Optimal Design of Cryogenic Air Separation Columns under Uncertainty

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Cryogenic air separation, while widely used in industry, is an energy intensive process. Effective design can improve efficiency and reduce energy consumption, however, uncertainties can make determination of the optimal design difficult. This paper addresses the conceptual design of cryogenic air separation process under uncertainty. A rigorous, highly nonlinear model of three integrated columns is developed to capture the coupled nature of the process. The multi-scenario approach is used to incorporate the uncertainty, giving rise to a nonlinear programming problem with over half a million variables. Nevertheless, this problem is solved efficiently using IPOPT, demonstrating the effectiveness of interior-point methods on complex, large-scale nonlinear programming problems. The optimal design from the multi-scenario approach is compared against the optimal design using nominal parameter values. As expected, the results using the multi-scenario approach are more conservative than the nominal case; however, they may be less conservative than traditional overdesign factors.

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Introduction

Large quantities of high purity air products are used in several industries, including the steel, chemical, semiconductor, aeronautical, refining, food processing, and medical industries. Methods of air separation include cryogenic and non-cryogenic approaches (Castle, 2002). Although non-cryogenic processes such as pressure swing adsorption and membrane separation have become more competitive, cryogenic distillation technology is still the dominant choice for producing large quantities of very high purity and liquefied air products (Baukal, 1998). Cryogenic air separation is an energy intensive process that consumes a tremendous amount of electrical energy. The U.S. industrial gas industry consumed approximately 31,460 million kilowatt hours in the USA in 1998, which accounts for 3.5% of the total electricity purchased by the manufacturing industry (Karwan et al., 2007).

Optimal operation and control of cryogenic air separation processes has received significant attention, with the primary goal of reducing energy consumption and improving economic performance during operation. Load switching in air separation columns are analyzed by White et al. (1996), and multivariable control schemes for cryogenic air separation are developed in Zhu et al. (2001) and Roffel et al. (2000). Trierweiler and Engell (2000) investigated the selection of an appropriate control...
structure based on dynamic behavior analysis. Seliger et al. (2006) integrated an air separation process model with an IGCC power plant and analyzed the combined process dynamics. Control strategies such as nonlinear model predictive control (NMPC) are difficult to implement for these systems because of the high computational cost associated with optimization of a large, complex dynamic model. Approaches have been developed that promote efficient NMPC for these systems by reducing the size and complexity of the model. Bian et al. (2005) developed a strategy for nonlinear model predictive control by adopting a dynamic wave model for the single nitrogen column. The advanced step NMPC controller (Zavala and Biegler, 2009), an alternative approach based on NLP sensitivity, has also been used in Huang et al. (2009) to perform efficient nonlinear model predictive control of a cryogenic air separation column as a part of an IGCC. Considering offline dynamic optimization, Zhu and Laird (2008) proposed an effective parallel nonlinear solution to deal with optimal control and operation under uncertainty for two highly coupled cryogenic air separation columns.

In addition to process control, previous research has also focused on planning and scheduling for air separation columns. Karwan and Keblis (2007) use a mixed integer programming formulation to optimize operating decisions under real time pricing. Miller et al. (2008) use thermodynamic ideal work to predict the energy requirements when production rates of cryogenic air separation columns change under varying electrical prices. Ierapetritou et al. (2002) use an ARIMA model to predict future
power prices and minimize operating cost using a two stage stochastic programming formulation. Because of the complexity associated with handling uncertainty, much of the existing research regarding design and operation under uncertainty of air separation systems makes use of simplified or linear process models. However, when considering the entire coupled system and the potential for varying operating conditions, air separation plants can exhibit highly nonlinear behavior. There is a need for strategies that can consider uncertainties and provide rigorous optimization of these complex nonlinear models.

Optimizing the design of the cryogenic air separation system has the potential to significantly affect not only the capital investment, but also the future economic performance. In practice, most current design schemes focus on specialized column structures and opportunities for energy and mass integration. Agrawal and coworkers simulate and analyze various thermal coupling methods (Agrawal and Yee, 1994), structured packing on packed columns for argon production (Agrawal, et al., 1993), and multiple component distillation sequences (Agrawal, 1995, 1996) in order to improve energy efficiency and separation performance. Egoshi and coworkers (Egoshi et al., 2002) address the problem of predicting practical separation performance and obtaining the optimal design of cryogenic air separation plants using a rigorous transport model for structured packing. Regardless of the design strategy used, in order to retain future process flexibility it is important to consider potential uncertainties during the design phase. These include uncertainty in process
performance, uncertainty in product demands and pricing, and uncertainty in availability and pricing of process inputs.

One example of uncertainty in the model arises in the selection of thermodynamic methods and parameters. The primary components are separated under extremely low temperatures, and standard packages may not adequately describe the behavior of the system under these conditions. Indeed, many companies specializing in air separation have spent significant resources developing specialized thermodynamic methods for their systems.

A second form of uncertainty relates to unknown demands on the process. Air separation systems can produce three component products of various grades in both vapor and liquid phases. Different customers have different product and purity demands, and these demands can change with seasons and other external factors. It is important to consider this product demand uncertainty during the design phase and develop a process that is flexible enough to meet future product demands.

A third form of uncertainty comes from unknown or varying availability of process inputs and pricing. The dominant operating expense in cryogenic air separation systems is the electricity required by the process. Peak versus off-peak costs and real-time pricing changes, can significantly affect the economic performance of the process. This uncertainty is well studied in a number of articles (Ierapetritou et al., 2002; Karwan and Keblis 2007; Miller et al., 2008).
To handle potential uncertainties in the design phase, the traditional approach is to design the process according to nominal values of the uncertain parameters and then overdesign based on empirical factors. However, this approach may result in infeasible or conservative design decisions. The development of systematic design methods that explicitly consider process uncertainty has been an important research topic for many years (Grossmann and Sargent, 1978; Halemane and Grossmann, 1983). The two dominant approaches for rigorous consideration of uncertainty in optimization are the stochastic programming approach and the chance-constrained approach. Grossmann and Guillén-Gosálbez (2009) recently discussed the opportunities for the use of these approaches in the synthesis and planning of sustainable processes.

In the stochastic programming approach, individual scenarios are included in the optimization formulation for each discrete realization of the uncertain parameters. Continuous uncertainty spaces are usually approximated by appropriate sampling. The problem can be formulated using multiple stages with potential for decisions (or recourse) at each stage. Several good textbooks describe this approach in detail (e.g. Birge and Louveaux, 2000).

In chance-constrained programming, constraints need not be satisfied over the entire uncertainty space, but instead they are required to be satisfied with a given probability. While this explicit description is often desirable, these formulations can be very difficult to solve in the general case.
Multi-scenario optimization is a popular approach for design of chemical processes under uncertainty. Several researchers have investigated effective formulation and solution strategies for this class of problems (Pistikopoulos and Grossmann, 1988a, 1988b; Pistikopoulos and Ierapetritou, 1995; Paules and Floudas, 1992; Varvarezos et al., 1994; Rooney and Biegler, 1999, 2001, 2003; Raspanti et al., 2000), and several well known reviews are available (Pistikopoulos, 1995; Biegler, Grossmann, and Westerberg, 1997; Sahinidis, 2004). Two stages are typically considered in these formulations: the design stage and the operation stage. Values for the design variables must be determined, whereas values of the control variables can be determined during the operational stage when some uncertainties may have been resolved.

Rooney and Biegler (2003) generalize the multi-scenario approach and classify the uncertainties into process variability and process uncertainty. Process uncertainty refers to quantities that are unknown at both the design stage and the operation stage. The design itself should ensure feasibility across these uncertainties. Process uncertainty includes, for example, unmeasured disturbances and uncertain model parameters. Process variability refers to quantities that are uncertain at the design stage but measureable during operation. Process control variables are allowed to change in order to compensate for this variability. While multi-scenario programming is a popular approach, challenges still include efficient solution of these large-scale problems, especially in the general nonlinear case.
The multi-scenario approach is generally viewed as focusing on reliability more than profitability since it requires feasibility of all scenarios. However, when the uncertainty space is continuous, the discrete scenarios represent only a sample of the continuous space. Recent research has demonstrated that this approach represents an approximation of the probabilistic approach and, in special cases, rigorous confidence intervals can be established (Luedtke and Ahmed, 2008; Nemirovski and Shapiro, 2006). These developments, coupled with improvements in optimization tools and computational capability, serve to increase the importance of this approach and its practical applicability.

Considering cryogenic air separation systems, uncertainty can arise from several sources. Process uncertainty (which is unknown during operation) can arise from unknown physical properties. For example, activity coefficient models for N₂–Ar–O₂ systems contain binary interaction parameters that are sensitive to argon purities and pressures (Harmens, 1970). Process variability (or measurable uncertainty) can arise because of changing product demands. In order to satisfy variable product demands, the cryogenic air separation system may be required to switch among different operating conditions. The argon product variability is often ignored; however, it can affect the optimal design significantly.

This paper addresses the conceptual design of cryogenic air separation systems considering an example of both process variability (uncertain thermodynamic parameters) and process uncertainty (uncertain product demands). A rigorous
nonlinear model is developed that includes all three primary separation columns (with recycle) to capture the coupled nature of these systems. A multi-scenario approach is used to discretize the uncertainty space and formulate a large-scale nonlinear optimization problem. The paper compares the nominal optimal design with that resulting from the multi-scenario formulation. This paper further demonstrates that the multi-scenario formulation is an effective tool for optimization under uncertainty, even when the process models are nonlinear and highly complex. The capabilities of off-the-shelf nonlinear programming tools have increased dramatically in the last five to ten years. The nonlinear interior-point solver, IPOPT (Wächter and Biegler, 2006), provides efficient solution of these large-scale problems, and this paper also shows the scalability of this approach as a function of the number of scenarios considered.

The paper is structured as follows. The next section describes the general multi-scenario formulation approach along with an overview of the interior-point solution approach. Next, the cryogenic air separation plant is described, followed by a description of the optimization formulation. The section Numerical Results shows the scalability of IPOPT as the problem size is increased, and discusses the optimal design. Finally, the paper closes with some conclusions about the design of air separation systems considering uncertainty, the use of multi-scenario programming with rigorous nonlinear programming models, and potential decomposition strategies for more efficient solution on parallel computing architectures.
Multi-scenario Programming Approach

The multi-scenario formulation can be expressed in general form as,

$$\min_{d, u, l} P = f_0(d) + \sum_{k \in K} \sum_{q \in Q} \omega_{qk} f_{qk}(d, u_k, l_{qk}, \theta_k^v, \theta_q^u)$$

subject to

$$h_{qk}(d, u_k, l_{qk}, \theta_k^v, \theta_q^u) = 0 \quad k \in K, q \in Q,$$

$$g_{qk}(d, u_k, l_{qk}, \theta_k^v, \theta_q^u) \leq 0 \quad k \in K, q \in Q,$$

where the design variables are given by $d$, control variables are given by $u$, and the state variables are given by $l$. The general equality and inequality constraints are given by $h$ and $g$ respectively. The objective allows for a general function associated with the design variables, as well as a weighted sum of terms from each scenario. The index set $K$ is defined for discrete values of variable parameters, $\theta^v$, and the index set $Q$ is defined for discrete values of unknown parameters, $\theta^u$. We assume that the control variables $u$ can be used to compensate for measured variable parameters, $\theta^v$, but not the uncertainty associated with unknown parameters, $\theta^u$. Thus, the control variables are indexed over the set $K$, while the state variables, are indexed over both $Q$ and $K$.

The objective function includes fixed costs related to the design variables and a weighted sum arising from a quadrature representation of the expected value of the objective over the uncertainty space. Discretization points are selected for this quadrature, however, realizations can be added to enforce feasibility at additional points. This gives a large-scale nonlinear multi-scenario problem with significant coupling or interaction induced by both the control and design variables.
In this paper, we solve the large-scale multi-scenario programming problem using the nonlinear interior-point algorithm IPOPT (Wächter and Biegler, 2006). This algorithm provides efficient handling of large numbers of inequality constraints by shifting variable bounds to the objective in the form of a log-barrier term. The solution of the overall NLP problem is obtained by approximately solving a sequence of these barrier sub-problems where the barrier parameter approaches zero.

To solve each barrier sub-problem, this algorithm calculates the step using a Newton-based approach on the primal-dual optimality conditions. Global convergence is ensured using a filter-based line search strategy. More details on this algorithm can be found in Wächter and Biegler (2006). The software is completely open-source and freely available through the COIN-OR foundation. In this paper, AMPL (Fourer, Gay, and Kernighan, 2006) is used to formulate the optimization problems. AMPL is a powerful modeling language that provides efficient 1st and 2nd order derivative information through automatic differentiation. All optimization problems in this paper were solved using IPOPT version 3.7.1.

Process Description

This paper addresses optimization of a rigorous model of a cryogenic air separation plant, including three coupled primary separation columns. The plant studied includes a double-effect heat integrated distillation column with a side crude argon column (CAC). The CAC introduces one energy integration structure and one recycle loop to the cryogenic air separation problem, making first-principle modeling and
optimization of the system significantly more difficult because of the additional coupling. Figure 2 shows the simplified structure of this particular cryogenic air separation system. The air feed stream is first compressed and pre-purified to remove primary impurities such as water and carbon dioxide. After being cooled by a primary heat exchanger, a portion of the air feed stream is introduced into the low pressure distillation column (LPC). The LPC is designed with 70 theoretical stages. The remaining feed enters the bottom of the high pressure distillation column (HPC), which contains 36 theoretical stages. A side vapor stream is withdrawn at the 28th tray of the LPC and is fed into the CAC for distillation. The liquid from the bottom of the CAC is returned to the LPC at the location of vapor stream withdrawal. The
oxygen-rich liquid from the bottom of the HPC contains approximately 62% nitrogen. This stream passes through a heat exchanger then is split into two streams, where one portion of this liquid flow will enter into the LPC for further distillation, and the remaining portion of this liquid is used as cold resource for the condensation of argon products at the top of the CAC before re-entering the LPC as feed flow. The reflux flow of the LPC is withdrawn from the top of the HPC and passes through the same heat exchanger as the oxygen-rich liquid described previously.

Liquid oxygen product is withdrawn from the combined condenser/reboiler, and vapor oxygen product is withdrawn from the bottom of the LPC. Liquid nitrogen product is withdrawn from the top of the HPC, while gas nitrogen product is withdrawn from the top of the LPC. Crude argon product is withdrawn from the top of the CAC.

This is a highly integrated system that can be very difficult to design and operate. Typically, there are a large number of design variables which need to be determined in the detailed design phase of a cryogenic air separation plant. However, this study mainly focuses on conceptual design under uncertainty. Therefore all valves are assumed to be throttle expansion valves, the mass loss in pipelines are assumed to be negligible, and constant heat transfer area and coefficients are used in the heat exchanger calculations.

Based on process dynamics of the cryogenic air separation system, five main control variables, $u = [U_1, U_2, U_3, U_4, U_5]$, are selected to compensate for variability of
argon product demands. These variables are defined as the feed air stream of the HPC ($U_1$), the feed air stream of the LPC ($U_2$), the reflux flow from the HPC to the LPC ($U_3$), the waste nitrogen stream ($U_4$), and the side withdrawal from the LPC to the CAC ($U_5$). The five main design variables are the diameters of the three distillation columns (the HPC, LPC and CAC), the heat transfer area of the combined condenser/reboiler, and the brake horsepower of the compressor. Table 1 shows the nominal operating conditions of the plant used in the case study.

<table>
<thead>
<tr>
<th>Variables (Units)</th>
<th>Nominal Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas oxygen product, mol/s</td>
<td>2.44</td>
</tr>
<tr>
<td>Liquid oxygen product, mol/s</td>
<td>0.64</td>
</tr>
<tr>
<td>Oxygen product purity</td>
<td>≥98%</td>
</tr>
<tr>
<td>Gas nitrogen product, mol/s</td>
<td>13.13</td>
</tr>
<tr>
<td>Nitrogen product purity</td>
<td>≥99.99%</td>
</tr>
<tr>
<td>Argon product purity</td>
<td>≥96%</td>
</tr>
<tr>
<td>Pressure of LPC, MPa</td>
<td>0.13-0.14</td>
</tr>
<tr>
<td>Pressure of HPC, MPa</td>
<td>0.68-0.69</td>
</tr>
<tr>
<td>Pressure of CAC, MPa</td>
<td>0.12-0.13</td>
</tr>
</tbody>
</table>

**Mathematical Model of the Process**

The distillation column (the LPC, the HPC, and the CAC) models are derived from the mass and energy balances coupled with the equilibrium relationships. In this rigorous model, key assumptions include: 1) complete mixing on each tray and 100% tray efficiency; 2) negligible heat losses in the tray; 3) constant pressure drop on each tray; 4) uniform pressure and temperature on each tray.
The mass balances for each tray are given by,

\[ F_j^V + F_j^L + V_{j-1} + L_{j-1} - V_j - S_j^V - L_j - S_j^L = 0 \]  

where \( j \) is the index of each tray from the top of each column, \( F_j^V \) and \( F_j^L \) are the vapor and liquid molar feed flows entering into the \( j^{th} \) tray, and \( S_j^V \) and \( S_j^L \) are the vapor and liquid molar side flows out of the \( j^{th} \) tray. The vapor and liquid flow rates are given by \( V_j \) and \( L_j \), respectively. Component mass balances are given by,

\[ V_{j+1} y_{i,j+1} + L_{j-1} x_{i,j-1} + F_j^V y_{i,j} + F_j^L z_{i,j} - \left( V_j + S_j^V \right) y_{i,j} - \left( L_j + S_j^L \right) x_{i,j} = 0 \]  

where \( i \) is the index of each component (1-nitrogen, 2-argon, 3-oxygen), and the liquid and vapor compositions are given by \( x_{i,j} \) and \( y_{i,j} \), respectively. The vapor and liquid compositions of feed flows entering the \( j^{th} \) tray are \( z_{i,j}^V \) and \( z_{i,j}^L \), respectively.

The model includes tray by tray energy balances, expressed by,

\[ V_{j+1} H_j^V + L_{j-1} H_j^L + F_j^V H_j^F + F_j^L H_j^{FL} - \left( V_j + S_j^V \right) H_j^V - \left( L_j + S_j^L \right) H_j^L = 0 \]  

where \( H_j^F \) and \( H_j^{FL} \) are the vapor and liquid enthalpies of feed flow entering into the \( j^{th} \) tray. The vapor and liquid enthalpies in the \( j^{th} \) tray are \( H_j^V \) and \( H_j^L \) respectively. The temperature dependence of the enthalpies were represented using a high-order polynomial fit to simulation data.

The vapor-liquid equilibrium expressions for each tray are given by,

\[ y_{\theta} = y_{\theta}^0 K_{\theta} x_{\theta} \]  

\[ K_{\theta} = P_j^0 \left( T_j \right) / P_j \]  

\[ \log y_{i,j} = \left( \alpha_{12} x_{\theta}^2 + \alpha_{31} x_{\theta}^2 + \left( \alpha_{12} + \alpha_{31} - \alpha_{23} \right) x_{\theta} \right) T_j \]
log \gamma_{2j} = \left( a_{12} x_{1j}^2 + a_{23} x_{3j}^2 + (a_{12} + a_{23} - a_{13}) x_{1j} x_{3j} \right) / T_j \quad (8)

log \gamma_{3j} = \left( a_{23} x_{2j}^2 + a_{13} x_{1j}^2 + (a_{23} + a_{13} - a_{23}) x_{2j} x_{1j} \right) / T_j \quad (9)

where \( T_j \) is the temperature of the \( j^{th} \) tray. The activity coefficients, \( \gamma \), are calculated using Margule’s equation, and ideal vapor-liquid equilibrium constants \( K_{ij} \) are calculated using Antoine’s equation with saturation pressure \( P_i^s \). The variables \( \alpha_{mn} \) are the binary interaction parameters of activity coefficients.

The following expressions are used to capture design relationships (Douglas, 1988; Peters et al., 2002). Column diameters are given by,

\[
D_{m,j} = 0.0164 V_m^0.5 \left[ 378 M_g^2 \left( \frac{T_{m,j}}{520} \right) 14.7 \right]^{1/4} \quad (10)
\]

where \( P_m \) is the tray pressure of each column, and \( M_g \) is the molecular weight of distillate.

\[
D_m = \max \left( D_{m,j} \right), m \in \{ LPC, HPC, CAC \}
\]

The height of each column is,

\[
H_m = 2.4 n_m, \quad (12)
\]

where \( n_m \) is the number of stages in each column and the heat transfer area in the combined condenser/reboiler can be described by,

\[
A = \frac{Q_i}{(U \Delta T)} \quad (13)
\]

where \( \Delta T \) is the temperature driving force. \( Q_i \) is the transferred heat between the LPC and the HPC, and \( U \) is the heat transfer coefficient.
The capital costs of column shells and trays (CSC and CTC) are estimated with the following equations:

\[
CSC_m = \left( \frac{M \ & \ S}{280} \right) 102 D_m^{1.066} H_m^{0.802} \left( c_m + c_m c_p \right)
\]

\[
CTC_m = \left( \frac{M \ & \ S}{280} \right) 4.7 D_m^{1.55} H_m \left( c_s + c_t + c_m \right)
\]

Here, M&S is the Marshall and Swift index. The parameters \(c_p, c_m\) and \(c_m\) are the pressure range, construction material and installation cost coefficients. The parameters \(c_s\) and \(c_t\) are the tray spacing and design cost coefficients, respectively.

The capital cost of heat exchanger (HEC) in combined condenser/reboiler is,

\[
HEC = \left( \frac{M \ & \ S}{280} \right) 102 A^{0.65} \left( c_m + c_m (c_s + c_p) \right)
\]

and the capital cost of the main compressor (CPC) is,

\[
BHP = \left[ \frac{(U_1 + U_2)}{1 - \Delta F_i} \right] \frac{k}{k - 1} \frac{\text{RT}}{P_m} \left[ \left( \frac{P_{out}}{P_{in}} \right)^{\frac{k-1}{k}} - 1 \right]
\]

\[
CPC = \left( \frac{M \ & \ S}{280} \right) 518 \left( BHP \right)^{0.82} \left( c_m + c_s \right)
\]

where \(BHP\) is the brake horsepower of the compressor. The entrance and exit pressures of the compressor are \(P_{in}\) and \(P_{out}\), and \(\Delta F_i\) is the loss amount of the feed flow in the compressor. The adiabatic index number of the gas is given by \(k\).

Because the major operating cost of cryogenic air separation processes is required electrical power, we assume the other operating costs can be ignored. Here, we also assume that a liquefier is not installed in the system. The power price is
assumed to be constant in this study, however, more complex formulations considering uncertain power prices will be investigated in future work.

The electricity cost ($EC$) is given by,

$$EC = \frac{C_{ele} \cdot BHP}{\eta},$$

(19)

where $C_{ele}$ is electricity price ($0.0574/(kWh)$), and $\eta$ is the efficiency of the compressor (0.75). The total annual cost (TAC) of our air separation process is given by the following form,

$$TAC = \frac{\sum_m (CSC_m + CTC_m + HEC + CPC)}{t_p} + \sum_{k \in K} \sum_{q \in Q} \left( \omega_{qk} E_{C_qk} \right),$$

(20)

where $t_p$ is the payback time, which is assumed to be 3 years. The last term in Eq. (20) is a numerical integration for the expected value of the operating cost. In the case studies, we assume that variability and uncertainty are both uniformly distributed. Therefore, the weights $\omega_{qk}$ are all equal. More accurate quadrature rules could be used along with other distributions. Other costs such as pipelines and valves are not included in this study.

**Numerical Results**

The base formulation described in the previous section is used to find the optimal design for the nominal case. In addition, a multi-scenario formulation is developed that considers uncertainty in argon product demands and the thermodynamic parameter $\alpha_{12}$, and evaluates the objective using the expected value of the operating
costs. Before discussing the optimization results in detail, we first present the timing results showing the scalability of the multi-scenario approach with IPOPT.

Argon product demands are assumed to be uniformly distributed between 0.1063 (-20%) mol/s and 0.1595 (+20%) mol/s, and the binary interaction parameter, $a_{12}$, is assumed to be uniformly distributed between 7.0 and 9.5 (Harmens, 1970). Figure 2 shows the IPOPT solution times using the default options. The same number of discretizations is used for each uncertain parameter, and the category labels give the total number of scenarios considered for each run. The white bars on the left list the average CPU time for each iteration. The grey bars on the right list the total CPU time in seconds. Note that the number of iterations need not be the same for each case. Furthermore, by default IPOPT uses exact first and second derivative information, and the number of iterations remains relatively constant as the size of the problem increases.
Figure 2. Timing Results for Multi-scenario Approach (Default Options)

Figure 3 shows the timing information using the quasi-Newton approach within IPOPT. With this option, the Hessian information is approximated using a limited memory BFGS update. Similar scaling is seen for this approach. The number of variables in the 4 scenario case is approximately 8,000, while the number of variables in the 196 scenario case is approximately 675,000. These results demonstrate that off-the-shelf nonlinear programming tools are able to scale effectively to reasonably large problems, even when the models are highly coupled and nonlinear.
Figure 3. Timing Results for Multi-scenario Approach (L-BFGS)

Taking the largest number of scenarios (196 scenarios), optimal results from the multi-scenario formulations are compared with optimal results for the nominal case in Table 2. As expected, the design is more conservative when uncertainty is considered. The optimal diameter of the HPC is the least sensitive to the uncertainty considered here.

<table>
<thead>
<tr>
<th>Variables</th>
<th>Nominal Case</th>
<th>Multi-Scenario Formulation</th>
<th>Difference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Dia. of LPC, m</td>
<td>0.66</td>
<td>0.76</td>
<td>15.65%</td>
</tr>
<tr>
<td>Dia. of HPC, m</td>
<td>0.88</td>
<td>0.95</td>
<td>8.24%</td>
</tr>
<tr>
<td>Dia. of CAC, m</td>
<td>0.44</td>
<td>0.54</td>
<td>23.42%</td>
</tr>
<tr>
<td>BHP, Kw</td>
<td>90</td>
<td>113</td>
<td>25.572%</td>
</tr>
<tr>
<td>Heat exch. area, m²</td>
<td>24</td>
<td>26</td>
<td>11.45%</td>
</tr>
<tr>
<td>TAC, $10⁵</td>
<td>1.412</td>
<td>1.586</td>
<td>12.35%</td>
</tr>
</tbody>
</table>

The diameter of the CAC and the brake horsepower are significantly affected. This is reasonable, since the variability in argon demands will require greater process flexibility.
This result also shows that it is not optimal (and may not be feasible) to absorb potential argon variability by operational changes alone. Both design and operation changes should be considered. The effects of these uncertainties on the diameter of the LPC are more dramatic than on the diameter of the HPC. This is expected given the variability in argon production and the integration between the LPC and the CAC. Increased withdrawal from the LPC to the CAC, coupled with variability in recycle from the CAC, requires increased flexibility in the LPC. In contrast, the CAC is less tightly integrated with the HPC.

Figure 4 shows how the optimal design changes as a function of the number of scenarios considered. The values for the argon demand and the uncertain binary interaction parameter were selected randomly from the ranges given previously. While it is difficult to guarantee that the scenarios sufficiently span the space of variability and uncertainty, it can be seen that the multi-scenario design solution converges as we increase the number of scenarios.
Figure 4. Dependence between multi-scenario design and increasing scenario number

Summary & Conclusions

This work uses a multi-scenario approach to determine the optimal design of a cryogenic air separation process considering two classes of uncertainty. Process variability describes uncertainty that is measurable during operation, and control variables can be used to compensate for this uncertainty. Process uncertainty represents unmeasurable quantities like uncertain model parameters or unmeasured disturbances. In this paper, argon product demands were selected as an example of process variability, and unknown activity coefficients were selected as an example of process uncertainty.

As expected, the optimal design is more conservative when uncertainties are considered. However, the multi-scenario approach provides a more rigorous treatment of
uncertainty than applying traditional overdesign factors. The approach allows for a more efficient design by capturing the potential for operational changes in the control variables as a function of process variability. Furthermore, nonlinear interactions between the uncertainties, the design decisions, and these potential control possibilities are rigorously captured.

While multi-scenario programming is a popular approach for treatment of uncertainty in optimization, it can be challenging to find efficient solution strategies for these large-scale problems, especially in the general nonlinear case. Nevertheless, there have been significant advancements in nonlinear programming algorithms, and the capabilities of general off-the-shelf solvers (e.g. IPOPT) have increased dramatically. The largest multi-scenario problem considered in this paper includes 196 scenarios and more than 675,000 variables. Nevertheless, this formulation solves in under 20 minutes on a standard desktop computer. These results show that recent algorithm improvements, coupled with continued increases in computational capability, allow practical application of the multi-scenario approach with rigorous, large-scale nonlinear models. This will be even more evident as we continue to develop algorithms that can exploit modern computing architectures to promote efficient solution in parallel.

**Future Work**

In this study, a rigorous model of an air separation process was developed that considered three highly integrated columns. The two uncertainties considered were a thermodynamic interaction parameter and the argon product demand. Future work is
needed to include treatment of additional uncertainties. A key variability during operation is the price of electricity, which is the dominant operating cost for the process. A careful analysis will help engineers further quantify the impact of this and other uncertainties on design and operation.

This work used a steady-state model and assumed perfect control was possible. Given the potential control challenges with such a highly integrated process, these optimization formulations should consider integrated design and control.

Finally, the main challenge in multi-scenario optimization is still efficient solution of the large-scale problem. The dominant computational expense of the IPOPT algorithm is the solution of the augmented linear system resulting from a Newton iteration of the primal-dual equations. Given a problem with a specialized structure, decompositions are possible that can exploit this structure and produce efficient solutions in parallel. We have developed a package, SCHUR-IPOPT, that uses an internal decomposition approach for the parallel solution of structured nonlinear programming problems based on the serial IPOPT algorithm. For the general design under uncertainty formulation, previous results on a large distributed cluster have demonstrated that the solution time is almost constant as scenarios and processors are added (Laird and Biegler, 2008; Zavala et al., 2008; Zhu and Laird, 2008). In the general multi-scenario formulation considering both process variability and process uncertainty, there is additional structure. If the problem is decomposed with a single scenario for each processor, then the common variables in the parallel decomposition
include both the control variables and the design variables. However, there is no restriction that each individual block needs to consider only a single scenario. If the problem is decomposed over the process variabilities only, then the number of common variables considered in the parallel decomposition includes only the design variables. With this scheme, the coupling induced by the control variables is handled internally by the serial linear solver. Furthermore, nested decomposition strategies are possible to promote further parallelization. Future work will include the development of specialized decomposition strategies for this nested structure.

**Nomenclature**

\( F = \text{feed flow rate, mol/s} \)
\( K = \text{ideal vapor-liquid equilibrium constant} \)
\( V = \text{vapor flow rate, mol/s} \)
\( L = \text{liquid flow rate, mol/s} \)
\( S = \text{side flow rate, mol/s} \)
\( H = \text{vapor or liquid enthalpies} \)
\( T = \text{tray temperature} \)
\( P = \text{tray pressure} \)
\( Q = \text{transferred heat} \)
\( UA = \text{heat transfer rate, W/K} \)
\( x = \text{liquid flow composition} \)
\( y = \text{vapor flow composition} \)
\( z = \text{feed flow composition} \)
\( U_1 = \text{air feed flow rate in HPC, mol/s} \)
\( U_2 = \text{expand air flow, mol/s} \)
\( U_3 = \text{nitrogen reflux from HPC to LPC, mol/s} \)
\( U_4 = \text{waste nitrogen, mol/s} \)
\( U_5 = \text{feed flow rate of crude argon column, mol/s} \)
\( D = \text{Diameter of distillation columns, m} \)

**Greek letter**

\( \gamma = \text{Activity coefficient} \)
\( \eta = \text{Compression efficiency} \)
\( k = \text{Adiabatic index number in compressor} \)
Abbreviation

HPC = High Pressure Distillation Column
LPC = Low Pressure Distillation Column
CAC = Crude Argon Column
TAC = Total Annual Cost
EC = Electricity Cost
BHP = Brake Horsepower of the Compressor
CPC = Capital Cost of Main Compressor
HEC = Capital Cost of Heat Exchanger
CSC = Capital Cost of Column Shells
CTC = Capital Cost of Column Trays

References


