	6.3%	16	21	-47	-44	-51	-45	-36	202	183	7.7	199	6
5520.2	415.7	29.2	386.5	9.2%	58.0%	6.3%	15	20	-47	-44	-51	-45	-33	199	180	7.4	196	6
5544.8	451.6	67.0	384.6	17.7%	80.2%	10.0%						-45		199	180	10.0	196	6
5638.3	518.6	85.4	433.2	18.4%	68.7%	10.8%	18	19	-3	-1	-3	-2	13	181	146	12.5	177	5