Framework for embedding black-box simulation into mathematical programming via kriging surrogate model applied to natural gas liquefaction process optimization

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Abstract

This paper presents a framework to solve the constrained black-box simulation optimization problem that arises from the optimal energy-efficient design of single-mixed refrigerant natural gas liquefaction process using reliable process simulator. Kriging surrogate model is used to introduce simple, computationally inexpensive, and effective algebraic formulations with reliable derivatives to the black-box objective and constraints functions. The algebraic surrogate optimization problem is embed into a nonlinear programming (NLP) model in GAMS. The NLP problem is solved using efficient multi-start gradient-based optimization with CONOPT local solver to determine a candidate of decision variables for which the true functions are calculated in the rigorous simulation. The single-mixed refrigerant process is analyzed considering 1-to-3-stage expansion and phase separation to assess potential energy savings. The present approach results show that more expansion stages can provide energy savings from 10.02 to 14.71 % comparing 2-stage and 3-stage expansion system with 1-stage. This optimization framework is more effective and consistent than Particle Swarm Optimization and Genetic Algorithm given the same budget of simulation evaluations for the considered simulation optimization problems resulting in 12.02 to 34.69 % savings.

Keywords: Simulation optimization, Kriging, Process simulation, Surrogate-based optimization, Natural gas liquefaction, Mathematical programming.

1. Introduction

Modeling and simulating complex systems rigorously with pure symbolic formulations and analytical methods can become very complicated with increasing size and complexity of the model, which can be a system of nonlinear algebraic or algebraic-differential equations. Therefore, much of today's engineering applications uses rigorous computer codes (simulation) to describe complex systems employing state-of-the-art numerical methods, in a black-box fashion [1]. In other words, for a given input $\mathbf{x} \in \mathbb{R}^n$, the simulation calculates the response variables of interest $\mathbf{y} \in \mathbb{R}^p$, in which n and p are the number of independent and dependent variables in the simulation, respectively. Although very efficient to describe in details complex

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systems that would otherwise have to be simplified or approximated, a drawback of using black-box models is the lack of symbolic formulation of the model equations and the analytical derivatives that are useful for optimization, for example. The use of simulation may also introduce noise to the calculations due to convergence and approximations of numerical methods [2]. In that sense, the optimization models that require simulations to calculate the objective function and/or constraints are often referred to as simulation optimization problem [3]. A simplified version of this class of problems can be described as to find an $\mathbf{x}^* \in \mathbb{R}^n$ that solves globally the following constrained problem

$$\begin{aligned} \min_{\mathbf{x}\in\mathcal{D}} \quad & f(\mathbf{x}) \\ s.t. \quad & \mathbf{g}(\mathbf{x}) \leq 0, \end{aligned} \tag{1}$$

in which the objective function $f : \mathbb{R}^n \mapsto \mathbb{R}$ and constraints $\mathbf{g} : \mathbb{R}^n \mapsto \mathbb{R}^q$, in which q is the number of constraints, are somewhat expensive to calculate, slightly noisy, and black-box functions, *i.e.* there is no available mathematical expression for f or $\mathbf{g}$, but for a given $\mathbf{x} \in \mathcal{D} \subseteq \mathbb{R}^n$ the value of $f(\mathbf{x})$ and $\mathbf{g}(\mathbf{x})$ are calculated in a computer code simulation with some noise. Besides being expansive to calculate and black-box, these functions that relies on the simulation can be noisy due to numerical approximations and convergence tolerance, which can jeopardize the calculation of accurate approximate derivatives and, therefore, the use of gradient-based optimization methods directly [2]. Also, the lack of analytical formulations of the optimization problem prevents the derivation of rigorous upper and lower bounds of the functions that are used for deterministic global optimization [4].

Knowing that the functions in the simulation optimization problem as in Eq. (1) are expensive to calculate, black-box, and noisy, methods like gradient-based with stochastic approximation, direct search, random search, and response surface are suited to solve it [3]. The latter group of methods, also known as surrogate-based optimization, has shown promising results in recent years [5]. The main idea behind these methods is to construct surrogate models, also known as meta-models or response surfaces, of the simulation-dependent, black-box functions from the optimization problem. These cheaper-to-evaluate surrogate functions are used to determine a candidate solution of the black-box optimization problem via optimization of either an acquisition function or the surrogate functions directly [6]. The latter approach has been investigated extensively with important classic works. For instance, Kushner [7] considered the probability of improvement to find promising next iterates to optimize the unknown function. Sacks et al. [8] analyzed maximizing the integrated mean squared error, the maximum mean squared error, and entropy of the surrogate model to guide the search. Jones el al. [9] used an efficient global optimization approach to the expected improvement acquisition function. Schonlau et al. [10] developed a general acquisition function for the constrained problem, which is a generalization of the expected improvement criterion averaged by the joint probability of constraints being feasible.

More recently, boosted by recent advances in machine learning models, the surrogate-based optimization approaches that solves the surrogate optimization problem directly to guide the search toward the opti-

mization of the true functions have gained popularity. Besides machine learning models, such as artificial neural networks, random forests, radial basis functions, polynomials, etc., the kriging [11] surrogate model has driven researches' attention as it is an interpolating model, which has few parameters to adjust, is cheap to evaluate, and has explicit algebraic formulation. Some important contributions to this surrogate-based optimization field of research includes Davis and Ierapetritou [12], which developed a kriging-based optimization with response surface model optimization for local refinement. Caballero and Grossmann [2] proposed a trust-region algorithm using kriging surrogate models of slightly noisy black-box functions embedded with implicit (non-noisy simulation functions) and algebraic equations in nonlinear programming (NLP) problems and solved each sub-problem with SNOPT solver. The approach was applied to the design of distillation columns, sequence of distillation columns, and production of phthalic anhydride from o-xylene. Cozad et al. [13] proposed the ALAMO software, which uses a global mixed-integer nonlinear programming (MINLP) approach to symbolic regression considering an ensemble of surrogates and aiming simple function formulation that can be efficiently used for optimization. Boukouvala and Floudas [4] developed an iterative framework, called ARGONAUT, composed by bounds tightening, sampling, surrogate function selection, global optimization of the surrogate-embedded NLP problem using ANTIGONE solver, and collection of new sampling points until convergence. Wang and Iearapetritou [14] proposed a kriging-based framework for the optimization of stochastically constrained problems. The main contribution of this work was to present the "feasibility-enhanced Expected Improvement" acquisition function, which explicitly improves the feasibility knowledge while searching for a new sample point. Quirante et al. [15] embedded process simulator to generalized disjunctive programming problems considering integer variables and using kriging surrogate model for noisy black-box functions and implicit equations for non-noisy ones. The logic-based Outer Approximation solver was employed with SNOPT for NLP sub-problems using TOMLAB-MATLAB interface, and the approach was applied to the synthesis of vinyl chloride monomer production process. Schweidtmann and Mistos [16] proposed the MAiNGO software, which is a deterministic global optimization solver for NLP problems with artificial neural networks surrogate models embedded. The solver uses McCormick relaxations in a reduced space employing the convex and concave envelopes of the nonlinear activation function. The framework was tested on four optimization examples: an illustrative function, a fermentation process, a compressor plant and the cumene production process. Thebelt et al. [17] developed a framework, called ENTMOOT, for embedding trained gradient boosted trees surrogate models into larger NLP problems and tested it on constrained global optimization test problems and on a fermentation process. Kim and Boukouvala [18] proposed a framework to simulation-based MINLP problems that uses adaptive sampling and surrogate modeling with one-hot encoding (without integer variables relaxation) that resulted in accurate and robust mixed-variable kriging and neural network models, which were effective surrogates for optimization. The approach was tested on MINLP benchmark problems and a chemical process synthesis case study.

One application of simulation optimization problem is in the design of natural gas liquefaction processes. These cryogenic refrigeration processes consist of cooling down the natural gas to about -160 °C at slightly above ambient pressure to liquefy, store, transport, and commercialize it safely [19]. These processes consume significant amounts of energy and their optimal design are very important to reduce the cost of liquefied natural gas (LNG) as its liquefaction is responsible for 40–60 % of the costs of the LNG value chain, depending on the site conditions and available liquefaction technology [20]. Also, they are of extreme importance currently as the natural gas demand is expected to increase 29.4 % from 2019 to 2040, accordingly to the International Energy Agency [21]. This expected increase in natural gas demand is mainly due to the increasingly global energy consumption, which is expected to go from 603 to 715 trillion MJ per year from 2019 to 2040 [21], and for it being a cleaner and economically competitive energy source compared to other fossil fuels, such as oil and coal.

The natural gas liquefaction process that uses a mixture operating in a single refrigeration cycle and explores its temperature range of evaporation as heat sink to cool and liquefy both the natural gas stream and itself in a multi-stream heat exchanger (MSHE) is called single-mixed refrigerant (SMR) process. From the simulation point of view, the main modeling challenges in this process come from the vapor-liquid equilibrium calculations at below ambient temperature and the Pinch-like calculation in the multi-stream heat exchangers considering phase change and rigorous thermodynamic calculations at cryogenic conditions. Therefore, it is useful to employ chemical process simulators, where all of these difficult calculations are performed with state-of-the-art methods, equations, and empirical coefficients. The optimization part of the problem consists of choosing the process degrees of freedom, such as refrigerant composition and the thermodynamic cycle conditions to improve a process metric of interest. For that, optimization techniques have been used extensively in the past decade, as reported in the annotated bibliography from Austbø et al. [22].

Some of the important contributions on optimal design of SMR natural gas liquefaction process that used optimization techniques to reliable black-box process simulator are now reviewed. Lee [23] developed a sequential methodology for the systematic synthesis of mixed-refrigerant systems by a combined mathematical programming and thermodynamic approach and applied to the SMR liquefaction process with 1-stage compression and multi-stage expansion. Nogal et al. [24] investigated, using a Genetic Algorithm (GA), the optimal design of mixed refrigerant cycles that included multistage refrigerant compression, 1-to-4-stage expansion, multiple refrigeration cycles, full enforcement of the minimum temperature difference in heat exchangers, simultaneous optimization of variables, and consideration of capital costs. Aspelund et al. [25] tackled the optimal design of SMR process for natural gas liquefaction with 1-stage compression and 1-stage expansion using a hybrid optimization approach of Taboo Search and Nelder-Mead algorithm. Wahl et al. [26] investigated the optimization of a simple 1-stage compression and 1-stage expansion SMR natural gas liquefaction process using a time-efficient Sequential Quadratic Programming routine connected to the process

simulator. Hwang et al. [27] proposed a generic liquefaction superstructure model to express various types of liquefaction cycles, from which 27 feasible configurations were optimized to reduce compression power consumption. Thermodynamic calculations were in sequential modular fashion, using the Peng-Robinson equation of state, and a hybrid optimization framework consisting of GA and SQP was used. Khan and Lee [28] investigated the effectiveness of Particle Swarm Optimization (PSO) in simulation optimization problems in the optimal design of SMR natural gas liquefaction process with 4-stage compression and 1-stage expansion. He et al. [29] proposed and optimized a novel SMR cycle integrated with natural gas liquids recovery process for small-scale LNG plant using a GA. Khan et al. [30] investigated the performance of a sequential coordinate randomization search method for optimizing SMR natural gas liquefaction process. Moein et al. [31] minimized total required work of the commercial APCI-SMR natural gas liquefaction process, which includes a 3-stage compression with phase separation and a 3-stage expansion systems, using a GA. Park et al. [32] optimized the Korea SMR process, which considers a 3-stage compression with phase separation and 2-stage expansion systems and 2-phase expander for LNG stream, using a modified coordinate descent methodology. Austbø and Gundersen [22] minimized the power consumption of a simple 1-stage compression and 1-stage expansion SMR process for natural gas liquefaction using four different constraint formulations to handle the trade-off between investment and operating costs using SQP with multiple starting points.

More recently, Pham et al. [33] developed a knowledge-inspired hybrid optimization of a modified SMR natural gas liquefaction process with 2-stage compression with phase separation and 2-stage expansion without phase mixing targeted for offshore applications. Na et al. [34] analyzed the performance of a modified DIRECT algorithm to optimize an SMR natural gas liquefaction process with 3-stage compression and 2-stage expansion. Pham et al. [35] investigated the energy enhancement of SMR natural gas liquefaction process with 4-stage compression with phase separation and 1-stage expansion using knowledge-based optimization. Qyyum et al. [36] investigated the effect of replacing the throttling valve of Joule-Thomson effect with hydraulic turbine in the energy efficiency enhancement of an SMR natural gas liquefaction process with 4-stage compression and 1-stage expansion. Qyyum et al. [37] analyzed the performance of a hybrid modified coordinate descent algorithm to cope with the optimization of an SMR natural gas liquefaction process with 4-stage compression with phase separation and 1-stage expansion. Ali et al. [38] investigated the performance of a meta-heuristic vortex search algorithm for the optimization of an SMR natural gas liquefaction process with 4-stage compression with phase separation and 1-stage expansion. Ali et al. [39] examined surrogate-assisted modeling and optimization of the SMR natural gas liquefaction process with 4-stage compression and 1-stage expansion. The process optimization was carried out using a surrogate-assisted modeling approach that was optimized using GA and PSO. Lee et al. [40] performed, using a GA, the design and optimization of 3-stage compression with phase separation and 1-stage expansion SMR natural gas liquefaction process with several steady-state operation regimes, depending on the load variation using a GA. He et al. [41] proposed a comprehensive optimization and comparison between 2-stage compression with phase separation

and 2-stage expansion SMR and parallel nitrogen expansion natural gas liquefaction processes from the perspectives of specific energy consumption, exergy efficiency, techno-economy, and operational flexibility using a GA. Qyyum et al. [42] proposed an energy-and-cost-efficient 2-stage expansion SMR process for natural gas liquefaction and compared it to the dual-mixed-refrigerant process. Nikkho et al. [43] optimized two mini-scale modified 3-stage compression with phase separation, 2-stage expansion SMR natural gas liquefaction processes using a GA. Santos et al. [44] investigated the optimization of a 4-stage compression with phase separation, 1-stage expansion SMR natural gas liquefaction process employing an augmented number of decision variables with Nelder-Mead derivative-free optimization method, considering valve and hydraulic turbine expansion. Later the methodology was improved in Santos et al. [45], in which a kriging-assisted global search scheme that included the optimization of the probability of feasible improvement acquisition function to find promising candidates to run the local search with Nelder-Mead algorithm.

Most of the present literature on optimal design of the SMR natural gas liquefaction process have relied on global optimization meta-heuristics, mainly GA, to investigate more sophisticated aspects of the processes, such as energy-efficient, robust, and flexible design, process flow diagram modifications, and economic analysis. Although meta-heuristics are powerful tools for complex optimization problems [46], these methods usually require lots of functions evaluations and lack deterministic convergence proof. Given the present review on methods for black-box optimization problems and single-mixed refrigerant natural gas liquefaction process design and considering the particularities of the simulation optimization problem from this design task toward minimum energy consumption using reliable chemical process simulators, surrogate modeling can be used to introduce symbolic formulation to the optimization problem functions that then can be embedded in mathematical programming setup and solved using classical and efficient gradient-based optimization or deterministic global optimization. Differently from what was done in [39] and [45], which optimized the surrogate optimization problem or acquisition function based on the surrogate models using global optimization meta-heuristics, the present approach explore the mathematical information introduced by the surrogate models. It means that, generic regression models are fitted to data generated from the rigorous simulation and used to replace the black-box functions f and $\mathbf{g}$ by surrogates $\hat{f}$ and $\hat{\boldsymbol{g}}$ that introduce analytical formulation to those functions with reliable derivatives that can be used for efficient gradient-based optimization of the resulted nonlinear programming problem.

The objective of the present paper is to propose an effective utilization of kriging surrogate models to replace the process-simulator-dependent, black-box objective and constraints functions and introduce explicit algebraic formulation to the optimization problem. The proposed framework consists of a three-piece program: the main program in MATLAB that stores the sampled data at the rigorous simulation-dependent functions, fits and updates the kriging models, calls the process simulator for rigorous function evaluations of candidates, and calls GAMS for solving the surrogate optimization nonlinear programming problem; the simulator program in Aspen HYSYS that contains the processes models and calculates the

rigorous functions of the optimization problem; and the algebraic modeling system in GAMS that contains the surrogate optimization problem implemented explicitly, receives the current NLP problem parameters from the main program, and returns the solution to it. One novelty of the present approach to make the liquefaction process simulation optimization effective using surrogate models is to divide the multi-stream heat exchangers, which are modeled like Pinch calculations with phase change and non-ideal solutions, so that the temperature driving force between hot and cold composite curves are calculated for each segment of the heat exchanger instead of the whole. This leads to better behaved functions that are well adjusted by the surrogate models for the sake of optimization.

To test the optimization methodology, the single-mixed refrigerant natural gas liquefaction process design considering 1, 2, and 3 expansion stages is investigated. The decision variables in these simulation optimization problems are the refrigerant component flow rate, the condensation and evaporation pressure, and the expansion temperature of the multi-component refrigerant in the refrigeration cycle. The considered constraint is that a minimum temperature driving force of 3 °C must be assured throughout every multi-stream heat exchanger. Also, phase separation is considered in between compression stages to explore the energy savings from condensation along the refrigeration cycle. The optimization results from the present methodology are compared with two global meta-heuristic optimization approach, which are Particle Swarm Optimization and Genetic Algorithm. The energy efficiency of multi-stage expansion is investigated jointly with a thermodynamic analysis of entropy generation. The computational aspects of the present approach is analyzed with respect to prediction time of the kriging models, and convergence of the proposed algorithm.

This paper is organized so that in Section 2 the kriging model and the surrogate optimization problem are defined and derived. Section 3 describes in detail the single-mixed refrigerant natural gas liquefaction process with 1-to-3-stage expansion system as well as its simulation considerations, constraints, degrees of freedom, and optimization. Section 4 presents the optimization framework that includes sampling, kriging model fitting, and efficient derivative-based optimization of the surrogate problem. Section 5 provides the results of the present optimization approach to the natural gas liquefaction design problem, jointly with an energy and thermodynamic efficiency analysis of multi-stage expansion, a performance comparison with two well-established meta-heuristic for global optimization, Particle Swarm Optimization and Genetic Algorithm, and the computational aspects of the algorithm.

2. Kriging-based constrained optimization

To solve the black-box constrained simulation optimization problems as in Eq. (1), a framework that uses kriging models as surrogates of the objective function f and constraints $\mathbf{g}$ to introduce a simple and efficient algebraic formulation of the black-box functions for efficient gradient-based optimization is presented in this section. The first step of this method is to derive the kriging model of f and $\mathbf{g}$. For simplicity, in Sections 2.1, the kriging model is derived only for f, but the same process can be extended easily for $\mathbf{g}_i,\ i = 1, .., q$. In

Section 2.2 the surrogate optimization problem in algebraic form is derived, which is implemented in GAMS to be solved with state-of-the-art optimization solvers.

2.1. Kriging Model

For mathematical background on the surrogate model used in this surrogate-based optimization framework, the kriging model is derived as the best linear unbiased predictor, Gaussian process regression model following the derivation of Sacks et al. [8], Lophaven et al. [47], Stein [48], and Santos et al. [45].

First, consider the following regression

$$\hat{f}(\mathbf{x}) = \boldsymbol{\beta}^T \boldsymbol{\mathcal{F}}(\mathbf{x}) + z(\mathbf{x}) \tag{2}$$

in which $\boldsymbol{\mathcal{F}} : \mathbb{R}^n \mapsto \mathbb{R}^p$ is a combination of p linear or nonlinear functional forms that approximates f, $\boldsymbol{\beta} \in \mathbb{R}^p$ are the p regression coefficients, and z is the error between the true function and the regression model given by a stochastic function with zero mean and covariance between two points $z(\mathbf{x}^{(i)})$ and $z(\mathbf{x}^{(j)})$ given by

$$\text{cov}\left(z(\mathbf{x}^{(i)}), z(\mathbf{x}^{(j)})\right) = \sigma^2 \mathcal{R}(\boldsymbol{\theta}, \mathbf{x}^{(i)}, \mathbf{x}^{(j)}),$$

where σ^2 is the process variance and $\mathcal{R}(\boldsymbol{\theta}, \boldsymbol{p}, \mathbf{x}^{(i)}, \mathbf{x}^{(j)})$ is the correlation model with parameters $\boldsymbol{\theta} \in \mathbb{R}^n$. Now, assuming some continuity about the function f, one would expect that the correlation between points that are closer to each other is greater than those that are far apart. That notion is translated into the correlation model. There are many correlation models, also known as kernels, that obey this intuition (see [8] for more details), and in this work the Gaussian correlation is used

$$\mathcal{R}(\boldsymbol{\theta}, \mathbf{x}^{(i)}, \mathbf{x}^{(j)}) = \exp\left[-\sum_{h=1}^{n} \boldsymbol{\theta}_h \left(\mathbf{x}_h^{(i)} - \mathbf{x}_h^{(j)}\right)^2\right], \tag{3}$$

where $\boldsymbol{\theta}_h > 0$ is the h^{th} component of the parameters that scales how the correlation between points changes with respect to the h^{th} component of their distance squared.

In the present paper, the regression model chosen is $\boldsymbol{\mathcal{F}}(\mathbf{x}) = 1$, so that $p = 1$ and the kriging predictor is called ordinary kriging. This regression model simplicity usually is enough for good prediction [2] because the behavior of the data is incorporated in the error model $z(\mathbf{x})$. Now, suppose there are m sampled points $\mathbf{X} = [\mathbf{x}^{(1)} \ ... \ \mathbf{x}^{(m)}]^T$, where $\mathbf{x}^{(i)} \in \mathcal{D} \subseteq \mathbb{R}^n, \forall i = 1, ..., m$ is the i^{th} sampled point. And, for all these points, the value of $y^{(i)} \in \mathbb{R} |\ y^{(i)} = f(\mathbf{x}^{(i)}), \forall i = 1, ..., m$ so that $\mathbf{Y} = [y^{(1)} \ ... \ y^{(m)}]^T$ and $\mathbf{g}^{(i)} \in \mathbb{R}^q |\ \mathbf{g}^{(i)} = \mathbf{g}(\mathbf{x}^{(i)}), \forall i = 1, ..., m$ so that $\mathbf{G} = [\mathbf{g}^{(1)} \ ... \ \mathbf{g}^{(m)}]^T$ are available. Then, it is possible to define $\mathbf{R}(\boldsymbol{\theta}) \in \mathbb{R}^{m \times m}$ as the matrix of stochastic-process correlations between z at sampled points, which is $\mathbf{R}_{i,j}(\boldsymbol{\theta}) = \mathcal{R}(\boldsymbol{\theta}, \boldsymbol{p}, \mathbf{x}^{(i)}, \mathbf{x}^{(j)})$, $i, j = 1, ..., m$, and $\mathbf{r}(\mathbf{x}, \boldsymbol{\theta}) \in \mathbb{R}^m$ such that $\mathbf{r}(\mathbf{x}, \boldsymbol{\theta}) = [\mathcal{R}(\boldsymbol{\theta}, \mathbf{x}^{(1)}, \mathbf{x}) \ ... \ \mathcal{R}(\boldsymbol{\theta}, \mathbf{x}^{(m)}, \mathbf{x})]^T$. Finally, deriving the kriging model as the best linear unbiased predictor (see Stein [48], Lophaven et al. [47] or Santos et al. [45] for a complete derivation) in Eq. (2) becomes

$$\hat{f}(\mathbf{x}) = \hat{\beta} + \mathbf{r}(\mathbf{x})^T \mathbf{R}^{-1} (\mathbf{Y} - \mathbf{1}\hat{\beta}), \tag{4}$$

where, $\mathbf{1}$ is a column vector of m entries of ones,

$$\hat{\beta} = \frac{\mathbf{1}^T \mathbf{R}^{-1} \mathbf{Y}}{\mathbf{1}^T \mathbf{R}^{-1} \mathbf{1}}, \tag{5}$$

is the generalized least square solution of the regression coefficient,

$$\hat{\sigma}^2 = \frac{1}{m} (\mathbf{Y} - \mathbf{1}\hat{\beta})^T \mathbf{R}^{-1} (\mathbf{Y} - \mathbf{1}\hat{\beta}), \tag{6}$$

is the process variance, and

$$\hat{s}^2(\mathbf{x}) = \hat{\sigma}^2 \left(1 - \mathbf{r}(\mathbf{x})^T \mathbf{R}^{-1} \mathbf{r}(\mathbf{x}) + \frac{(1 - \mathbf{1}^T \mathbf{R}^{-1} \mathbf{r}(\mathbf{x}))^2}{\mathbf{1}^T \mathbf{R}^{-1} \mathbf{1}} \right), \tag{7}$$

is the expected mean squared error of the predictor.

With Eqs. (4), (5), (6), and (7) it is possible to predict the value of the function f at untried points $\mathbf{x}$ and estimate the prediction error. It remains unknown, however, the parameters $\boldsymbol{\theta}$ of the correlation matrix in Eq. (3). Notice that an approach of minimization of prediction error to determine the model parameters is not possible because kriging as in Eq. (4) interpolates the data for any $\boldsymbol{\theta} > 0$, see Santos et al. [45] for proof to this remark. Then, those parameters are determined by maximum likelihood of the model, *i.e.* maximizing the probability of the data given the model, $p(\hat{f}(\mathbf{x})|\mathbf{x}, \boldsymbol{\theta}, \boldsymbol{p})$. The likelihood is given by

$$\mathcal{L}(\boldsymbol{\theta}, \boldsymbol{p}|\mathbf{X}, \mathbf{Y}) = \frac{1}{(2\pi)^{m/2} (\hat{\sigma}^2)^{m/2} |\mathbf{R}|^{1/2}} \exp \left[\frac{-(\mathbf{Y} - \mathbf{1}\hat{\beta})^T \mathbf{R}^{-1} (\mathbf{Y} - \mathbf{1}\hat{\beta})}{2\hat{\sigma}^2} \right]. \tag{8}$$

Taking the natural log of Eq. (8), substituting $\hat{\beta}$, inverting the sign, removing the constant terms and after some algebra, the maximum likelihood problem becomes

$$\min_{\boldsymbol{\theta}} \quad \psi(\boldsymbol{\theta}) = |\mathbf{R}|^{1/m} \hat{\sigma}^2, \tag{9}$$

in which $|\mathbf{R}|$ is the determinant of $\mathbf{R}$.

2.2. Surrogate Optimization Problem

The surrogate model defined by Eqs. (4) and (5) with $\boldsymbol{\theta}$ parameters given by the solution of the problem in Eq. (9) can be written easily in symbolic formulation to be used explicitly in algebraic modeling language software, such as GAMS, where state-of-the-art global optimization solvers can be used. The kriging model in algebraic formulation becomes

$$\hat{f}(\mathbf{x}) = \hat{\beta} + \sum_{i=1}^{m} \boldsymbol{\alpha}_i \; e^{-\sum_{j=1}^{n} \boldsymbol{\theta}_j (\mathbf{x}_j - \mathbf{X}_{i,j})^2}, \tag{10}$$

where $\hat{\beta}$ is given by Eq. (5), and $\boldsymbol{\alpha} = \mathbf{R}^{-1}(\mathbf{Y} - \mathbf{1}\hat{\beta})$.

Nevertheless, the problem of interest is the constrained optimization in Eq. (1), so that one wants to find a candidate that not only is promising for minimizing f, but also meets the inequality constraints $\mathbf{g}$. For

this reason, the kriging constraints that are $\hat{\boldsymbol{g}}(\mathbf{x}) \leq 0$ has to be satisfied. Thus, the constrained surrogate optimization problem in algebraic form becomes

$$\begin{aligned} \min_{\mathbf{x}\in\mathcal{D}} \quad & \hat{\beta} + \sum_{i=1}^{m} \boldsymbol{\alpha}_i \; e^{-\sum_{j=1}^{n} \boldsymbol{\theta}_j (\mathbf{x}_j - \mathbf{X}_{i,j})^2} \\ s.t. \quad & \hat{\beta}_c^{(c)} + \sum_{i=1}^{m} \boldsymbol{\alpha}_{\boldsymbol{c}_i}^{(c)} \; e^{-\sum_{j=1}^{n} \boldsymbol{\theta}_{\boldsymbol{c}_j}^{(c)} (\mathbf{x}_j - \mathbf{X}_{i,j})^2} \leq 0, \qquad c = 1, ..., q. \end{aligned} \tag{11}$$

It is worth mentioning that for a given set of data $\mathbf{X}$, $\mathbf{Y}$, and $\mathbf{G}$ and trained kriging models for f and $\mathbf{g}$, *i.e.* $\boldsymbol{\theta}$ and $\boldsymbol{\theta_c}$, the parameters $\hat{\beta}$, $\hat{\beta}_c$, $\boldsymbol{\alpha}$, and $\boldsymbol{\alpha_c}$ are constant. Therefore, the proposed kriging-based constrained optimization methodology transforms the black-box constrained simulation optimization problem given in Eq. (1) into two simpler optimization problems that are to adjust the models parameters to the data as in Eq. (9) and optimize the constrained surrogate optimization problem to find a promising candidate as in Eq. (11).

3. Natural gas liquefaction process

The case study and motivating problem for the proposed kriging-based optimization framework is the optimal design of single-mixed refrigerant natural gas liquefaction processes. These processes consist of using a multi-component refrigerant operating in a refrigeration cycle to produce heat sink to cool down and liquefy the natural gas stream. The considered process flow diagram of 1-to-3-stage expansion single-mixed refrigerant natural gas liquefaction is illustrated in Figure 1. The refrigeration cycle includes a 4-stage compression system with intermediate cooling and phase separation for possible condensate. The liquid phase is compressed in the pumps P-1, P-2, P-3, and P-4 and the vapor phase in the compressors K-1, K-2, K-3 and K-4. No phase mixing is considered, which means that the refrigerant heavy condensate is mixed together and goes through the hot pass in the multi-stream heat exchanger separately from the light vapor phase. Then, the streams are mixed back together in the cold pass inside the cryogenic heat exchanger as they vaporize. The third expansion stage is possible thanks to a phase separation of stream 11v, for which the condensed phase is sub-cooled and expanded in valve V-3, whereas the lighter vapor phase is first liquefied then sub-cooled to be expanded in valve V-4.

3.1. Process Simulation

The described single-mixed refrigerant natural gas liquefaction processes is rather difficult to model and simulate mainly because it involves Pinch-like calculations in each multi-stream heat exchangers considering phase change, cryogenic conditions, and non-ideal mixtures. In other words, these heat exchangers are discretized in energy segments, where vapor-liquid equilibrium calculations are performed to determine the temperature of every stream and, therefore, the temperature profiles in these operation units. And, for the sake of process feasibility considering the Second Law of Thermodynamics, the hot streams temperatures

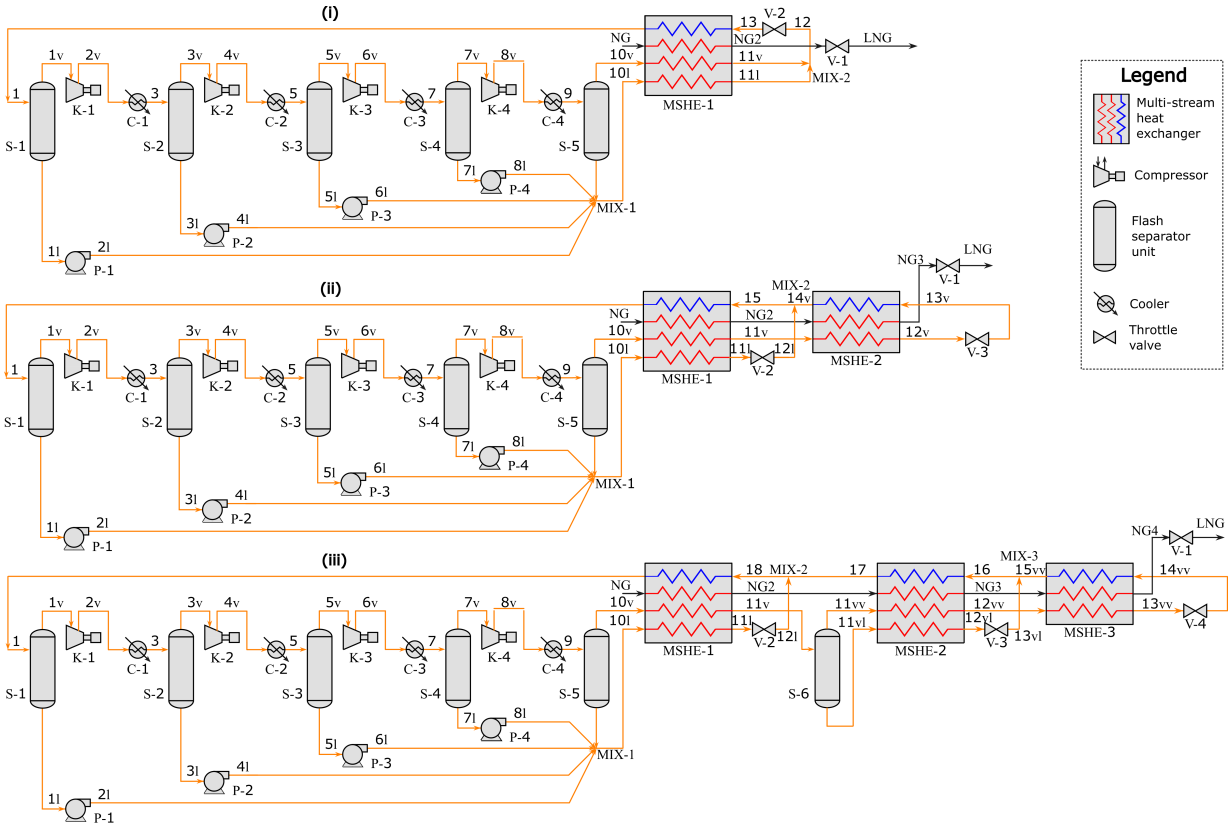


Figure 1: Process flow diagram of single-mixed refrigerant natural gas liquefaction process with 4-stage, phase-separated compression system and 1-to-3-stage expansion system, respectively in (i), (ii), and (iii).

have to be effectively higher than the cold ones throughout the heat exchangers. For rigorous calculations, these processes are modeled and simulated in Aspen HYSYS® V9 using Peng-Robinson equation of state, which is appropriate for hydrocarbons mixtures, such as the natural gas and the refrigerant mixtures.

The natural gas stream NG is considered to be at 8,000 kPa and 32 °C, and its composition is presented in Table 1 as well as other simulation parameters and considerations. A basis of calculation of 1 kg h^{-1} for the natural gas mass flow rate is used. The refrigerant is a mixture of nitrogen, methane, ethane, propane, and i-pentane, and their component mass flow rates are optimization decision variables. The discharge and suction pressures of the multi-stage compression P_{dis} and P_{suc} are decision variables of the optimization problem, and the pressure ratio of each compression stage is given by $(P_{dis}/P_{suc})^{1/4}$, which is the ratio between the discharge and suction pressures divided by 4 stages in series. Notice that the pressure of vapor streams –subscript v– matches the liquid ones –subscript l– from 1 to 8. Finally, the temperatures of every hot stream leaving a multi-stream heat exchanger are considered to be the same to match practical constructions. The expansion temperatures are also degrees of freedom in the simulation and decision variables in the optimization problem, except the temperature of the last expansion, which is fixed to -149.2 °C to guarantee the LNG pressure, molar vapor fraction, and temperature requirements exposed in Table 1.

Table 1: Summary of simulation parameters and considerations, adapted from Pham et al. [35]

Natural gas feed condition	
Property	Condition
Temperature	32 °C
Pressure	8,000 kPa
Flow rate	1.0 kg h^{-1}
Composition	Molar fraction
Nitrogen	0.0022
Methane	0.9133
Ethane	0.0536
Propane	0.0214
i-Butane	0.0046
n-Butane	0.0047
i-Pentane	0.0001
n-Pentane	0.0001
Design parameters and considerations	
Intermediate cooling temperature	40 °C
Intermediate cooling pressure drop	0.0 kPa
LNG molar vapor fraction	8.0 %
LNG temperature	-158.6 °C
LNG pressure	120.0 kPa
Compressor adiabatic efficiency	0.75
Pump adiabatic efficiency	0.75
Thermodynamic property package	Peng-Robinson
MSHE pressure drop (hot stream)	100.0 kPa
MSHE pressure drop (cold stream)	10.0 kPa
Minimum temperature approach	3 °C

One practical constraint in the natural gas liquefaction process is that the minimum temperature approach between hot and cold composite curves throughout the multi-stream heat exchangers must be greater than or equal to 3 °C to avoid Second Law of Thermodynamic violation and impractically big heat exchange area [20]. Another constraint is that all molar vapor fraction in compression inlet streams must be 1, which means that only vapor is allowed in compressors to avoid physical damage. However, this constraint is always assured once flash separators are considered before each compression stage. Even though this adds cost to the process, these separators are considered unpenalized to investigate possible energy savings due to phase separation.

3.2. Process Optimization

Given the process description, considerations, constraints, and degrees of freedom, it is possible to define an optimization problem from which the solution are decisions for the optimal natural gas liquefaction processes. As elaborated in Section 3.1, the decision variables of this optimization are $\mathbf{m}_i$, $i \in REFR$ ={nitrogen (N), methane (C_1), ethane (C_2), propane (C_3), i-pentane (iC_5)} the mass flow rate of component i in the set of refrigerants $REFR$, and P_{suc}, P_{dis}, and $T_{exp_{\ell=1,\dots,Ne-1}}$ the suction and discharge pressures and the mixed-refrigerant temperatures of expansion. The index ℓ refers to the expansion stage, and Ne is the total number of expansion stages. Then, the decision variables for all expansion scenarios are $\mathbf{x} = [\mathbf{m}_{i\in REFR}, P_{suc}, P_{dis}, T_{exp_{\ell=1,\dots,Ne-1}}]$, so that $\mathbf{x} \in \mathbb{R}^n$ is the input vector of decision variables to the simulation and $n = 7 + Ne - 1$ is the dimension of the problem.

Knowing that the work consumption is the most relevant spending in the natural gas liquefaction process, then the design problem is to find $\mathbf{x}^* \in \mathbb{R}^n$ that minimizes the following optimization problem

$$\begin{aligned} \min_{\mathbf{x}\in\mathcal{D}} \quad & f(\mathbf{x}) = \frac{\sum_{p\in PM} W_p(\mathbf{x})}{\dot{m}_{NG}} \\ s.t. \quad & \mathbf{g}_\ell(\mathbf{x}) = 1 - \frac{\min_{k=1,\dots,Nk}\{Th_{\ell,k}(\mathbf{x}) - Tc_{\ell,k}(\mathbf{x})\}}{3} \leq 0, \quad \ell = 1, \dots, Ne \\ & \mathcal{D} = [\mathbf{x}^{lb},\ \mathbf{x}^{ub}], \end{aligned} \tag{12}$$

in which, for a given $\mathbf{x}$, $W_p(\mathbf{x})$ is the work consumption of the pressure manipulator unit p in the set of compressors and pumps PM, $Th_{\ell,k}(\mathbf{x})$ and $Tc_{\ell,k}(\mathbf{x})$ are the temperature of hot and cold composite curves in the ℓ^{th} MSHE at energy segment k, Nk is the number of energy segments in the MSHE composite curves calculations, $\mathcal{D}$ is a box constraint for the decision variables bounded by $\mathbf{x}^{lb}$ and $\mathbf{x}^{ub}$, and $\dot{m}_{NG}$ is the mass flow rate of the natural gas stream. Notice that the value of $W_p(\mathbf{x})$ as well as $Th_{\ell,k}(\mathbf{x})$ and $Tc_{\ell,k}(\mathbf{x})$ are obtained in the black-box chemical process simulator, and so is $f(\mathbf{x})$ and $\mathbf{g}(\mathbf{x})$. Then, $f(\mathbf{x})$ and $\mathbf{g}(\mathbf{x})$ are known only at sampled points and make the optimization problem equivalent to Eq. (1). Table 2 presents the values chosen for lower and upper bounds of $\mathbf{x}$ for scenarios (i), (ii), and (iii) given by $[0.33\mathbf{x}_{base}, 1.66\mathbf{x}_{base}]$, where $\mathbf{x}_{base}$ is a heuristically determined base case. In other words, the bounds are determined to be between $2/3$ below and above the base case. Note also that most of these bounds are selected to diminish the search region toward promising regions, rather than for physical or operational constraint, and it implicates in easier optimization problem and more stable simulations convergence. In addition, the lower bound of m_N is rounded to 0 to consider the case of a heavier multi-component refrigerant, *i.e.* without nitrogen, and the expansion temperatures are adjusted to guarantee decreasing temperature from up to downstream expansion stage.

The kriging model requires some continuity about the black-box function. That may not be the case for $\mathbf{g}$ as reported in Santos et al. [45] because this function is the minimum temperature driving force between the hot and cold composite curves throughout the whole multi-stream heat exchanger, where multi-component streams undergo phase change. For that, we propose to discretize each $\mathbf{g}_\ell$ function in K sections, which

Table 2: Lower and upper bounds on the decision variables for the three scenarios

		1-stage expansion		2-stage expansion		3-stage expansion	
$\mathbf{x}$	$\mathbf{x}_{base}$	$\mathbf{x}^{lb}$	$\mathbf{x}^{ub}$	$\mathbf{x}^{lb}$	$\mathbf{x}^{ub}$	$\mathbf{x}^{lb}$	$\mathbf{x}^{ub}$
m_N [kg·h^{-1}]	0.250	0.000	0.415	0.000	0.415	0.000	0.415
m_{C1} [kg·h^{-1}]	0.600	0.198	0.996	0.198	0.996	0.198	0.996
m_{C2} [kg·h^{-1}]	1.000	0.330	1.660	0.330	1.660	0.330	1.660
m_{C3} [kg·h^{-1}]	1.200	0.396	1.992	0.396	1.992	0.396	1.992
m_{iC5} [kg·h^{-1}]	1.800	0.594	2.988	0.594	2.988	0.594	2.988
P_{suc} [kPa]	250.0	82.50	415.0	82.50	415.0	82.50	415.0
P_{dis} [kPa]	4000	1320	6640	1320	6640	1320	6640
T_{exp_1} [°C]	-50	-	-	-83.00	-16.50	-80.00	-16.50
T_{exp_2} [°C]	-110	-	-	-	-	-130.0	-80.10

makes it more likely to be well behaved functions. Then, the optimization problem becomes

$$\begin{aligned} \min_{\mathbf{x}\in\mathcal{D}} \quad & f(\mathbf{x}) = \frac{\sum_{p\in PM} W_p(\mathbf{x})}{\dot{m}_{NG}} \\ s.t. \quad & \mathbf{g}_\kappa(\mathbf{x}) = 1 - \frac{\min_{k\in\Omega_\kappa}\{Th_{\ell,k}(\mathbf{x}) - Tc_{\ell,k}(\mathbf{x})\}}{3} \leq 0, \quad \kappa = 1, ..., K \times Ne \\ & \mathcal{D} = [\mathbf{x}^{lb},\ \mathbf{x}^{ub}], \end{aligned} \tag{13}$$

in which κ is the set of $K \times Ne$ divisions in $\mathbf{g}$, and Ω_κ is the set of the Nk points from composite curves calculations that belongs to section κ. Also, the number of constraints increases from Ne to $K \times Ne$ in the proposed optimization formulation.

4. Optimization Framework

A framework is proposed in this paper to solve black-box constrained optimization problems as in Eq. (1), which is tested in the energy-efficient optimal design of single-mixed refrigerant natural gas liquefaction processes with 1-to-3 expansion stages. This approach uses kriging surrogate models as presented in Section 2.1 to substitute the black-box objective function and constraints and introduce algebraic formulation to the black-box problem as presented in Section 2.2. The surrogate optimization model is, then, implemented in GAMS, where it is solved with state-of-the-art, derivative-based, global solvers. Figure 2 illustrates how the optimization framework operates.

Before anything in this optimization framework, MATLAB and Aspen HYSYS are connected via object linking and embedding technology using the MATLAB built-in function "actxserver" to create in its environment a component object model (COM) server of the HYSYS application with the process simulation

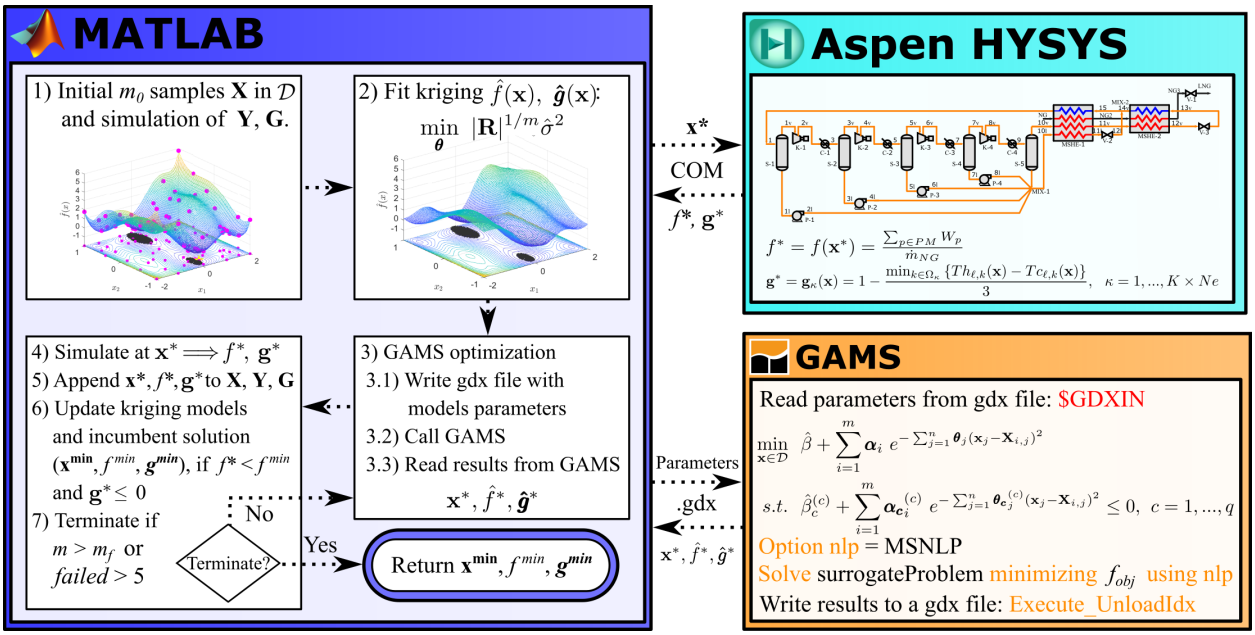


Figure 2: Algorithmic building blocks of present kriging-based optimization framework.

variables and methods. In other words, the simulation objects like streams, operations, and solver become exposed to the programming environment so that each $\mathbf{x}^*$ generated in the MATLAB is sent to the simulation by setting their values in the respective objects features (mass flow rate, temperature, pressure). Then, after converging the simulation, the objects features that contain the values to calculate the objective and constraints functions are accessed in the programming environment to compute f^* and $\mathbf{g}^*$.

Then, the first step of this approach is to make an initial sample in $\mathcal{D}$ to adjust the surrogate models. For that, Latin Hypercube algorithm is used to maximize the minimum distance between points and fill in m_0 points in the design space. For each sample point in $\mathbf{X}$, the rigorous simulation is performed to calculate $\mathbf{Y}$ and $\mathbf{G}$. Also, the simulation evaluations counter m receives m_0, $k \leftarrow 0$.

Given the initial data $\mathbf{X}$, $\mathbf{Y}$, and $\mathbf{G}$, the kriging models are adjusted for f and $\mathbf{g}$ as in Eq. (10) to maximize the likelihood of the model given the data. The maximum likelihood optimization problem in Eq. (9) is solved with the MATLAB implementation of a interior-point method in the built-in function `fmincon` to determine the $\boldsymbol{\theta}$ and $\boldsymbol{\theta_c}$ parameters of the kriging models. The initial guess for the algorithm is $\boldsymbol{\theta}_h = 1$ for $h = 1, ..., n$. Notice that these parameters are not readjusted, but in between iterations the kriging models are updated with the same $\boldsymbol{\theta}$ to the current $\mathbf{X}$, $\mathbf{Y}$, and $\mathbf{G}$, unless log-likelihood function $\psi(\boldsymbol{\theta})$ becomes at least 10 times greater than the value obtained in the adjusting parameter optimization problem in Eq. (9). In other words, if the model with current parameters is not likely to have generated the available data, a new parameter adjustment will be performed.

Given the kriging models jointly with the maximum likelihood $\boldsymbol{\theta}$ parameters, the surrogate optimization problem as in Eq. (11) is solved in GAMS, where the algebraic kriging models are implemented explicitly.

To do that, the programming environment has to communicate with the algebraic modeling language system to send the model parameters that change every iteration ($\hat{\beta}$, $\hat{\beta}_c$, $\boldsymbol{\alpha}$, $\boldsymbol{\alpha_c}$, $\mathbf{X}$, and possibly $\boldsymbol{\theta}$ and $\boldsymbol{\theta_c}$). This communication is performed via GDX (GAMS Data eXchange) files that provide an interface to read and write values of GAMS symbols such as sets, parameters, variables, and equations. Then, the GAMS program, which has the surrogate optimization problem implemented, is run from MATLAB to solve the NLP problem and to write the solution results ($\mathbf{x}^*, \hat{f}^*, \hat{\boldsymbol{g}}^*$) also in GDX files that are read in MATLAB. Any global solver that deals with nonconvex NLP problems can be used for this optimization problem. However, there is no point to spent too much resource and time on global NLP solvers with provable global optimality, such as Baron [49], because the solution of the surrogate optimization problem in Eq. (11) is an approximation of the true black-box constrained optimization problem in Eq. (1). Therefore, the multi-start NLP solver MSNLP from Ugray et al. [50] with CONOPT [51] solver for local search is selected, and the reasoning behind this choice is that it is fast to converge to (at least) a good local minimizer. The starting points in the multi-start approach are generated using a normal probability distribution from an initial coarse search to define a promising region within which random starting points are concentrated.

After solving the surrogate optimization problem, the rigorous simulation is performed at the solution returned to the programming environment from GAMS $\mathbf{x}^*$ to calculate f^* and $\mathbf{g}^*$, and the simulation evaluations counter m is iterated. Then, the current values of $\mathbf{x}^*$, f^*, and $\mathbf{g}^*$ are appended to the data $\mathbf{X}$, $\mathbf{Y}$, and $\mathbf{G}$, and the incumbent solution ($\boldsymbol{x^{min}}, f^{min}, \boldsymbol{g^{min}}$) is updated if the solution is improved ($f^* < f^{min}$ and $\mathbf{g}^* \leq 0$). From the extended data, the kriging models parameters $\hat{\beta}$, $\hat{\beta}_c$, $\boldsymbol{\alpha}$, and $\boldsymbol{\alpha_c}$ are updated and the algorithm iterates until the maximum number of sampled points m_f is achieved or if the surrogate optimization fails ($f^* > f^{min}$ even for infeasible $\mathbf{g}^* > 0$) five times to provide a promising candidate to solve the true black-box optimization problem.

5. Results

In this section, the results from the kriging-based optimization framework proposed in Section 4 applied to the optimal design of single-mixed refrigerant natural gas liquefaction processes with 1, 2, and 3 expansion stages are reported. The considered parameters of the optimization approach are initial sample size $m_0 = 10n$, which is a classical number for the kriging surrogate model [9], maximum number of samples (function evaluation budget) $m_f = 20n$, number of decision variables $n = 7 + Ne - 1$, number of sections into which each multi-stream heat exchanger is divided $K = 10$. The box-constrained design space $\mathcal{D}$ is given by the lower and upper bounds on the decision variables at Table 2, and the computer program to compute f and $\mathbf{g}$ is the natural gas liquefaction processes simulations in Aspen HYSYS. The computer used to run this framework has a Intel Core i7-9750H processor with 16 GB of RAM.

Table 3 presents the best results of decision variables, and objective function from five optimization runs for each of the three scenarios that accounts for the only randomness present in the proposed framework, which is the initial m_0 samples of $\mathbf{X}$. The best net work consumption found for these processes are 0.2571, 0.2262, and 0.2193 kW per kilogram of natural gas being liquefied per hour, respectively for scenarios (i), (ii), and (iii). It represents a specific net work consumption of 925.5, 814.2, and 789.3 kJ per kilogram of natural gas. These quantitative results show the effectiveness of the optimization framework to find energy-effective alternatives to the single-mixed refrigerant natural gas liquefaction process.

From the optimization results in Table 3, it is possible to conclude that the single-mixed refrigerant natural gas liquefaction process benefits from more expansion stages from the energy consumption point of view. Not only the net work consumption decreased with the number of expansion stages, but also the total expected size of the multi-stream heat exchangers (UA) decreased from 657.7 to 550.0 and 543.0 kJ/(°C h) and their total heat duty from 2912 to 2545 and 2406 kJ/h. In other words, the inclusion of more expansion stages in the designed liquefaction processes diminished the work consumption, therefore the electricity and compressors size, and diminished the multi-stream heat exchangers with respect to total heat transfer and expected area. These results are both factors of cost-effective as well as energy-efficient process. The main reason behind this improvement is the improved thermodynamic efficiency of the process with more expansion stages. Table 4 presents the results of entropy generation in each operating unit. The reader is invited to see Santos et al. [45] or Smith et al. [52] for more details on the calculation of entropy generation in this analysis.

In fact, the total entropy generation is significantly smaller with more expansion stages, from 0.5634 to 0.4377 W/°C for scenario (i) and (iii). The most relevant thermodynamic efficiency gain takes place in the compressors, coolers, multi-stream heat exchangers, and mixers. Therefore, the additional expansion stages with phase separation enable formation of intermediary mixed-refrigerant with different composition in each multi-stream heat exchanger and guarantee a better match between composite curves, as presented in Figure 3, more efficient inter-cooled compression system, and smoother mixing processes. The phase separation in

Table 3: Optimization results for single-mixed refrigerant natural gas liquefaction with 1, 2, and 3 stages of expansion

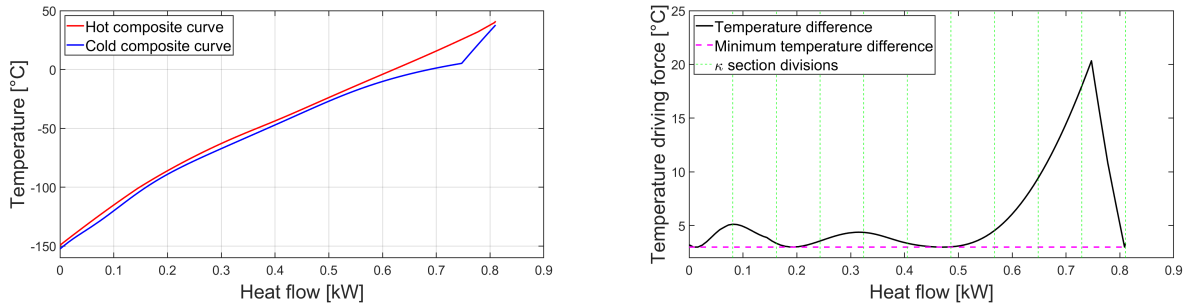
Decision variables	Scenario (i)	Scenario (ii)	Scenario (iii)
m_N [kg·h^{-1}]	0.1429	0.1582	4.633E-2
m_{C1} [kg·h^{-1}]	0.4167	0.4426	0.4512
m_{C2} [kg·h^{-1}]	0.8586	0.9533	1.004
m_{C3} [kg·h^{-1}]	0.8173	1.198	0.8333
m_{iC5} [kg·h^{-1}]	1.805	1.735	1.609
P_{suc} [kPa]	350.3	282.8	302.2
P_{dis} [kPa]	2944	2660	3654
T_{exp_1} [°C]	-	-24.31	-30.08
T_{exp_2} [°C]	-		-120.2
Optimization results			
Net work consumption $\left[\frac{\text{kJ}}{\text{kg NG}}\right]$	925.5	814.2	789.3
MSHE-1 minimum temperature approach [°C]	3.000	3.003	3.010
MSHE-2 minimum temperature approach [°C]	-	3.003	3.008
MSHE-3 minimum temperature approach [°C]	-	-	3.010
MSHE-1 expected area (UA) $\left[\frac{\text{kJ}}{\text{°C·h}}\right]$	657.7	223.2	286.1
MSHE-2 expected area (UA) $\left[\frac{\text{kJ}}{\text{°C·h}}\right]$	-	326.8	231.8
MSHE-3 expected area (UA) $\left[\frac{\text{kJ}}{\text{°C·h}}\right]$	-	-	25.07
MSHE-1 heat duty [kJ/h]	2912	1315	1271
MSHE-2 heat duty [kJ/h]	-	1230	1007
MSHE-3 heat duty [kJ/h]	-	-	127.7

between compression stages appeared only in the last two stages in accordance with previous works [45]. It is because the multi-component refrigerant is too light to condense at lower pressure levels, even though the optimal values of decision variables in Table 3 show preference to heavier compositions of mixed-refrigerant. In scenario (i), 9.34 % of the mass flow rate is compressed in pump P-3 and 27.82 % in P-4. In scenario (ii), 11.51 % of the mass flow rate is compressed in P-3 and 21.08 % in the P-4. In scenario (iii), 20.56 % of the mixed-refrigerant mass flow rate is compressed in P-3 and 18.19 % in the P-4. These results are conclusive to show the importance of phase separation in the energy saving in the compressors K-3 and K-4 as 32.59 up to 38.75 % of the mixed refrigerant flow rate is removed from compressors system.

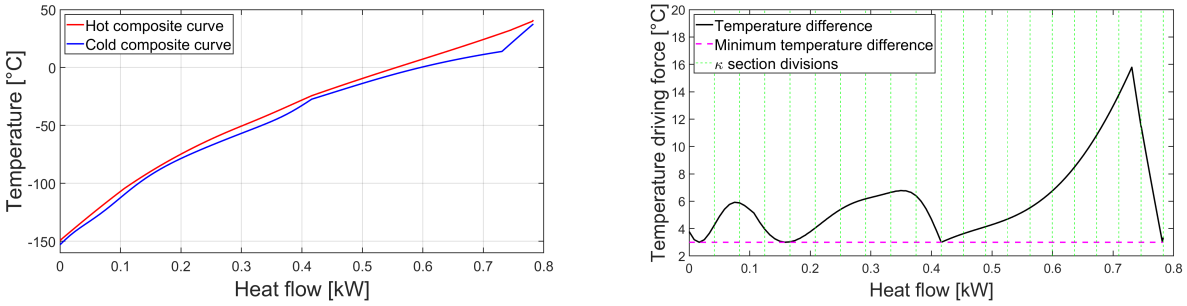
Optimization comparison. Now, in order to evaluate the performance of the proposed optimization methodology applied to the optimal design of SMR natural gas liquefaction process, its results are compared to the ones from the well-established global optimization meta-heuristics of Particle Swarm Optimization and

Table 4: Entropy generation analysis

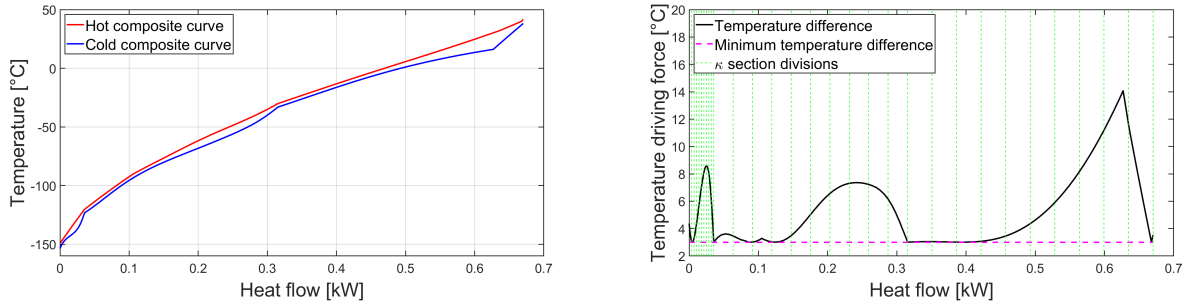
Op. Name	Scenario (i)		Scenario (ii)		Scenario (iii)	
	Sgen [W/°C]	%	Sgen [W/°C]	%	Sgen [W/°C]	%
K-1	5.21E-02	9.25	4.66E-02	10.13	4.60E-02	10.51
K-2	5.09E-02	9.03	4.53E-02	9.84	4.44E-02	10.14
K-3	4.58E-02	8.13	4.04E-02	8.78	3.71E-02	8.48
K-4	3.51E-02	6.23	3.27E-02	7.11	2.98E-02	6.81
C-1	2.15E-02	3.82	1.69E-02	3.67	1.88E-02	4.30
C-2	3.03E-02	5.38	2.81E-02	6.11	3.63E-02	8.29
C-3	5.02E-02	8.91	3.89E-02	8.45	3.54E-02	8.09
C-4	3.49E-02	6.19	2.99E-02	6.50	2.93E-02	6.69
MSHE-1	1.06E-01	18.87	4.10E-02	8.91	2.93E-02	6.69
MSHE-2	-	-	6.14E-02	13.34	3.52E-02	8.04
MSHE-3	-	-	-	-	1.14E-02	2.60
V-1	3.84E-02	6.82	3.84E-02	8.35	3.89E-02	8.89
V-2	4.43E-02	7.86	1.04E-02	2.26	1.47E-02	3.36
V-3	-	-	2.53E-02	5.50	1.61E-02	3.68
V-4	-	-	-	-	6.10E-03	1.39
S-1	-	-	-	-	-	-
S-2	-	-	-	-	-	-
S-3	0.00E+00	0.00	0.00E+00	0.00	0.00E+00	0.00
S-4	0.00E+00	0.00	0.00E+00	0.00	0.00E+00	0.00
S-5	0.00E+00	0.00	0.00E+00	0.00	0.00E+00	0.00
S-6	-	-	-	-	0.00E+00	0.00
P-1	-	-	-	-	-	-
P-2	-	-	-	-	-	-
P-3	4.70E-04	0.08	5.25E-04	0.11	1.20E-03	0.27
P-4	9.72E-04	0.17	6.36E-04	0.14	7.31E-04	0.17
MIX-1	3.50E-03	0.62	3.20E-03	0.70	4.80E-03	1.10
MIX-2	4.87E-02	8.64	4.81E-04	0.10	6.80E-04	0.16
MIX-3	-	-	-	-	1.50E-03	0.34
TOTAL	**0.5634**	**100.0**	**0.4601**	**100.0**	**0.4377**	**100.0**



(a) Scenario (i).



(b) Scenario (ii).



(c) Scenario (iii).

Figure 3: Temperature profiles in the multi-stream heat exchangers

Genetic algorithm with the same budget of simulation evaluations, as presented in Table 5. The parameters were set as default of the MATLAB implementation of these algorithms and with n number of particles, or individuals, and 20 iterations. To deal with the constraints of the optimization problem in Eq. (13) a simple barrier penalization is applied proportional to the constraint violation multiplied by a penalization parameter equal to 1000, as in [45], to guarantee feasible solutions. Knowing that these methods depend on randomness, each algorithm was applied 5 times for each simulation.

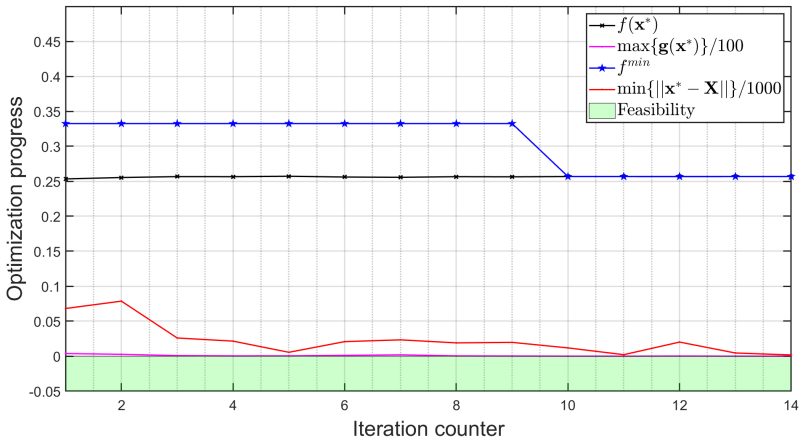
From the results in Table 5 it is possible to infer that the present methodology is more efficient (lower best result of objective function) and more consistent (lower mean value and standard deviation of objective function) for all three scenarios of the single-mixed refrigerant natural gas liquefaction process design. The energy savings of the results from the present optimization method in comparison with the ones from Particle

Table 5: Comparison between present optimization framework with PSO and GA

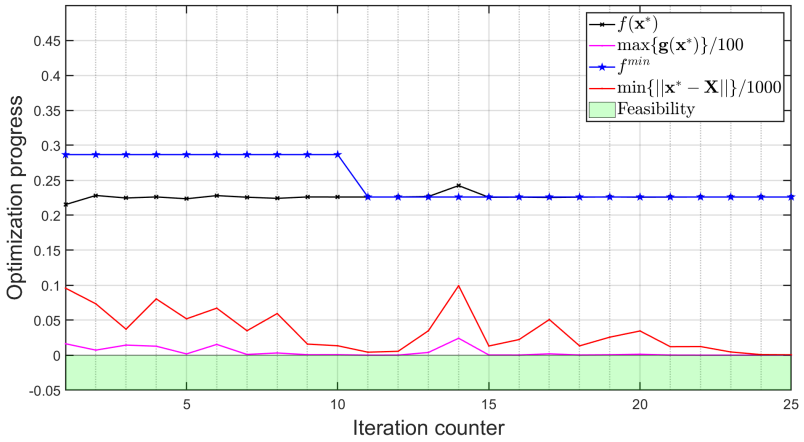
Obj. function / Opt. Run	1-stage-expansion SMR			2-stage-expansion SMR			3-stage-expansion SMR		
	Present	PSO	GA	Present	PSO	GA	Present	PSO	GA
Run 1	0.2571	0.2920	0.2954	0.2262	0.2998	0.3102	0.2198	0.3219	0.3403
Run 2	0.2572	0.3192	0.3926	0.2262	0.3018	0.2954	0.2200	0.3166	0.3361
Run 3	0.2571	0.3312	0.3449	0.2262	0.2836	0.2737	0.2203	0.2984	0.3760
Run 4	0.2571	0.3339	0.4166	0.2272	0.2903	0.4157	0.2197	0.2927	0.3665
Run 5	0.2571	0.3167	0.3300	0.2262	0.2907	0.3080	0.2193	0.2761	0.3443
Results analysis									
Best result	0.2571	0.2920	0.2954	0.2262	0.2836	0.2737	0.2193	0.2761	0.3361
Mean result	0.2571	0.3186	0.3559	0.2264	0.2932	0.3206	0.2198	0.3011	0.3526
Standard deviation	5.49E-05	1.49E-02	4.36E-02	3.89E-04	6.68E-03	4.93E-02	3.46E-04	1.66E-02	1.57E-02
Savings (%)	-	12.02	13.03	-	20.29	17.39	-	20.48	34.69
Execution time (min)	4.22	3.35	4.296	6.25	5.90	5.99	14.05	9.42	9.626

Swarm Optimization and Genetic Algorithm are significant, from 12.02 to 34.69 %.

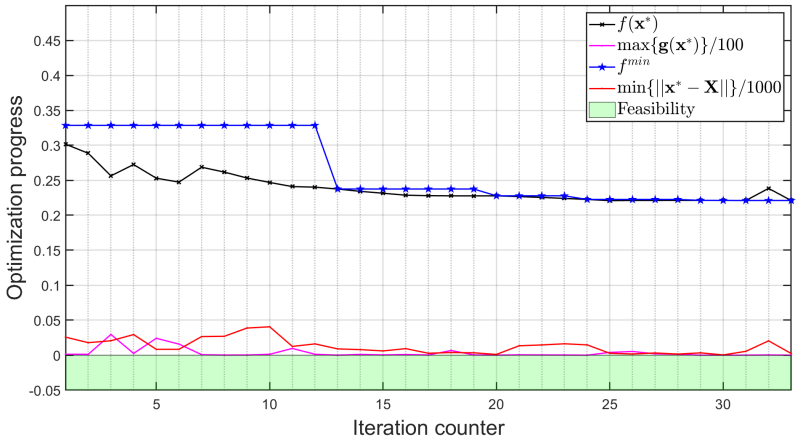
Even though the present methodology involves additional steps other than function evaluation and determination of next iteration steps, like kriging model fitting and surrogate problem optimization, the mean execution times are competitive with Particle Swarm Optimization and Genetic Algorithm. The first reason for that is due to the computation effort to predict value of f and $\mathbf{g}$ in the kriging surrogate model being way smaller than the simulation. For instance, the mean elapsed time of computing f and $\mathbf{g}$ at the $10n$ initial points goes from 1.4 to 3.1 seconds, depending on the simulation, whereas for $\hat{f}$ and $\hat{\boldsymbol{g}}$ it is in the order of magnitude of 10^{-4} seconds. This allows fast solution time of the surrogate problem in GAMS using multi-start gradient-based local search. The other reason is the convergence of the present approach, illustrated in Figure 4. This convergence is indicated by the objective function value at iterates $f(\mathbf{x}^*)$ (black curve) converging to the incumbent solution f^{min} (blue curve), and the minimum distance of the iterate $\mathbf{x}^*$ to the previous data $\mathbf{X}$ (red line) converging to zero. The optimization progress of the present method is fast because the kriging models capture the behavior of the black-box functions, specially in promising regions (feasible and low values of f). This happens as the sampling comes from the surrogate optimization solution, and is concentrates in these promising regions. One can think of this as an active learning process, where the surrogate models not only learn the black-box functions behavior but also choose where to learn more.



(a) Scenario (i).



(b) Scenario (ii).



(c) Scenario (iii).

Figure 4: Optimization progress of the proposed method for each considered simulation-optimization problem.

6. Conclusions

This paper presented an optimization framework to solve the constrained black-box simulation optimization problem that arises from the optimal energy-efficient design of the single-mixed refrigerant natural gas liquefaction process with 1, 2, and 3 expansion stages, using reliable process simulator. In this approach, the kriging model is adjusted to data generated from the process-simulator-dependent, black-box functions of the simulation optimization problem to introduce simple, computationally inexpensive, and effective algebraic formulations to the black-box objective and constraint functions. The constrained surrogate optimization problem is solved in GAMS using state-of-the-art efficient gradient-based multi-start local optimization with CONOPT local solver to determine a candidate of decision variables for which the true functions are calculated in the rigorous simulation. The surrogate problem optimization guides the sampling towards learning the rigorous functions near promising regions for solving the original simulation optimization problem.

This optimization framework was applied to the natural gas liquefaction design using the process simulator Aspen HYSYS for rigorous simulations, MATLAB for the main program that handles with linking, data storage, and model fitting, and GAMS for implementing and solving the NLP surrogate optimization problems. The best net work consumption found for these processes are 0.2571, 0.2262, and 0.2193 kW per kilogram of natural gas being liquefied per hour, respectively for scenarios (i), (ii), and (iii), and the total expected size of the multi-stream heat exchangers (UA) decreased from 657.7 to 550.0 and 543.0 kJ/(°C h) and their total heat duty from 2912 to 2545 and 2406 kJ/h. In other words, the inclusion of more expansion stages made the designed liquefaction processes more cost-effective as well as energy-efficient, mainly because of the thermodynamic efficiency of the process.

From comparing the present approach to global optimization meta-heuristics of Particle Swarm Optimization and Genetic Algorithm, it is evident that, for the same budget of simulation evaluations, the present approach is more efficient and more consistent with significant numerical improvement of 12.02 to 34.69 % of energy savings. The main reasons for the better efficiency is the low computation effort to predict the functions f and $\mathbf{g}$ using the kriging surrogate model (from 1.4 to 3.1 seconds in the simulation to the order of magnitude of 10^{-4} seconds for $\hat{f}$ and $\hat{\boldsymbol{g}}$) that allows fast execution time of the surrogate optimization problem in GAMS. The other reason is the convergence of the present approach, which is relatively fast because the kriging models capture the behavior of the black-box functions, specially in promising regions, and uses it efficiently for optimization.

Although it was tested only for single-mixed refrigerant natural gas liquefaction process design, the proposed optimization approach is suitable for any constrained black-box simulation optimization problem, once it provides computationally cheap-to-evaluate surrogate models (kriging) of the objective and constraints functions with simple symbolic formulation that can be embedded to tradition NLP problems.

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