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From droplets to process: Multilevel research approach to reduce emissions from LNG processes

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Abstract

In the Enabling Low-Emission LNG Systems project, SINTEF Energy Research and NTNU have since 2009 been developing knowledge and tools with the aim to enable environmental safe, cost-effective, and energy efficient LNG processes. The project has employed a multilevel approach, where studies of fundamental phenomena, component modeling, and process optimization have been performed in parallel. Detailed models for two-phase flow in heat exchangers (HXs) have been developed, and falling films and droplet behavior relevant for LNG HXs have been studied experimentally. Further, the project work has generated robust and accurate HX models that can describe instabilities and include geometrical considerations. Finally, the project has improved the basis for optimization of LNG process through investigating of optimization tools and careful analysis and formulation of the optimization problem.

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Keywords: LNG, optimization, Heat exchanger modeling, Fluid Dynamics, Thermodynamics, Multi-phase flow

Nomenclature

LNG	Liquefied natural gas
LELNG	Enabling Low-Emission LNG Project
HX	Heat exchanger

1. Introduction

Natural gas will play an important role in the future energy system with its lover carbon footprint compared with other fossil fuels. By liquefying the gas the volume is reduced around 600 times. Liquefied natural gas (LNG) is therefore an energy-efficient and dominating solution for ship transportation over large distances. During the last decade, LNG penetration in the energy market has increased tremendously, and,

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in the IEA 450 (2°C) scenario, the use of natural gas will increase significantly in decades ahead [1]. Due to increased natural gas demands, and current large regional differences in gas prices, the use of LNG is expected to continue to increase rapidly in the years to come, and a large number of LNG carriers are under order [1].

Although LNG is an environment-friendly energy source, the liquefaction process is still costly in terms of investments required and energy consumption. Therefore, SINTEF Energy Research and NTNU are running a research project aiming to enable energy efficient and, hence, environment-friendly, LNG production. This project, called "Enabling Low-Emission LNG Systems" (LELNG), is funded by the PETROMAKS2 program of the Research Council of Norway, Statoil, and GDF SUEZ. The duration of the project is from the fall of 2009 to 2014.

In LELNG, the challenges related to developing and operating energy-efficient LNG plants are addressed on three levels, each with a dedicated subproject (SPs), as illustrated in Figure 1. On the most fundamental level, detailed phenomena of two-phase flow in LNG process plants and heat exchangers (HXs) are studied experimentally and theoretically. The heat exchangers are very important units of natural gas liquefaction processes, both in terms of investment cost, operational robustness and energy robustness of the process, necessitating a thorough understanding. Currently, however, the detailed behavior with regards to flow and heat transfer is not fully understood. In the project, droplet-film interactions have been experimentally investigated and the results are represented in new models. Further, flowing film patterns over tube bundles found in spiral wound heat exchangers are studied experimentally, and heat transfer has been modelled. As illustrated in Figure 1, this work will aid the development of flow regime maps for heat exchanger modeling. On the second level, a framework for HX models has been developed. The framework enables the inclusion of HX geometries and physics, and can and has been be adapted to different types of relevant HX designs. Unlike composite curve models that are often employed, this framework hence allows for rigorous inclusion of size and weight constraints, as well as modeling of undesirable phenomena like the Ledinegg instability and mal-distribution. Using this model during HX design and process design could reduce risks related to operational challenges and reduce energy consumption within realistic physical constraints. The model is so efficient and robust that it has been used for process optimization, leading us to the third level of detail in LELNG, the overall LNG process evaluation. LNG has been produced for the last 50 years and the plants worldwide utilize a large variety of processes. However, optimizing the LNG process is challenging due a large number of variables and non-convexity of the objective function and the constraints. In LELNG, the optimization problem has been characterized and different formulations and optimization techniques have been investigated. Both in the studies and modeling of heat exchangers and optimization of LNG processes, the project has had a generic focus. Hence, the focus has been on the development of tools and competence adapted to and tested on a range of different general process and heat exchanger designs rather than the study of specific commercial solutions.

Two PhD candidates have been educated in this project, looking theoretically at detailed flow phenomena found in HXs and HX network models and instabilities, respectively. A third PhD candidate will soon complete his studies in LNG process optimization.

2. Two-phase flow phenomena in LNG processes

As discussed in the introduction, the purpose of the work on two-phase flow phenomena has been to gain insight into fundamental phenomena occurring in heat exchangers in liquefaction plants. A basic hypothesis of the work in two-phase flow phenomena in LNG processes has been that a thorough understanding of the processes and phenomena occurring at a small-scale in the heat exchanger is necessary to obtain an improved understanding of the heat exchanger, its design, and operation. For instance, a very important quantity to determine in heat exchangers is the local heat transfer. In for in spiral wound heat exchanger, this heat transfer will be dependent on the thickness of the film, which again is related to droplet-film interaction and shear flow patterns. In this projects, these phenomena has been studied extensively both through experiments and modeling.

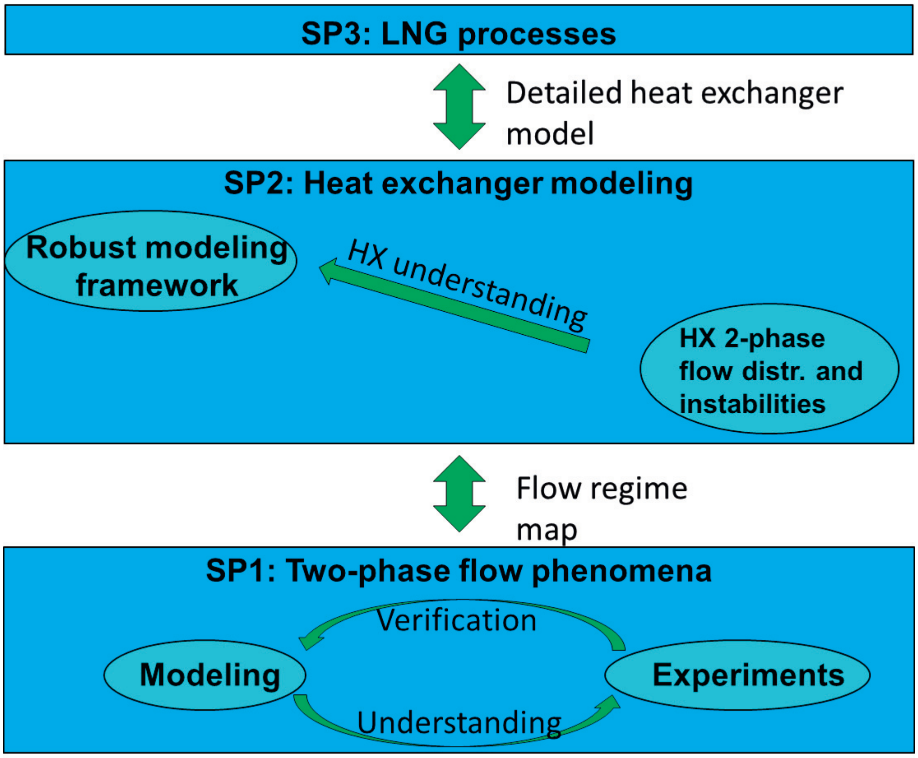


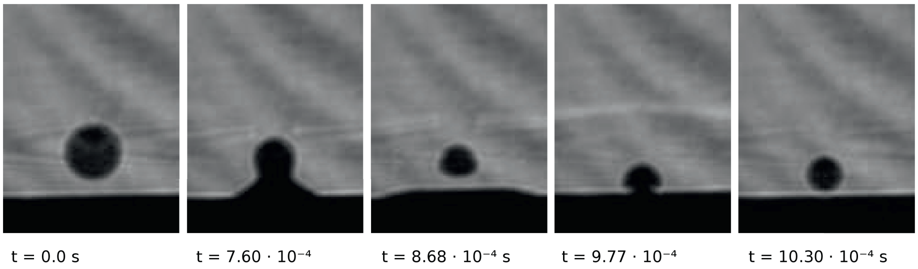
Fig. 1: Project structure of LELNG showing interconnections between the different activities

2.1. *Experimental study of detailed flow phenomena*

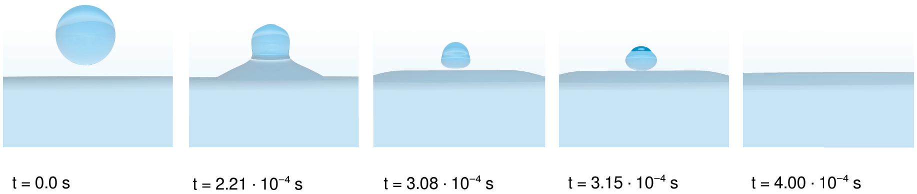
In the project, quite extensive work has been performed in order to understand the droplet behavior relevant to LNG heat exchangers. For maximum heat transfer coefficient leading to efficient and compact heat exchangers, the working liquid and the walls of the heat exchanger should be in contact, and hence the fraction of splashing droplets in the gas phase should be limited. Hence, the regimes for droplet-film coalescence, bouncing, and jetting should be identified in terms of parameters such as droplet size, velocity, and angle for conditions similar to what is found in heat exchangers. Although impingement studies of droplets on either a dry or a liquid-covered surface has been investigated over the past hundred years, most of these studies have been performed on non-moving targets and using water. However, the properties of water is significantly different from mixed refrigerants in LNG plants, in particular the surface tension and viscosity is much higher for the former.

In our work, we have progressed systematically to conditions that more realistically simulate heat exchangers, going from droplets falling on deep pools [2, 3, 4] to studies of droplets on flowing films at various angles [5]. The latter study showed that the threshold between coalescence and splashing of a flowing film can be well-characterized by using a modification of a model suggested by Cossali et. al. [6] meant for normal impacts. Hence, based on this result a systematic work with different horizontal film thicknesses has since been performed. n-pentane has been used as a model fluid, because of its quite similar properties to mixed refrigerants.

Lately the focus has been on characterization of shear flow on tube bundles found in spiral wound heat exchangers. Transitions between flow different flow regimes like droplet, column, and sheet flow, and hysteresis effects have been identified using n-pentane, and the transitions seem to differ from those of water when using common dimensionless empirical models [7]. The current experimental focus in the project is to measure film thickness on tube bundles with cross flow.



(a) Experimental result



(b) Simulation result

Fig. 2: Experimental results (top) and simulation results (bottom) for a 0.18 mm water droplet falling through air and impacting a deep pool of water at 0.29 m/s. Reprinted from [10] with permission from Elsevier.

2.2. Modeling

Two-phase flow modeling of the detailed phenomena is needed in order to understand the experimental results as well as to be able to generalize them. Relevant and carefully designed simulations may provide results for variable ranges that are difficult to test experimentally. Conversely, experiments are important for verification and further development of physical models.

The two-phase modeling work is based on an in-house code that solves the Navier-Stokes equations for two-phase immiscible and incompressible flow. The position of the interface between the phases is captured with the level-set method [8], and the ghost-fluid method [9] is used to handle interface discontinuities. For details of the model, solution techniques, and implementation, see [10].

Detailed modeling of the small-scale flow phenomena occurring in HXs still pose several challenges. One particular challenge that was met during the present project was to develop a robust and accurate method for calculating interface curvature of small droplets [10, 11, 12, 13]. More specifically, the new method improves robustness during topology changes, for instance when a droplet collides with a film to form a single body of liquid.

Comparisons between numerical and experimental results were performed for droplets falling onto flat films [10, 13]. Figure 2 shows a comparison between experimental results and simulation results for a water droplet falling through air and impacting on a deep water pool. The numerical results qualitatively capture the partial coalescence. However, the bouncing phenomenon in the final frame of the experimental results is not captured. In order to capture the bouncing phenomenon, one must either resolve extremely small length scales or employ numerical methods that prevent coalescence under certain conditions. Of these, the latter seems like the simplest approach and is a natural path for future work.

Recently, the two-phase flow model has been extended to support heat transport and mass transfer between phases due to vaporization and condensation. The model was verified against analytical solutions, and results for a vaporizing liquid methane droplet are shown to be consistent with qualitative expectations [14]. The model was also applied for simulating a lava lamp, and the results were found to be in agreement with experiments [15].

One goal in LELNG was to enable simulations of relevant flow phenomena inside complex geometries. This is needed for simulating the experiments of flow across tube bundles. To this end, a diffuse domain approach (DDA) with higher order correction terms was developed and analyzed [16]. The DDA allows complex geometries to be embedded in the governing equations, and may therefore be used for simulating flows inside and through complex geometries. The latter remains as a future task.

3. Heat exchanger modeling

In the LELNG project, a flexible heat exchanger modeling framework has been used, further developed, and adapted for simulation and optimization of LNG heat exchangers. The modeling framework makes it possible to create complex heat exchangers based on configuration of simple low level objects.

The objective of the modeling framework is that heat exchangers can be modeled with different level of details. In this project heat exchanger models are a link between fundamental laboratory measurements on flow and heat transfer phenomenas and process simulation using geometrically described heat exchangers. For a heat exchanger model to be useful for optimization purposes, it is necessary the this model is very robust and reasonably fast. The modeling framework are described by [17]. A short description of the hierarchy and modules is following.

3.1. Description of the modeling framework

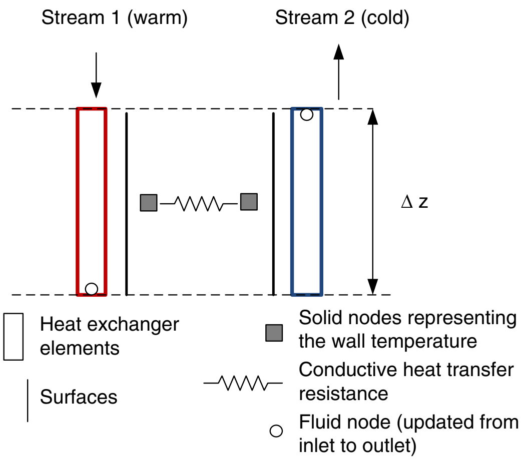
An overview of the important elements contained in the heat exchanger modeling framework is described in Table 1. At the top level, one find the fully described heat exchanger model. This is the interface to the use of a model for process simulation and optimization, and model specific geometry data need to be provided. This input data is processed an transformed to a more generic format for use at the intermediate and bottom level.

Table 1: Overview of the multi-level heat exchanger modeling framework

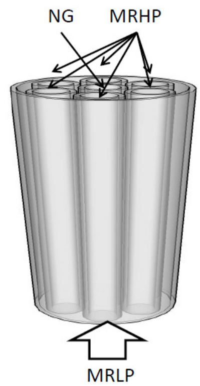
Bottom level	Basic elements	**HX element** - containing fluid nodes, geometry and correlation information **Solid-node** - representing the metal temperature and position coordinates (x,y,z) **Surface** - link between a solid-node and an element **Resistor** - link between two solid-nodes
Intermediate level	Model generator	**Circuit** - sequence of connected elements **Wall-pass** - sequence of solid nodes and their positions
	Numerical solvers	Non-linear solver for updating solid node temperatures ODE/DAE Solver for integration of the performance of the fluid pass(es)
Top level	Heat exchanger model(s)	Examples: Tube-in-tube, Tube-in-shell, Plate, Plate-Fin, Extended-surface

The actual heat exchanger model that is solved, consists only of basic elements. During model generation, all basic elements are created and connected according to a set of rules and depending of the heat exchanger type. This process creates the the fluid circuits and the wall-passes. In the finished heat exchanger model, all hx-elements belong to a circuit and all surfaces is linked to [between] one hx-element and one solid-node. The “map” of how the heat hx-elements, surfaces and solid-nodes are connected, determines the complexity of the heat exchanger. A simple illustration of two hx-elements exchanging heat is shown in Figure 3a with an example of a multi-stream LNG heat exchanger in Figure 3b.

A hx-element listed in Table 1, contains a pointer to a fluid node and to a geometry description. The fluid node stores information on all the local thermo-physical properties. The geometry description that is available can be quite complex, like for a Plate-Fin surface, but for the basic hx-element, only the channel perimeter and the cross flow area are required. In this description there is also pointers to methods for



(a) Simplified illustration of two heat exchanger elements



(b) Example of a multi-stream LNG heat exchanger used for testing purposes

Fig. 3: Basic elements in the modeling framework

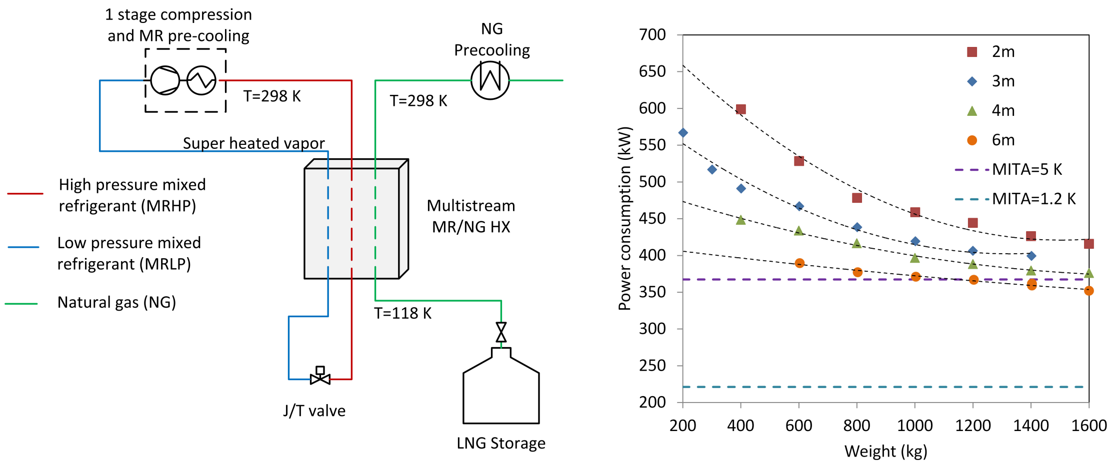
calculation of local heat transfer coefficient and wall friction. In this project fundamental measurements on droplet film interacion can contribute to improve such models.

The solution strategy used for solving the heat exchanger is based on finding the solid-node temperatures that result in a net heat flux (convective and conductive) to be zero around each node. This is a (large) system of non-linear equations, where the local convective fluxes are found by integrating the capacity and pressure through all hx-elements within each circuit, using a suitable ODE or DAE solver. The fluid-node properties for a hx-element will be updated from the inlet to the outlet due to the heat tranfer between its surface(s) and the fluid. This heat transfer is thus calculated independently of the other streams in the heat exchanger. After all fluid properties has been updatet, one iteration step for solid-node temperature can be performed. Such a scheme is very useful when calculating for instance a Plate-Fin heat exchanger layer by layer, since temperature crossing between a stream and a surface cannot occur. The scheme has also proved to be very robust regarding a wide range of operating conditions which is essential for process/component optimisation purposes.

In addition to the elements listed in Table 1, special elements like valves, flow distributors and collectors, bends, baffles and so on are also available to be included in a circuit.

3.2. Examples of use

All heat exchanger models created with this framework include a full geometrical descriptions. When such a model is parameterized and included as a sub-model for process simulation or optimisation, many of the geometrical parameters are available together with the operating conditions as “free” variables. Since geometry data like weight, surface area and foot-print area calculated by the model, such data can be used as objective function or constraints in process optimization. Such a feature was demonstrated by [18] where a generic compact heat exchanger were created and used as the main cryogenic heat exchanger in a single mixed refrigerant process used for liquefaction of natural gas. In this case, the objective function was to minimize the compressor power, but instead of using composite streams and specify a minimum temperature approach, a maximum weight was set as a constraint and the heat exchanger length was used as an parameter. The results showed that to avoid having an excessive large heat exchanger, a MITA between 4-5 was more realistic that 0.5-2 as often used in optimization literature. The SMR process layout and the main results are shown in Figure 4a and 4b

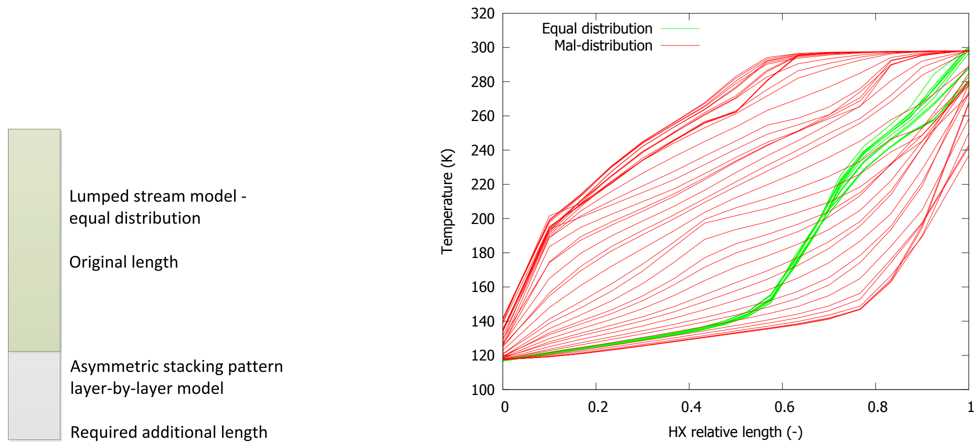


(a) Layout of the SMR process where the compressor suction and discharge pressure, and the refrigerant flow-rate was free variables

(b) Main results from the process optimization when weight constraints are imposed on the main heat exchanger

Fig. 4: Inclusion of a detailed heat exchanger model in process optimization

Another example showing the flexibility of the modeling framework is shown in [19] where a plate-fin heat exchanger is modelled and a configuration using a lumped-stream approach is compared to a configuration using individual layers. With individual layers, effects of stacking pattern on flow mal-distribution and heat exchanger performance can be studied as well situationa where multiple solutions can exsist, known as static or Ledinegg instability. The latter has also been thourougly discussed in [20]. Examples from simulations with a multi-layer plate-fin heat exchanger are shown in Figure 5a and 5b



(a) Required oversizing to achieve equal outlet condition after liquefaction of Natural Gas for a layer-by-layer with asymmetric stacking pattern when compared to an ideal case

(b) Illustration of two diffent solutions for the refrigerant flow distribution when operating in a region where static instabilities may occur

Fig. 5: Examples from simulations on a layer-by-layer configure plate-fin heat exchanger

3.3. Mathematical modeling of fluid flows in pipe networks

To gain a deeper understanding of dynamic and static instabilities in heat exchangers with several parallel channels, a PhD on mathematical modeling of junction flow was part of this project. In [21] and a in series of articles [22, 23, 24, 25] the use of various network models very studied with special attention on the applied coupling condition like: Either, Pressure, momentum flux or Bernoulli invariant as constant in the junctions. The work mainly considers models derived for the isothermal and isentropic Euler equations. For the applied test cases it was shown both theoretically and numerically that only by using the Bernoulli invariant, mass was conserved in the junction when a generalized Riemann problem was solved.

4. Evaluation of LNG processes

An important goal of LELNG has been to develop tools and methods for optimization of LNG processes. We have been using a general approach and looked at both simple processes and multi-cycle processes with mixed refrigerants. In any case LNG processes are complex and controlled by a number of parameters, such as the molar component molar flows and intermediate pressure and temperature levels of each refrigerant used in the process. The number of variables increases quite rapidly with the complexity of the processes. When optimizing LNG processes, the objective function is often the compressor power consumption, but in addition constraints are needed to ensure that there are positive temperature differences in the HXs and that the feeds to the compressors are superheated. To illustrate why LNG processes are difficult to optimize, Figure 6 shows the temperature difference constraint in the cold end of two serially connected HXs in a given LNG process. The constraint is plotted as a function of the temperature of the natural gas entering at the warm side and molar component flow rates of the refrigerant. As seen, the plotted surfaces are highly nonlinear. Another important aspect is that the execution time for one flowsheet evaluation is substantially higher than the execution time needed for the optimization routines to calculate a new set of variables to be evaluated, even when using simplified composite curve HX models. Optimization routines based on many function evaluations will therefore not be suitable. Hence, optimization of LNG processes remains a difficult problem, although quite a lot of work has been reported on topic [26].

In our work we have found that using ordinary sequential quadratic programming (SQP) routines compared with a flowsheet simulator is feasible for simple processes as long as the optimization problem is formulated well [27]. For the same problems, convergence was much faster than other techniques reported in the literature [28, 29, 30]. To compare different optimization methods in a more rigorous way, a total of 16 different variations of optimization routines have been tested [31]. These include routines for both global and local optimization and include techniques like SQP, simulated annealing [32], and evolutionary search [30]. In general the local methods based on SQP are the most efficient ones, returning good solutions for simple and well formulated LNG problems for random initial conditions with only a fraction of the evaluations required by the global solvers. The global solvers in general work well for simple analytical problems. However, for LNG processes global solvers will require at least 10 times more evaluations than SQP routines in order to get to a comparable solution.

The formulation of the optimization problem is critical in order to get good results when optimizing LNG processes [33]. For instance, when using composite curve HX models, an important constraint is the need for a positive temperature difference between the hot and the cold streams throughout the HX. SQP techniques are particularly sensitive to how this requirement is implemented. Very often the minimum temperature difference (MITA) is used since this value is easiest to retrieve from the process simulator. However, for LNG HXs using mixed refrigerants, the minimum temperature difference may occur in different locations, leading to kinks in the derivatives of MITA which are difficult to handle for SQP techniques. In order to avoid this problem, each temperature difference could for instance be used as a constraint, or the HX could be virtually split such that each new HX only contains one location where the minimum value is obtained. For SQP techniques also the estimator chosen for the derivatives is important.

For processes with multiple cycles and mixed refrigerants, the investigated standard optimization techniques will generally have problems finding good solutions from randomly generated initial conditions. The currently most successful approach has been to combine existing routines where a non-gradient based method has been used to generate a feasible solution, and a SQP based method has been repeatedly used with alternating LNG flowsheet formulations.

In addition to commercial flowsheet simulators, we have also employed the robust and fast detailed HX model in the project instead of a composite curve model in full optimization of a single-cycle LNG process. Hence, the energy efficiency of the LNG process is optimized with respect to process parameters and HX

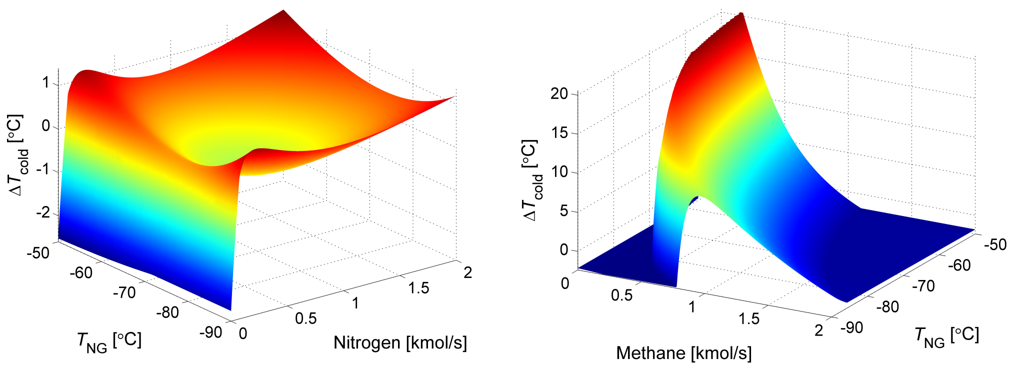


Fig. 6: Temperature difference constraint ΔT_{cold} in the cold ends of two serially connected HXs in an LNG process as a function of refrigerant composition and the natural gas inlet temperature of the first HX, T_{NG}. In the first HX shown to the left ΔT_{cold} is given as a function of nitrogen content of the refrigerant flow, whereas methane content is varied in the second HX shown to the right.

design while maintaining the weight and volume constraints. The HX is not treated like a black box but is optimized simultaneously with the process surrounding it. Preliminary results indicates that this approach is possible, but more computational expensive and somewhat less robust than when using simple HX models.

5. Conclusions

The Enabling Low-Emission LNG Systems project aims to improve design and operation of LNG plants using a multilevel research approach. In order to improve LNG processes, understanding of the heat exchangers (HXs) involved is vital, and hence these have been studied on a detailed level, both experimentally and through modeling. Furthermore, a flexible framework for robust and accurate HX modeling has been developed. Tools for optimization of LNG plants have been developed, and by careful formulation of the problem, sequential quadratic programming have been found to be most promising optimization technique. As a supplement to process optimization using composite curve HXs, work is underway to investigate the use of the new detailed HX models in optimization of simple LNG processes.

The project has been an academic success with 17 journal or proceedings articles accepted for publishing, and several more submitted, 22 conference papers presented, and not at least two PhDs completed so far, with one more due in 2014.

Acknowledgments

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