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Carbon Dioxide Capture by a Continuous, Regenerative Ammonia-Based Scrubbing Process

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OVERVIEW

To develop a knowledge/data base to determine whether an ammonia-based scrubbing process is a viable regenerable-capture technique that can simultaneously remove carbon dioxide, sulfur dioxide, nitric oxides, and trace pollutants from flue gas.

NETL CO_2 Sequestration Program Goal (by 2012)

- **Post-combustion flue gases**
  - < 20% increase in cost of energy services
    - Parasitic losses of capture
    - CO_2 compression, transportation, and injection
  - 90% CO_2 capture
- **Current monoethanolamine (MEA) scrubbing systems – estimated 67% increase in COE**

PROJECT OBJECTIVES

- To develop a low cost CO_2 capture process for flue gas.
- To integrate the control of pollutants (SO_2, NO_x, Hg, and fine particulate) with CO_2 control.
- To assure that the process is regenerable.
- To off-set costs with saleable by-products.

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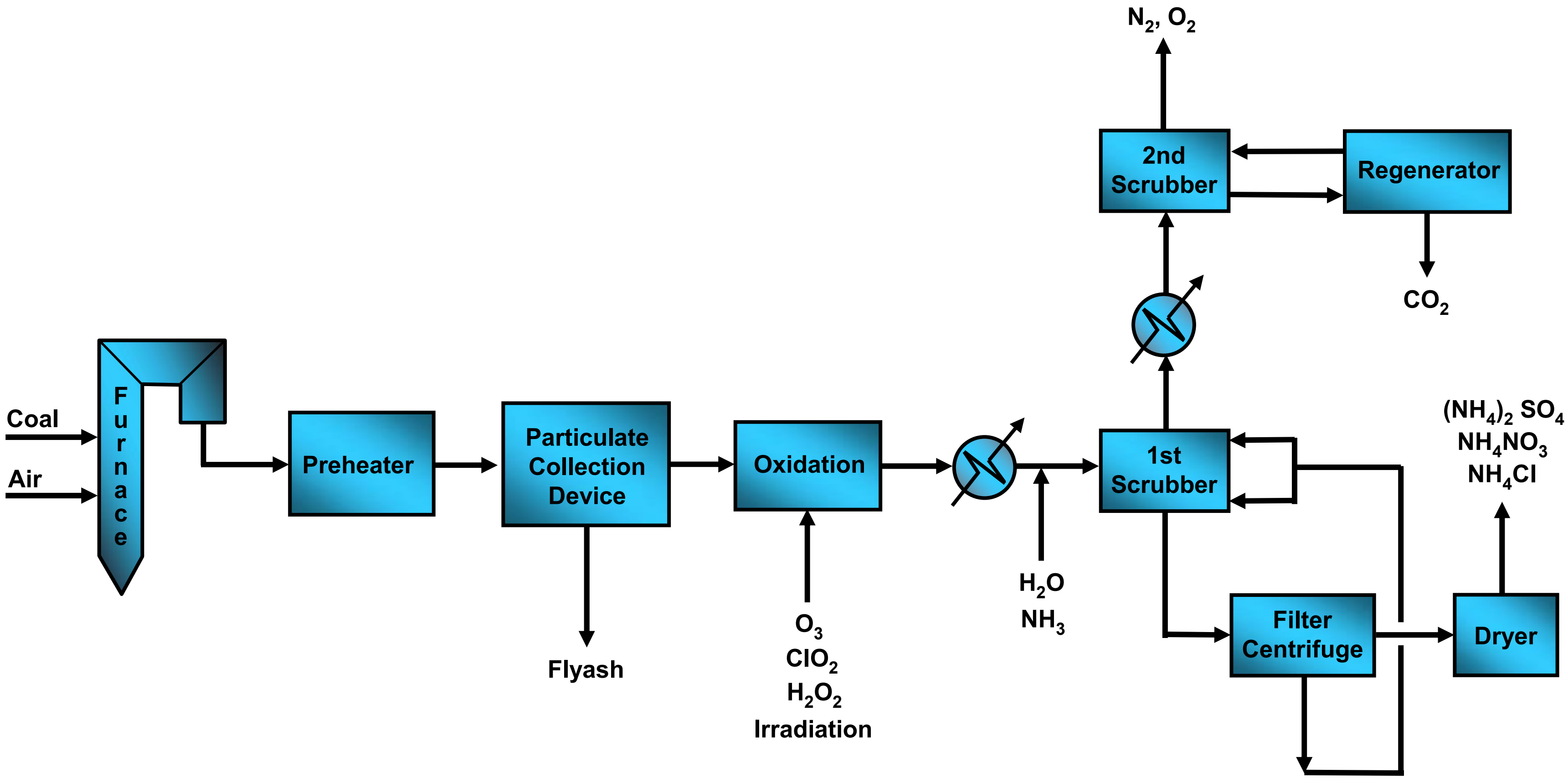
Aqua Ammonia Process



ADVANTAGES OF PROCESS

- Multi-component control of acid gases produced during coal combustion.
- Combination of oxidation step with ammonia wet scrubbing.
- Process is regenerable with respect to CO_2 scrubbing.
- Fabrication of a saleable commodity (fertilizer) out of waste materials (acid gases).
- Production of a pure CO_2 stream that can further be processed or sequestered.
- Lower energy cost.

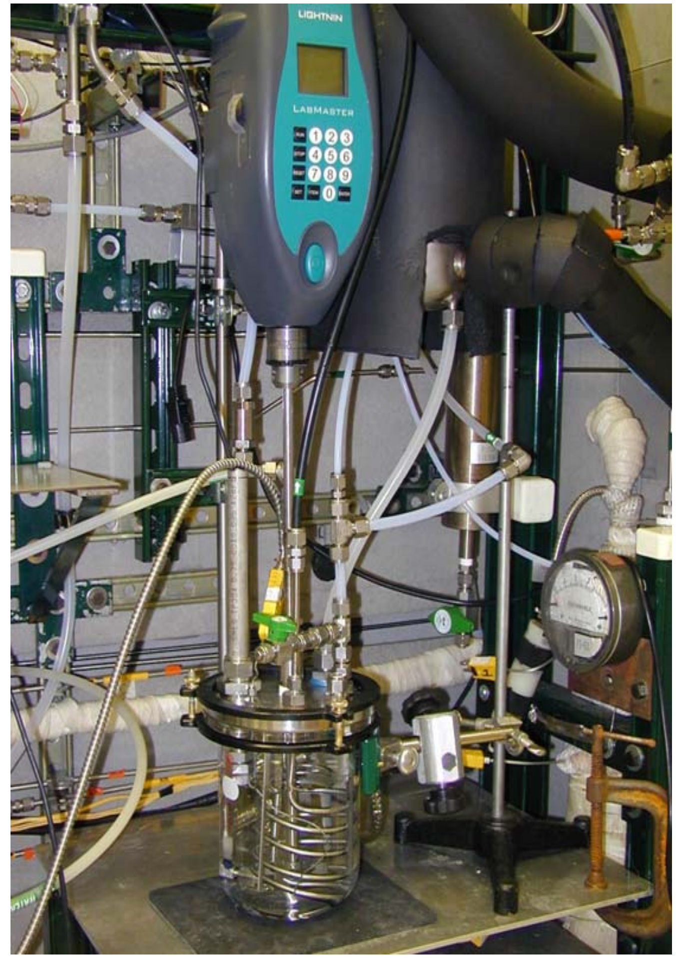
AQUA AMMONIA PROCESS CHEMISTRY (thermal regeneration)

- $2\ NH_4HCO_3(aq) \longrightarrow (NH_4)_2CO_3(aq) + CO_2(g) + H_2O$ $\Delta H = 6.4$ kcal/mol
- $NH_4HCO_3(aq) \longrightarrow NH_3(aq) + CO_2(g) + H_2O$ $\Delta H = 15.3$ kcal/mol
- $(NH_4)_2CO_3(aq) \longrightarrow 2\ NH_3(aq) + CO_2 + H_2O$ $\Delta H = 24.1$ kcal/mol

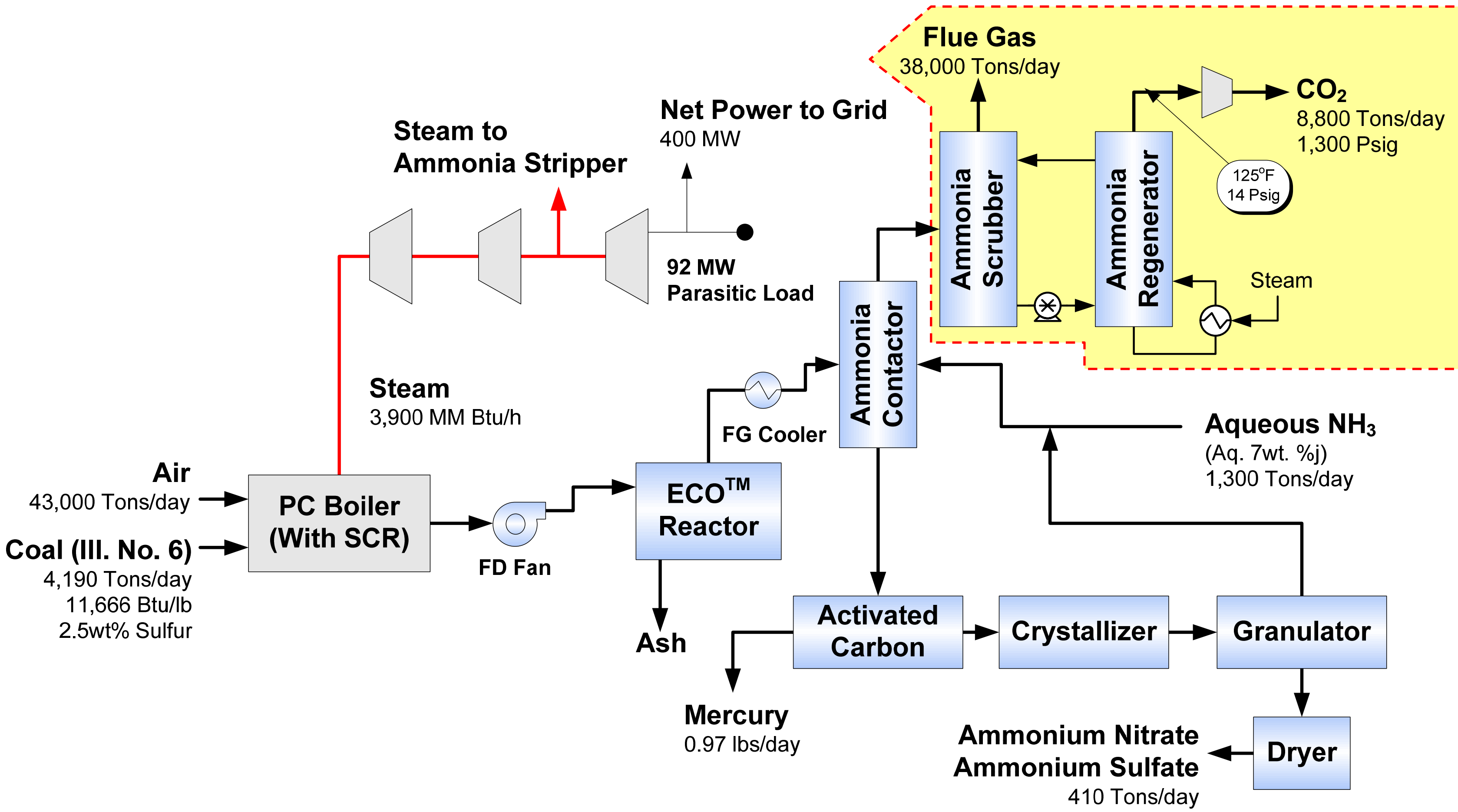
Values indicate enthalpy of dissociation of CO_2

SEMI-BATCH REACTOR RESULTS

- **Saturated solution with CO_2 in flue gas**
- **Regenerated to 180°F**
- **Five absorption/regeneration cycles**
- **Capacities to 0.068 g CO_2/g absorbent measured (vs 0.052 for 30% MEA)**
- **Above 170°F regeneration, conversion to NH_4OH observed**
- **Ammonia losses ~ 1/3 of initial inventory**



Aqueous Ammonia Systems Analysis



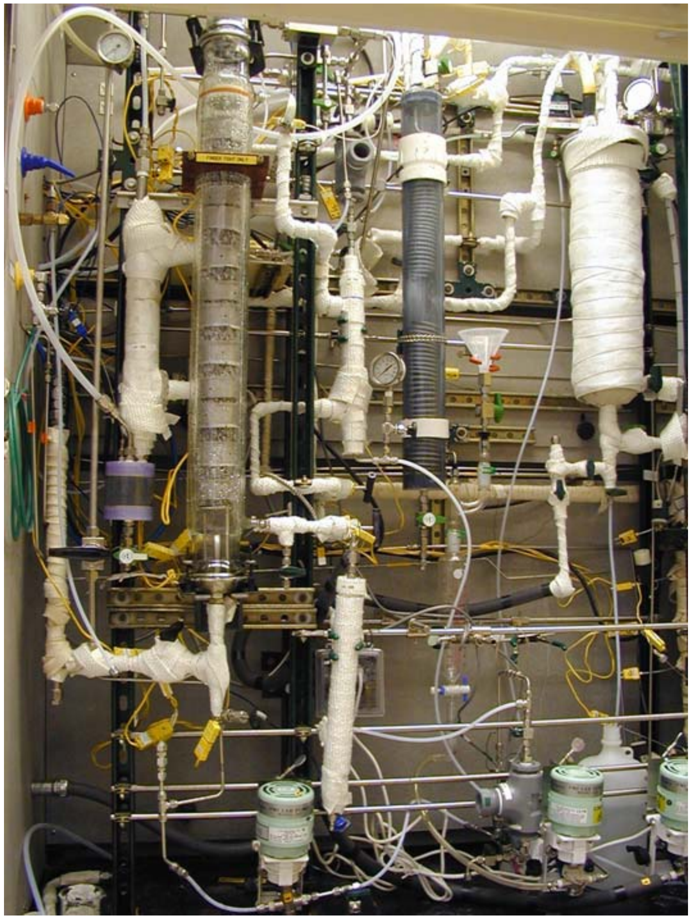
Integrated with Powerspan's multi-pollutant control ECO™ technology
Approaches programmatic goal of 20% increase in cost of energy service

Aqueous Ammonia Systems Analysis Results

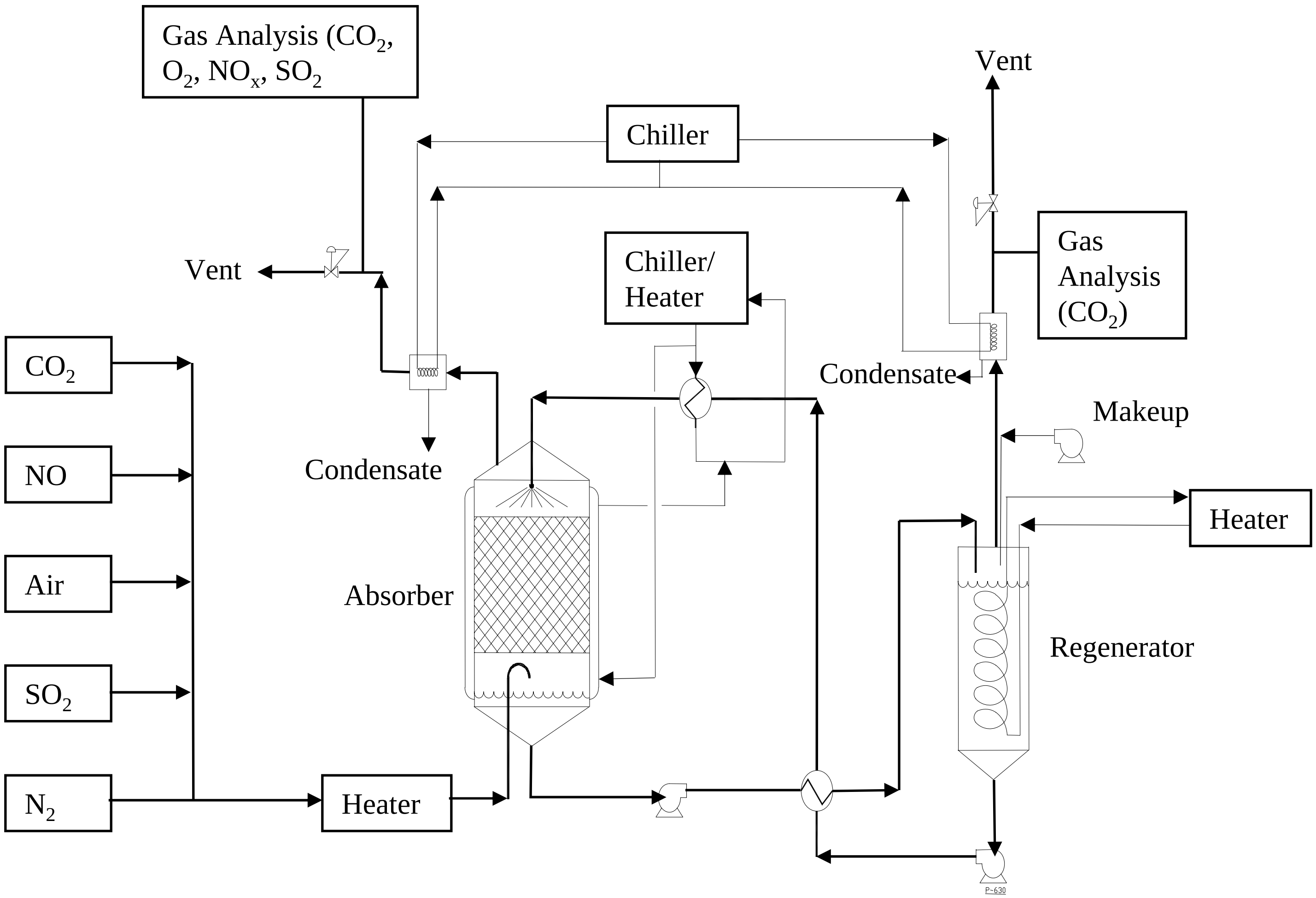
	Base Plant	**Amine**	**Ammonia**	**Ammonia**
Component Controlled	--	CO_2	CO_2	CO_2, SO_2, NO_x, Hg
$/tonne CO_2 avoided	--	47	27	14
Increase in COE, %	--	67	39	21

Continuous Reactor Testing

- **Results of semi-batch reactor system used as basis for continuous flow unit design coupling absorption and regeneration**
- **Perform parametric study**
  - Gas and liquid residence times in absorber and regenerator
  - Absorber and regenerator temperatures
  - Initial ammonia concentration
  - CO_2 concentration
  - Available mass transfer area
- **Identify species present**
- **No attempt to maximize CO_2 removal**



Continuous Process Flow Diagram



Aqua Ammonia Continuous Unit
Baseline Test Conditions

Absorber: Temperature: 80°F

Gas Flow: 23,500 sccm (8 s residence time)

Packing Height: 68 cm (4 sets BX Gauze packing)

Absorber Diameter: 7.6 cm

Solution: 3000 g of 14% NH_3 solution at startup

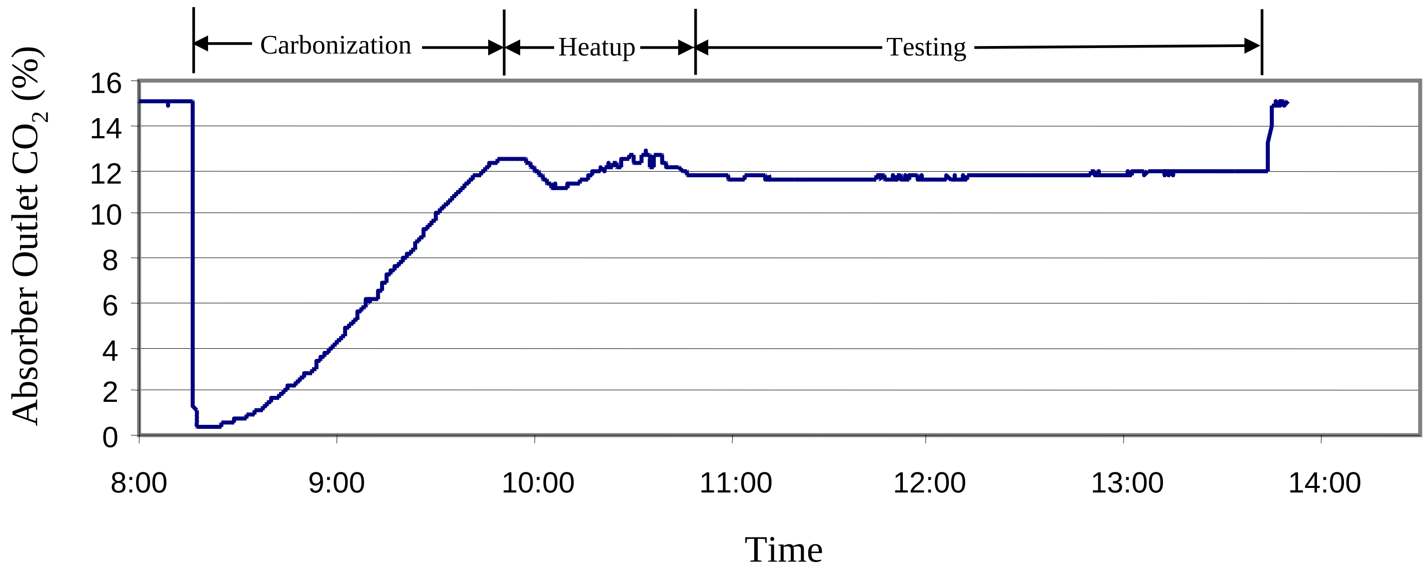
172 g/min (res. time = 17 minutes, L/G=55 gal/kcf)

Saturate with CO_2 in simulated flue gas at 50°F before heat-up

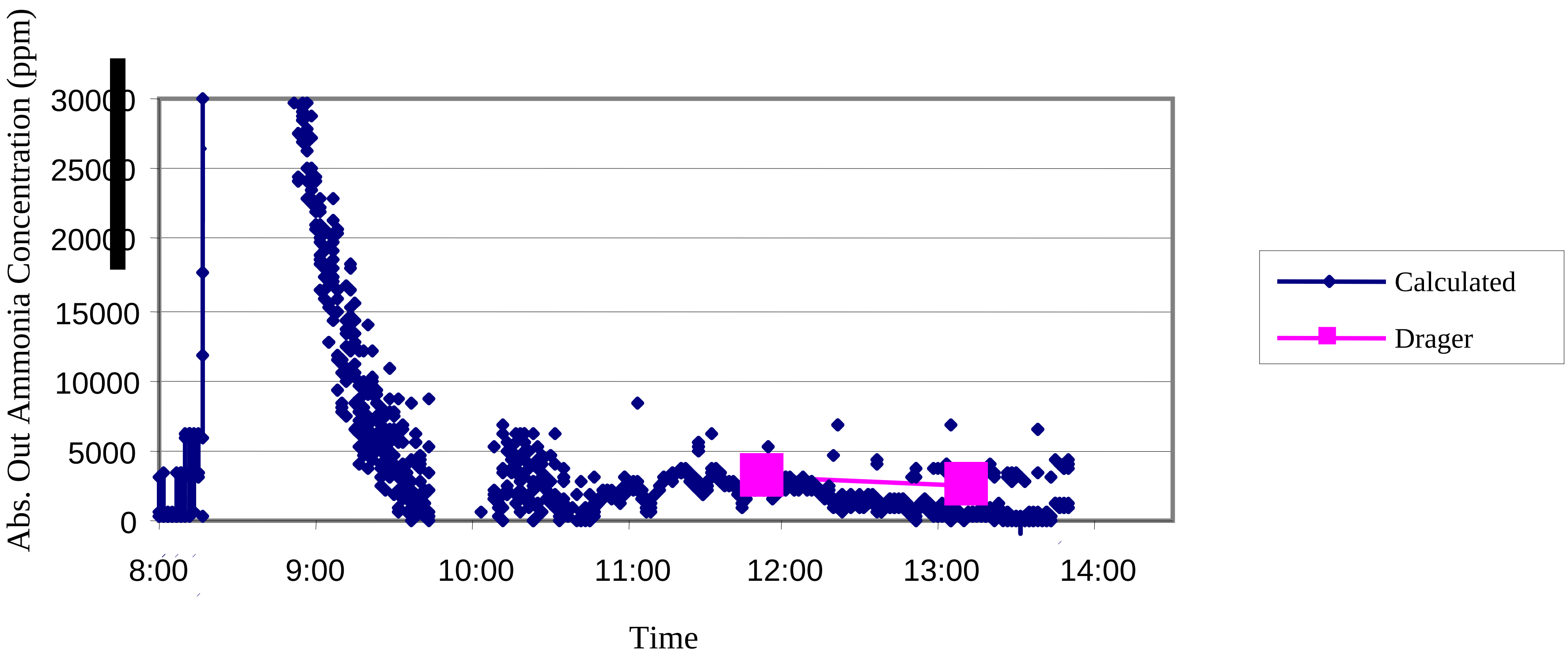
Regenerator: Temperature: 170°F

Post absorber and regenerator samples extracted and analyzed

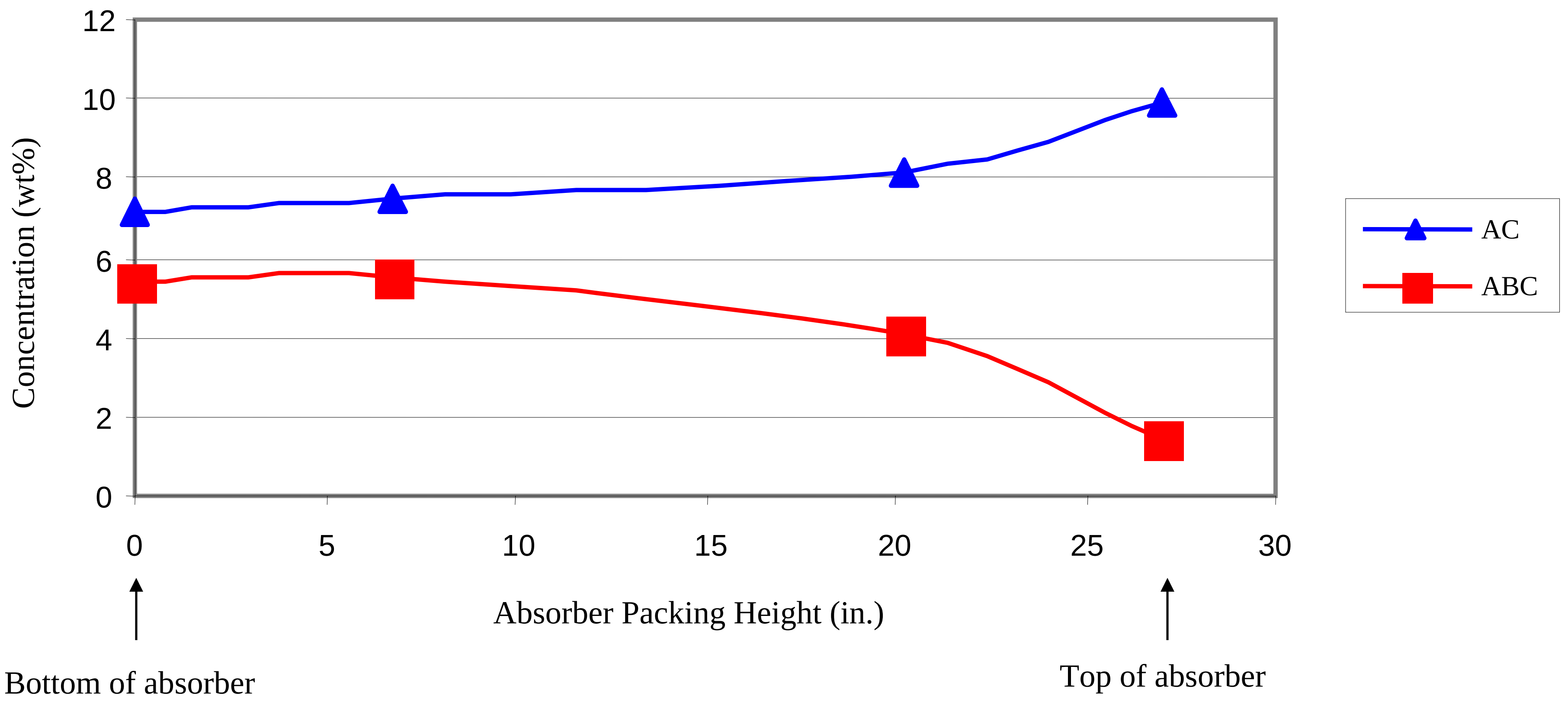
Aqua Ammonia Continuous Unit
Typical Test Results



Aqua Ammonia Continuous Unit
Typical Test Results



Aqua Ammonia Continuous Unit
Typical Absorber Sampling Test Results



Parametric Summary

- **Quantified effects of process parameters**
- **Identified key parameters for optimization**
  - Liquid inventory
  - Regenerator temperature
  - Absorber temperature
  - Gas flow
- **Parameters with lesser effect include:**
  - Initial NH_3 concentration
  - Liquid Flow
  - Packing Height
- **Presence of O_2 had no effect on performance**

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Detailed Parametric Testing

- **Initial parametric tests identified key parameters**
  - Regenerator residence time (liquid inventory)
  - Absorber temperature
  - Regenerator temperature
  - Flue gas flow rate

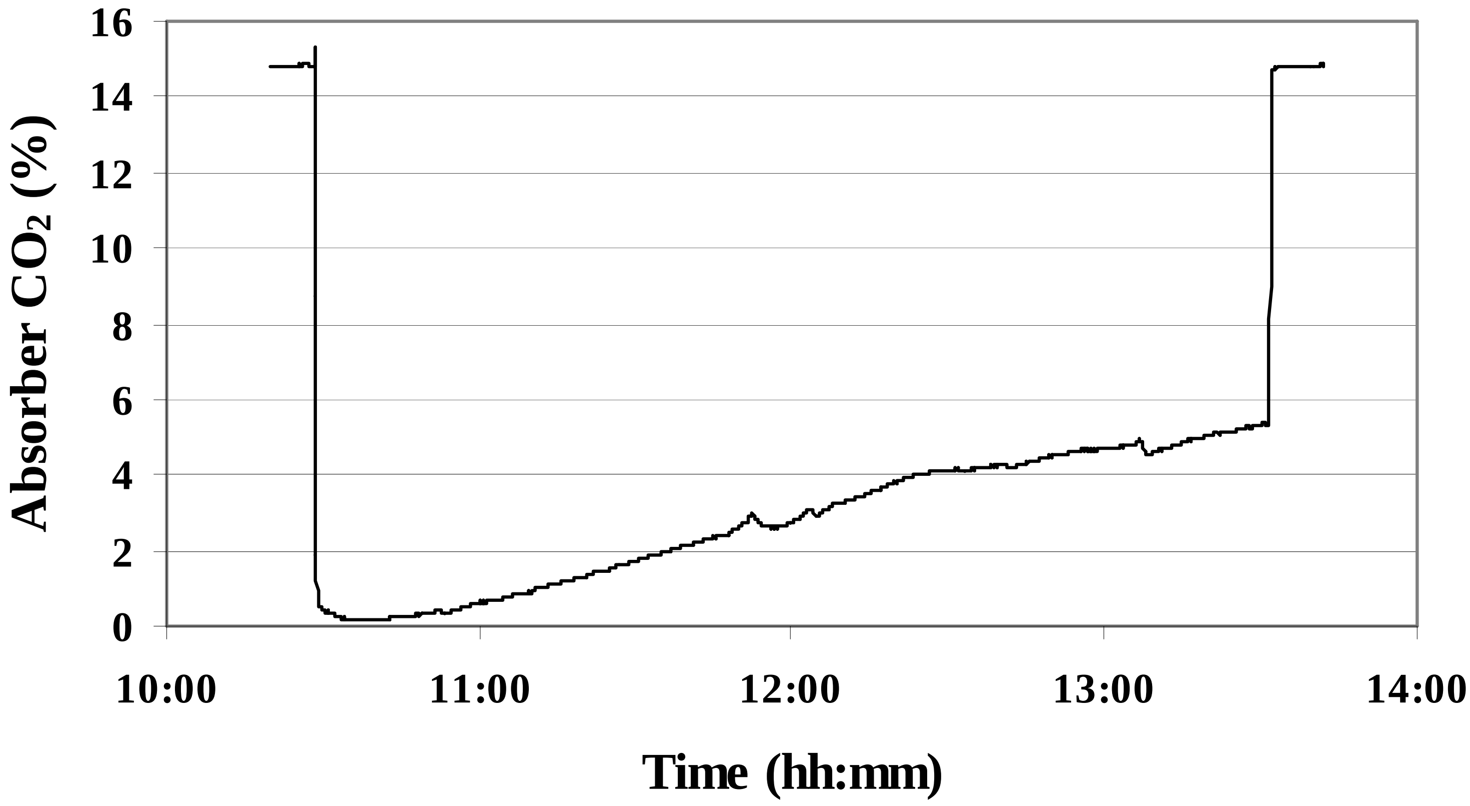
- **Changes in testing / procedure**
  - Carbonization with pure CO_2 to maintain NH_3 content
  - No solution circulation during heat-up
  - Larger regenerator

Detailed Testing – Baseline Conditions

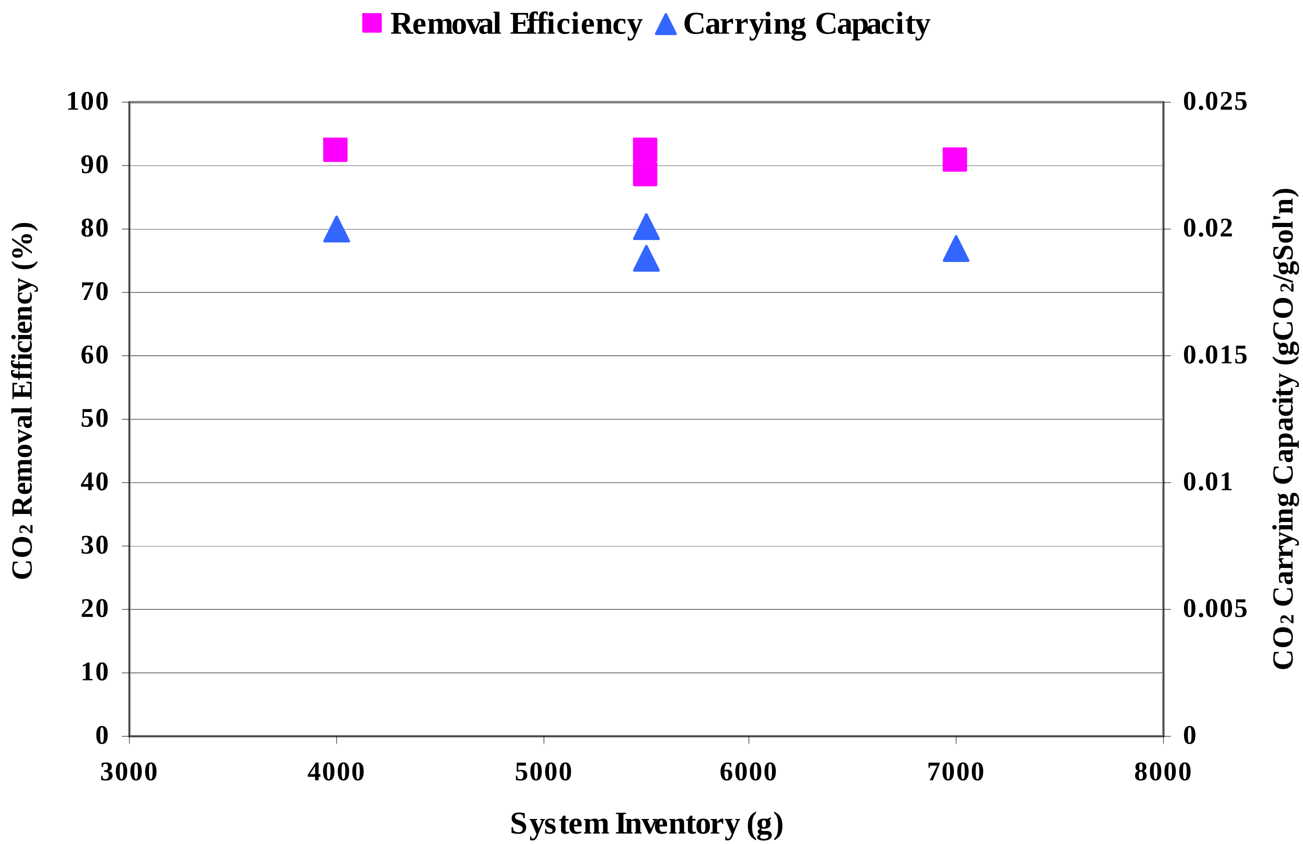
- **Absorber: Temperature: 80^{o}F**
- **Gas Flow: 12,500 sccm (15 s residence time, 15% CO_2)**
- **Packing Height: 68 cm (4 sets BX Gauze packing)**
- **Absorber Diameter: 7.6 cm**
- **Solution: 5500 g of 14% NH_3 solution at startup**

  172 g/min (res. time = 32 minutes, L/G=110 gal/kcf)
- **Saturate with CO_2 at 50^{o}F before heat-up**
- **Regenerator: Temperature: 180^{o}F**
- **Post absorber and regenerator samples extracted and analyzed**

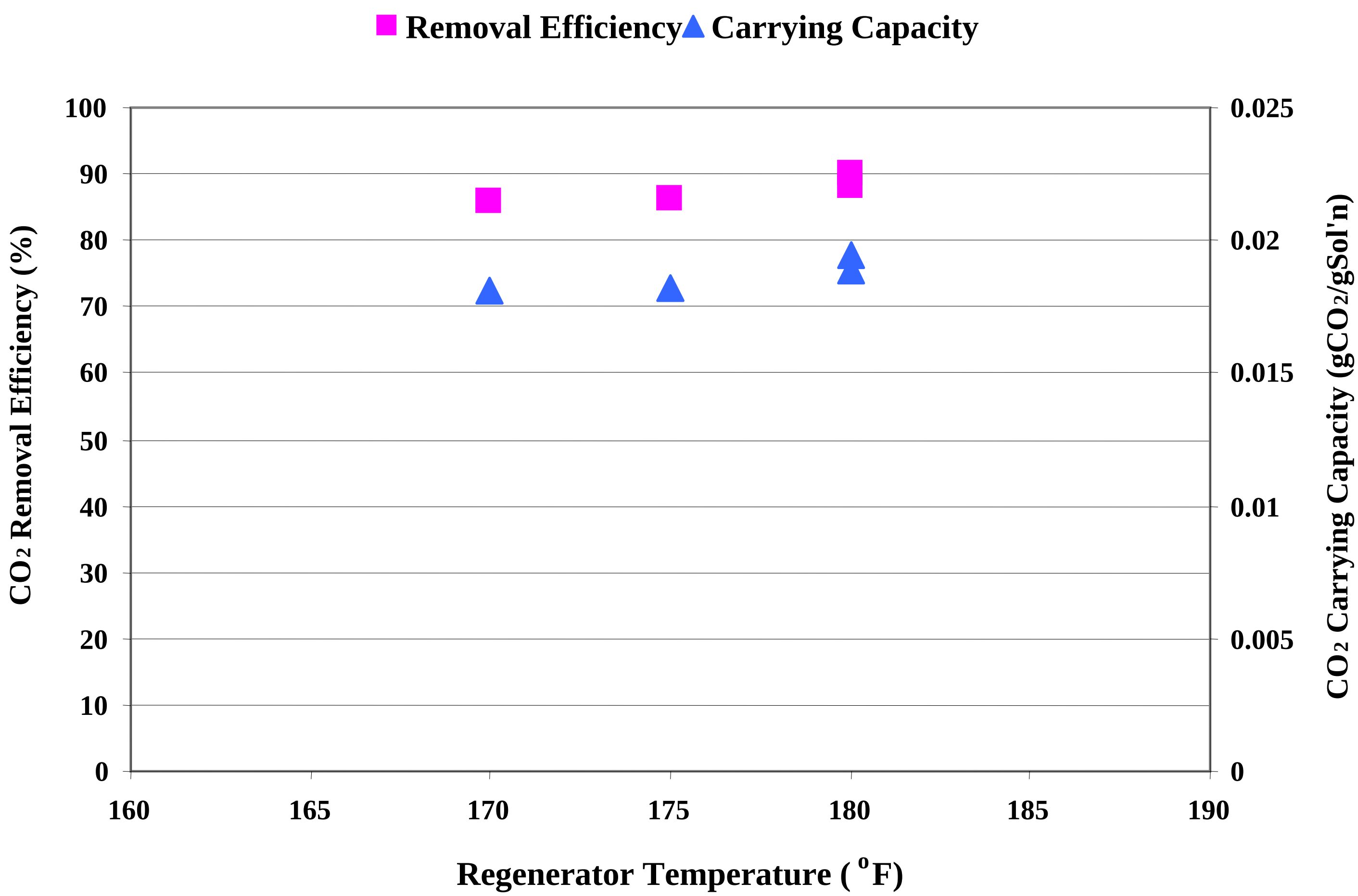
Typical Detailed Test Results



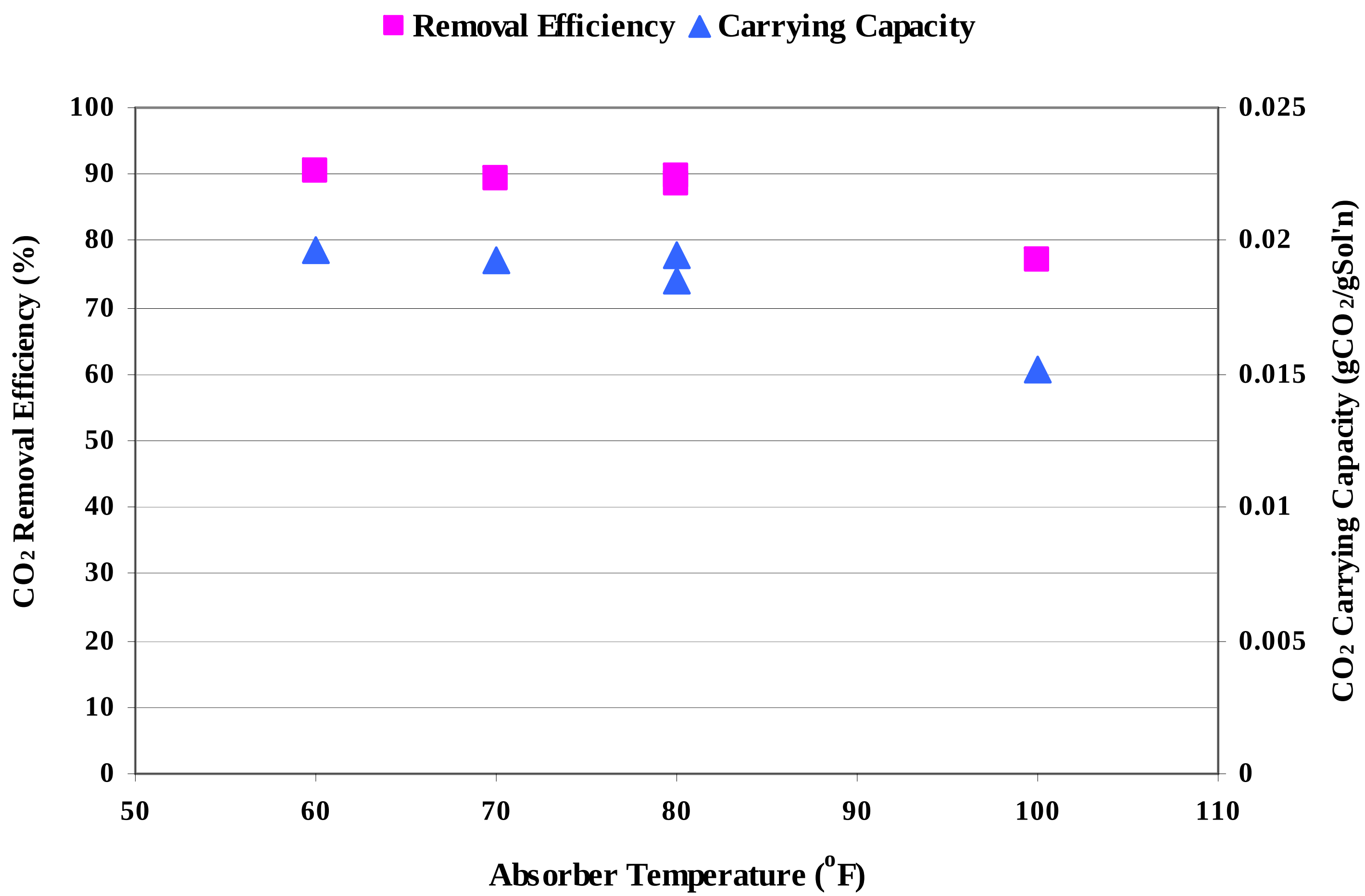
Inventory Effects – Detailed Parametric Testing



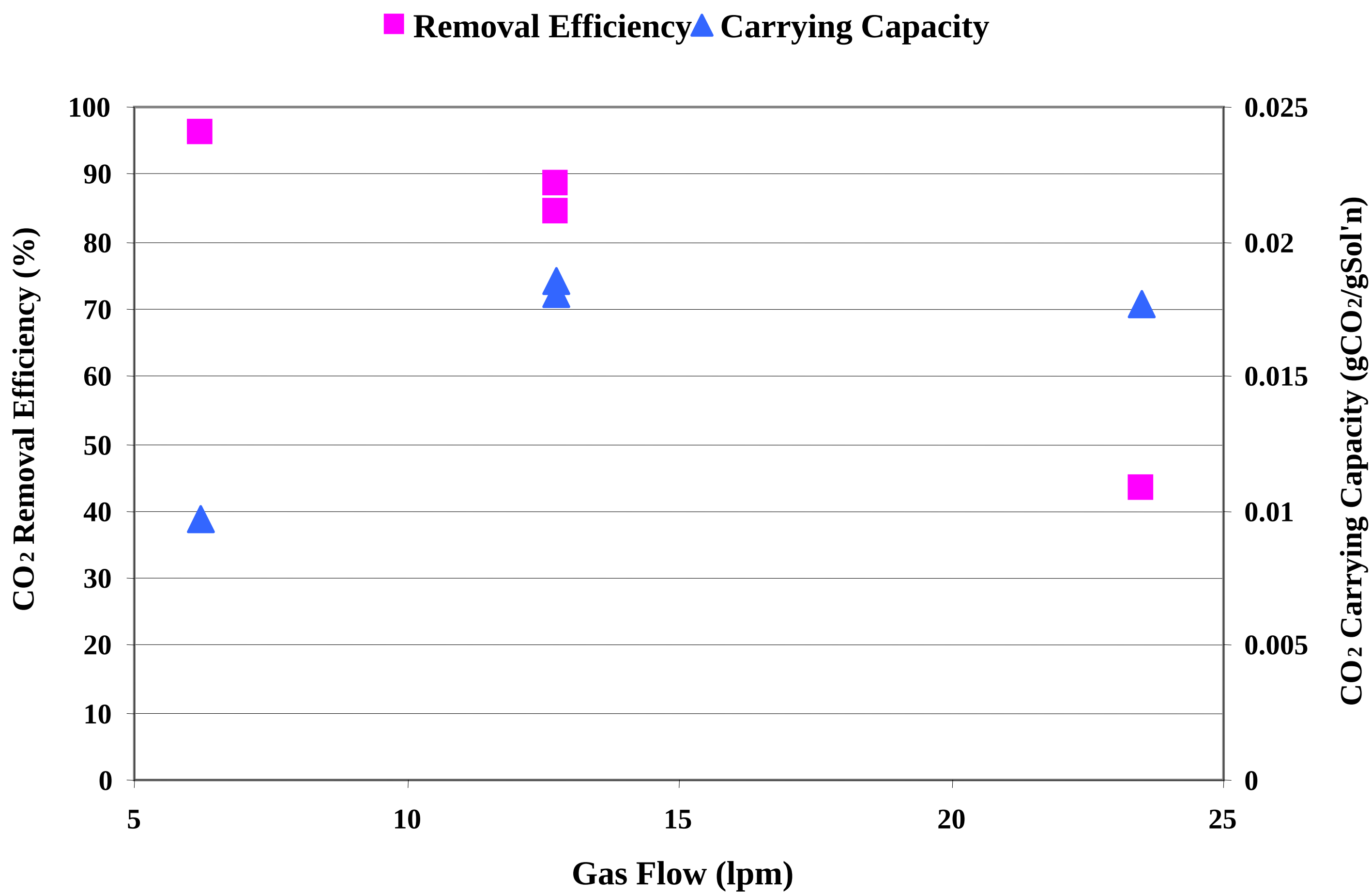
Regeneration Temperature Effects – Detailed Parametric Testing



Absorber Temperature Effects – Detailed Parametric Testing



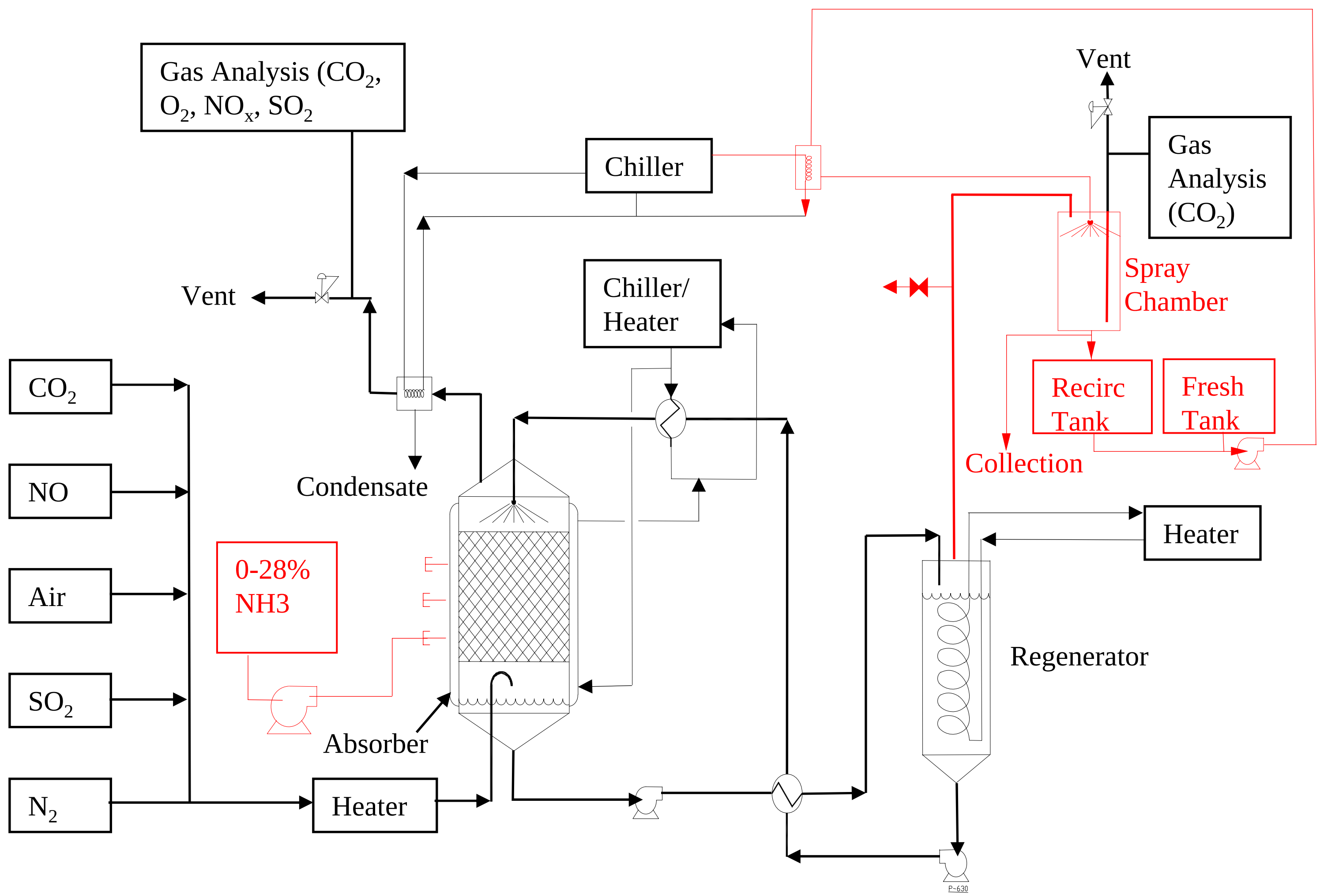
Gas Flow Effects – Detailed Parametric Testing



Plans

- **Modify continuous unit**
  - Add ammonia makeup system
  - Install spray chamber on regenerator to quantify ammonia loss at steady state
- **Run selected tests to define process conditions at steady-state**
- **Revisit systems analysis**

Continuous Process Flow Diagram



Conclusions

- **Continuous unit effective screening tool**
  – Absorber and regenerator temperature, gas flow, and regenerator residence time had greatest impact
  – Liquid flow rate, initial NH_3 concentration, packing had lesser impact
  – Presence of oxygen had no impact
- **Test conditions achieved > 90% CO_2 removal efficiencies**
- **Not true steady state due to ammonia losses**