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Development, modelling and evaluation of a small-scale gas liquefaction plant

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Abstract:
A small-scale gas liquefaction plant was developed and analysed based on process simulation tools and pilot tests. It will be installed in harbours, easing the penetration of liquefied natural gas (LNG) as a maritime fuel, in a sector facing more stringent environmental regulations. The proposed plant uses a multi-component refrigerant together with a propane precooling cycle and plate heat exchangers, to achieve a higher performance. This LNG production concept was modelled based on the Danish natural gas composition. Firstly, the total power consumption and heat transfer conductance were minimised by optimising the operating conditions and the refrigerant composition. The effects of varying feed and refrigerant compositions were analysed. Secondly, the system layouts were evaluated by conducting an exergetic assessment. Finally, the most promising layouts were validated by pilot plant measurements, for a feed processing rate of 2160 kg/h. The results indicate that the specific power consumption can be reduced to the 1400-1800 kJ/kg range, for an exergetic efficiency of 25-30%. A good agreement between the simulation and experimental results was found, which justifies the use of the property database of the Groupe Européen de Recherches Gazières for system analyses.

Keywords:
Energy, Gas liquefaction, Refrigeration, Process optimization, Exergy, Small-scale

1. Introduction
Liquefied natural gas (LNG) is a mixture of hydrocarbons consisting mainly of methane, with small fractions of ethane and propane, depending on the initial feed gas composition. Natural gas is usually processed at moderate to high pressures, cooled down to around -160°C, and stored at low to near atmospheric pressures. Compared to most hydrocarbon-based fuels, LNG has greater energy content, of about 45 to 50 MJ per kg on a higher heating value basis. Compared to compressed natural gas (CNG), LNG has an energy density greater by a factor 2.5, which reaches about 22 MJ/litre. At atmospheric pressure, LNG occupies a volume 600 times lower than conventional natural gas. These properties make LNG interesting for transportation and storage purposes along the gas chain, from the extraction step to the use phase. The use of LNG has gained interest in several sectors, among these the shipping industry. The new legislation related to the Annex VI of MARPOL 73/78 from the International Maritime Organization \cite{1} deals with reduction requirements of nitrogen (NO\textsubscript{x}) and sulphur (SO\textsubscript{x}) oxides emissions. These regulations set stringent limits for specific control areas such as the Baltic Sea: for example, as of 2015, ships should not use fuels with a sulphur content exceeding 0.1%. In this context, LNG may substitute heavy fuel oil, as it significantly reduces CO\textsubscript{2}, NO\textsubscript{x} and particulate matter emissions in addition to eliminating sulphur ones.
However, the promotion of LNG as marine fuel requires the development of small-scale gas liquefaction facilities in harbours, i.e. with a capacity smaller than 1 mtpa per unit. The development of cost- and energy-efficient small-scale units is challenging, because of (i) higher production cost per unit of LNG (no economy of scale effect), (ii) the profitability of the supply chain and (iii) design constraints. The most energy-efficient liquefaction process is a cascade system [2] based on propane, ethylene and methane in three refrigeration cycles. It requires a high equipment inventory and is therefore not suitable for small-scale applications.

Expander-based layouts build on different configurations of reverse Brayton cycles [3]. They are compact, simple, and inherently safe since nitrogen is the most common refrigerant medium. However, they are generally less efficient than mixed-refrigerant processes [4], which operate with a mixture of hydrocarbons and nitrogen as working fluid. The refrigerant changes phase in the cryogenic heat exchangers, which presents the advantage of high heat transfer coefficients but the drawback of possible maldistribution issues of the two-phase flow. The refrigerant composition may also be adjusted over time if there are leaks of the most volatile components or variations of the feed gas composition.

The scientific literature on small-scale LNG systems is less extensive than for large-capacity liquefaction plants. Barclay and Denton [5] compare mixed-refrigerant and expander-based processes for small-scale offshore applications, listing the relevant selection criteria. Finn [6] compares the same processes for applications of similar size, and concludes that mixed-refrigerant processes are cost-effective if single compressors and plate-fin exchangers are used. It is added that these cycles are competitive as well for such capacities. Cao et al. [7] assess the efficiency of a nitrogen-methane expander-cycle against a single mixed-refrigerant process, and it appears that the latter is more efficient with propane pre-cooling. Remeljej and Hoadley [8] also compare mixed-refrigerant to expander-based processes and conclude that the former are the least energy-intensive.

There are few, if any, published works on the modelling, optimisation and experimental validation of gas liquefaction systems. The most relevant work is the one of Nekså et al. on natural gas re-liquefaction plants for small gas carriers. The proposed plant is a small-scale unit for liquefying the boil-off gas from gas tankers, which contains methane and a non-negligible fraction of nitrogen. It is intended for installation on gas carriers between export and receiving terminals. They present the full development of their mini-LNG concept, from the modelling to the experimental validation.

The present work introduces the development of a small-scale LNG production unit, from the modelling phase to the design optimisation and experimental validation with a test facility. The aim is to implement such facilities in Danish harbours, such as the one of Frederikshavn, for promoting the use of LNG in the marine industry.

2. Methods

2.1. Mixed-refrigerant processes

The requirements and needs for small- and large-scale liquefaction plants are different. Their performance, i.e. the specific power consumption per unit of LNG, is of key importance in both cases, but other factors are also essential for small-scale units. The number of equipment items and footprint (size and compactness), and the ease of operation and maintenance, are important as well. Processes operating with mixtures as refrigerants are historically preferred for small-scale applications, and the unit developed and presented paper belongs to this category. The refrigerant is a multi-component mixture consisting of hydrocarbons (e.g. methane, ethane, ethylene, propane, i- and n-butane, i- and n-pentane) and nitrogen. The refrigeration effect is generated by the Joule-Thomson effect, i.e. an adiabatic expansion through a valve device, where the refrigerant in liquid phase is partially flashed into the vapour-liquid region. The refrigerant mixture is zeotropic: it does not evaporate at a single temperature point, but over a temperature glide. It is therefore possible to tune the refrigerant composition to match the temperature profiles between the hot (natural gas) and cold (refrigerant) sides in the cryogenic heat exchangers. These small temperature differences lead to a high system performance without the need for a complex heat exchanger setup.
2.2. System description

A sketch of the flow layout is shown in Fig. 1. The process may be seen as an alternative layout of the propane-precooled mixed-refrigerant process (C3MR) [9], which consists of two refrigeration cycles. The first cycle consists of a propane refrigeration cycle, which ensures precooling of the second refrigerant and of the natural gas down to the -35 – -40°C range. The second one is a mixed-refrigerant refrigeration cycle, which ensures liquefaction and subcooling down to -162°C. Compared to layouts with only one single refrigerant, these setups can achieve higher system efficiency thanks to the high coefficient of performance of propane cycles. Large-scale liquefaction processes require specific multi-stream heat exchangers (spiral-wound or plate-fin) that are manufactured only by specific companies. Conventional two-flow plate heat exchangers are preferred for conventional small-scale applications. Such heat exchangers are widely used because of their compactness and high heat transfer coefficients. The refrigerant and the natural gas streams are cooled in parallel, in different series of counter-flow heat exchangers. The present system is designed for a feed processing of 2160 kg/h.

Fig. 1. Simplified process flow diagram of the proposed gas liquefaction unit.
The feed is natural gas imported from the grid and purified in a pretreatment unit for carbon dioxide and water removal. Its initial composition, considering Danish natural gas, is in the range of 90-92% methane, 6-8% ethane and 2-4% propane on a molar basis. The carbon dioxide content is negligible in comparison, representing less than 50 ppm. The fraction of butanes and pentanes does not exceed 1%. The treated gas enters the liquefaction unit at moderate to high pressures, and is first cooled down close to the ambient conditions by cooling water. It is then precooled with propane evaporating, and then liquefied and subcooled with the mixed-refrigerant. It is finally flashed to the desired storage and transport pressure. The boil-off gas is re-liquefied separately and contains higher fractions of methane and nitrogen than the feed gas. Similarly, the high-pressure refrigerant is desuperheated and partly liquefied. It enters a two-phase separator where the vapour and liquid phases are separated and processed separately. The vapour phase contains mainly light-weight hydrocarbons and nitrogen, which are the most volatile components. The liquid phase contains higher fractions of the medium-weight hydrocarbons and negligible amounts of the light-weight ones. The gas flow enters the cold box, where it is liquefied and subcooled to the same temperature as the natural gas. It is finally flashed, reaching the lowest temperature level of the complete system, and used to cool down itself.

2.3. System modelling and simulation

The liquefaction system is modelled and simulated using the software Aspen Plus version 7.2, considering the REFPROP database. It builds on the use of the multi-parameter model of the Groupe Européen de Recherches Gazières (GERG) [10], which displays the highest accuracy of all thermodynamic models for hydrocarbons. The model is based on fundamental equations of state explicit in the Helmholtz energy, and the uncertainties fall within the range of the experimental errors. The GERG model is complemented by other equations of state for chemical components such as ethylene. Preliminary simulations with the Peng-Robinson [11] and Redlich-Kwong-Soave [12] equations of state showed some discrepancies in the prediction of the liquid densities. The process was simulated assuming perfect liquid-vapour separation in the two-phase separator, no heat losses or pressure drops, and a constant compressor isentropic efficiency of 72%, which is a reasonable estimate based on the literature. The work of Nekså et al. is based on an isentropic efficiency of 65%, but the authors underline that higher efficiencies may be achievable and lead therefore to a more efficient system.

2.4. System design and optimisation

The system presents several design parameters that should be selected adequately to minimize the power consumption of the complete system. They are also constrained by the manufacturers and customer requirements:

- the high-pressure level of the refrigerant;
- the low-pressure level (after expansion);
- the precooling (cooling water and propane) temperatures;
- the final subcooling temperature;

In practice, the low- and high-pressure levels are limited by the operating envelope of the chosen compressor. As the system is meant for small-scale applications, multi-stage compression with intercooling is not desired, and a single-stage screw compressor is the preferred option. Moreover, the low-pressure level should exceed atmospheric pressures, as vacuum conditions are not desired because of leakage issues. The final temperature and pressure are constrained by the customer requirements. However, the selection of the subcooling temperature has an impact on the amount of boil-off gas after flashing.

- the chemical components in the refrigerant; and
- their respective fractions.

There is no standard mixture composition applicable to all natural gas feeds. The fractions of each component should be chosen adequately depending on the system specifications. In general, methane is required because of its low boiling point (around -162°C at 1 atm) and high latent heat
Nitrogen (about 510 kJ/kg). Nitrogen (-195°C at 1 atm) is also added because flashing the refrigerant mixture under vacuum conditions is impracticable from an operational perspective. Ethane, ethylene and propane are added to tune the refrigerant composition and minimise the system power consumption. Butanes and pentanes may also be added, although they are usually not desirable because of their freezing points in the range of -130°C to -140°C. Additional issues may be addressed, such as the mixture stability and possible maldistribution in the phase change processes. The mixture composition and the system operating parameters were determined by mathematical optimization, linking a genetic algorithm developed at EPFL on Matlab [13] and the simulation software Aspen Plus [14]. The abovementioned parameters were taken as degrees of freedom (decision variables) in the optimization problem. Values suggested by the industrial partners were taken as boundary constraints. The objective function was to minimize the total power consumption, respecting a minimum temperature difference of 5K within the heat exchangers.

2.5. System test and validation

The proposed system was developed at test scale, using the same refrigerant mixture and overall design layout. However, for practical reasons, the fluid to liquefy in the test facility was nitrogen. As nitrogen does not liquefy in this temperature range. Several temperatures therefore had to be adapted and the size of the nitrogen-refrigerant heat exchanger was adapted.

2.6. System evaluation

The performance of the gas liquefaction system was evaluated by performing an energy and exergy analysis. The first law of thermodynamics states that energy cannot be created or destroyed, but is only transformed from one form to another (e.g. electrical to thermal energy, etc.). It gives useful insights on the processes taking place within the overall system – it shows as well the losses and dissipation to the environment. However, it cannot be used to assess the performance of a given system against an ideal one.

\[ \dot{Q} - \dot{W} = \sum \dot{m}_{out}h_{out} - \sum \dot{m}_{in}h_{in} \]

Where \( \dot{Q} \) and \( \dot{W} \) stand for the energy rates in the form of heat and power, \( \dot{m} \) for the flow rate of a given material stream (inflowing or outflowing) and \( h \) the specific enthalpy.

On the contrary, the second law of thermodynamics pinpoints that irreversible phenomena take place in real processes, such as heat transfer across finite temperature differences. These non-idealities should then be identified to assess and improve the overall system performance. Exergy can be defined as ‘the maximum theoretical useful work (shaft work or electrical work) as the system is brought into complete thermodynamic equilibrium with the thermodynamic environment while the system interacts with it only’ [15]. This concept can therefore be applied for quantifying and locating the sources of performance losses, and thus the potentials for improving the overall system. The exergy balance of an open control volume in steady-state and steady-flow processes can be expressed as:

\[ \dot{E}_d = \sum \dot{m}_{out}e_{out} - \sum \dot{m}_{in}e_{in} + \sum \dot{E}^Q - \dot{W} \]

where \( \dot{E}_d \) is the destroyed exergy, \( \sum \dot{m}_{out}e_{out} \) and \( \sum \dot{m}_{in}e_{in} \) are the exergy flows associated with material streams, and \( \sum \dot{E}^Q \) the exergy flow associated with heat transfer. The exergy flows associated with streams of matter are related to their physical and chemical properties, while the exergy associated with a heat flow is related to the temperature at which the heat transfer process takes place. The concept of exergy is intrinsically linked to a dead state, which was defined here as 1.013 bar, 15°C, and with the chemical composition of the environment defined by Morris and Szargut [16]. The temperature of 15°C was chosen as the water temperatures in the Baltic and North Seas usually do not exceed this threshold. The exergetic efficiency is defined as the ratio of the exergetic product, which is the gain of exergy associated with the production of LNG, to the exergetic fuel, which is the power consumption of the overall system.
3. Results and discussion

3.1. Process simulation of the LNG unit

The LNG unit was modelled, simulated and optimised with Aspen Plus 7.2, analysing the impact of the operating conditions and mixture composition on the overall system performance. A zone analysis of each heat exchanger was performed to verify the thermodynamic feasibility of the heat transfer process. Each heat exchanger was divided into 30 control volumes for which the temperatures and enthalpies on each side were calculated.

The temperature profiles were matched by adjusting the pressure levels and mixture composition. The temperature-enthalpy profiles are shown, in the optimised case (Fig. 2), for two of the cryogenic heat exchangers. The liquefaction of natural gas takes place between the dew and bubble points of the natural gas, at around -35°C and -85°C. It is then followed by the subcooling process down to -155°C and a flash expansion to the final temperature of -162°C. On the cold side, the refrigerant evaporates from about -160°C to -78°C and is finally superheated up to -45°C. The minimum temperature differences are found within the NG/refrigerant heat exchanger, at about 40% of the cooling process, and at the hot end, at around 100%.

![Fig. 2. Temperature profiles within the cryogenic heat exchangers – natural gas/refrigerant (left) and high-pressure/low-pressure refrigerant (right)](image)

The same type of analysis was carried out for the other heat exchangers, and the temperature profiles within the cold refrigerant-refrigerant heat exchanger are presented. The temperature difference is close to 5°C in the entire heat exchange process, which can be explained by the similarities in composition and the small pressure difference between the hot and cold sides. It is worth noting that the duties of these heat exchangers are not similar. They are usually higher for internal heat exchangers, as the refrigerant has a greater flowrate and undergoes phase change as well.

The power consumption of the LNG unit, without precooling, reaches 0.27 kWh/kg LNG, and amounts to 0.46 kWh/kg LNG with precooling. These numbers are deemed satisfactory, as the EU target is 0.75 kWh/kg LNG. They should nevertheless be considered as preliminary numbers based on several assumptions such as zero heat losses, which is unlikely in practice. However, as mentioned by Nekså et al., higher values of the compressor isentropic efficiency for state-of-the-art small compressors may be attained, resulting in even smaller numbers.
3.2. Sensitivity analyses of the LNG unit

Sensitivity analyses on the refrigerant mixture composition were conducted to analyse the overall system behaviour. For example, an increase of the methane partial flowrate (Fig. 3), while keeping the other partial flowrates constant, results in smaller temperature differences in the internal heat exchangers i.e. in greater heat exchange areas. It also leads to greater power consumption and to higher suction temperatures. Decreasing the methane flowrate may be beneficial, but entails lower suction temperatures at a rate of 1.5°C per 1% of CH₄, which may cause issues of thermal stress in the compressor.

Fig. 3. Sensitivity analyses with respect to the methane flowrate, for the approach temperatures (left) and power consumption (right)

3.3. Performance analysis of the LNG unit

The performance of the proposed LNG plant was analysed by analysing the energy flows and performing an exergetic assessment on the optimised case. A comparison of the heat exchangers (Fig. 4) shows that the heat duty of the LNG/refrigerant heat exchanger is small in comparison to the internal heat exchangers. This is expected for mixed-refrigerant processes because of the large refrigerant flow compared to the natural gas one. The duty of the boil-off heat exchanger is negligible, as little gas is actually flashed in the final expansion.

Fig. 4. Heat duties per heat exchanger, expressed in kW
The aim of the exergy analysis was to suggest possible improvements and discuss further which ones are feasible from an economic and practical perspective. The exergy losses associated with the cooling water are accounted as exergy destruction, since this water cannot be used for any practical purposes because of its low temperature. The term “cold box” refers to the liquefaction and subcooling step, by opposition to the precooling. It includes the internal heat exchangers and the refrigerant-natural gas one together with the cryogenic valves and mixers. The term “propane precooling” refers to the propane precooling cycle, without including the propane compressors. It includes the exergy destruction related to the discharge of exergy from the propane to the cooling water. The term “water precooling” refers to the precooling with cooling water, upstream the precooling cycle with propane, to cool down the mixed refrigerant and natural gas down to 30°C. Finally, the term “valves” refers to the final expansion valve to decrease the LNG pressure from the liquefaction step to the storage and distribution.

The distribution of the exergy destruction (Fig. 5) shows that those can be roughly distributed into the compressors, the cold box and precooling with propane. The exergy destruction associated with the water precooling and throttling valves are negligible in comparison. Significant quantities of heat dissipated from the superheated mixed refrigerant and natural gas are discharged near the ambient conditions, which translates into small exergy losses. By opposition, the exergy destruction in the cold box and propane precooling are much larger because the heat transfer takes place at cryogenic conditions.

The exergy destruction in the compressors is associated with the non-idealities of the compression process (e.g. friction). The compressor used for increasing the mixed-refrigerant pressure is responsible for the highest share of irreversibilities. The propane compressors rank second and third, while the boil-off gas compressor is fourth. The exergetic efficiency is about 25%, and this low value can be imputed to the large compressor inefficiencies and irreversibilities within the cold box.

Fig. 5. Exergy destruction within the LNG unit, sorted by sub-systems

The distribution of the exergy destruction within the cold box (Fig. 6), including the mixed-refrigerant and boil-off gas compressors, shows that the compression losses dominate. These losses can, in practice, be reduced only by selecting more efficient compressors, which may be realistic considering the low value assumed in the simulations. Multi-stage compression is not desired, as this would result in a bigger system.

The exergy destruction in the internal and LNG heat exchangers is associated with the heat transfer process. It can only be reduced by decreasing the minimum temperature difference between the hot and cold streams, at the expense of a higher heat exchanger area and greater capital costs. It is worth noting that a mixture judged optimum for a temperature difference of 5 K may not be optimum if
this number is changed to 3 or 4 K, as the temperature profiles are not linear. Temperature differences as small as 1 to 2 K may be expected for state-of-the-art heat exchangers in large-scale applications.

![Exergy destruction within the cold box, including the compressors, sorted by components](image1)

**Fig. 6.** Exergy destruction within the cold box, including the compressors, sorted by components

### 3.4. Preliminary simulations of the test facility

The proposed liquefaction system has been developed at test scale to analyse the mixture behaviour in terms of heat transfer characteristics, verify the feasibility of the complete layout and check the temperatures and pressures. As mentioned earlier, the fluid used for the test measurements is nitrogen instead of natural gas. Several operating conditions have been adjusted to ensure the system feasibility, such as the final temperature and refrigerant flowrates. A zone analysis was performed as well for each heat exchanger (Fig. 7). At the difference of a conventional gas liquefaction plant, the minimum temperature difference is not found at the hot end, after superheating of the refrigerant, but at the cold one, after flashing. This impacts as well the internal heat exchangers. A smaller flow of refrigerant is processed in the whole system (less refrigerant per kg of nitrogen than per kg of natural gas), but a greater flow of low-pressure refrigerant is processed through the internal heat exchanger (higher ratio of LP to HP refrigerant in the test facility).

![Temperature profiles within the cryogenic heat exchangers – nitrogen/refrigerant (left) and high-pressure/low-pressure refrigerant (right)](image2)

**Fig. 7.** Temperature profiles within the cryogenic heat exchangers – nitrogen/refrigerant (left) and high-pressure/low-pressure refrigerant (right)
3.5. Comparison of test and model results

The model of the LNG system made in Aspen was validated using data from the small-scale test setup. The flow layout of the test setup was similar to the model layout (Fig. 1). The design parameters in Aspen consisting of temperatures, pressures and mass flows were substituted with data measured during test of the small-scale test facility. The test data was obtained by averaging the data from 20 minutes steady state operation, ensuring that the system was in balance during the whole test interval. For validation of the Aspen model 3 tests were carried out and the data averaged for each test. The design parameters were inserted into the Aspen model for further simulations. The temperatures calculated in the Aspen model were compared to the temperatures acquired from the tests. Table 1 shows the input parameters for the model, underlined as red nodes. A comparison of the calculated temperature (Sim.) with the measured (Meas.) one for the free parameters is shown for each variable, together with the absolute difference (Diff).

Table 1. Comparison between the model results and test data for three tests. The state points are given in Figure 1. The nodes marked with red are test result used as model input.

<table>
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<th>Node</th>
<th>Test 1</th>
<th>Test 2</th>
<th>Test 3</th>
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The result of the validation shows small discrepancies between the model and test results regarding the temperatures inside the cold box (state points 1-9) with exception of the refrigerant temperature out of the nitrogen heat exchanger. However, it is worth noticing that the mass flow meter (m<sub>n</sub>) for nitrogen was inaccurate for small mass flow rates (designed for 250 kg/h but run at around 45 kg/h during the tests). A lower nitrogen mass flow rate would have resulted in a lower temperature of the refrigerant out of the heat exchanger.
The deviation for the temperature at the compressor outlet ($T_{10}$) is acceptable for Test 1, but goes up to 13°C for Tests 2 and 3. This could derive from (i) the lack of information on the compressor isentropic efficiency, or from (ii) the high ambient temperature at the compressor location, which would lead to smaller heat losses.

The comparison between the test and simulation program validates the model, which is particularly interesting as the behaviour of the refrigerant mixture was not well-known before the tests. The properties of the refrigerant mixture are correctly predicted at different pressure levels, as the temperature after expansion corresponds to the one returned by the model.

The quantities of energy absorbed and released in the heat exchangers are almost the same in the model and test, implying that the heat losses are small. A last observation is that the measuring equipment at the test station is accurate, because the results returned by the model correspond to the ones measured in the test.

4. Conclusion

A mixed-refrigerant process for small-scale natural gas liquefaction was modelled, optimised and built in a test facility. Fundamental equations of state explicit in the Helmholtz free energy were used to predict accurately the thermophysical properties of the refrigerant and feed, as well as the temperatures and pressures. The system performance was analysed by carrying out an exergy analysis, and the overall plant was optimised taking the mixture composition, operating temperatures and pressures as degrees of freedom. Several sensitivity analyses were carried out to analyse the system behaviour under variations of the refrigerant composition.

The present work shows that the power consumption of the proposed facility can be much lower than the threshold value suggested in the EU directives. This results from a close match of the temperature profiles in the cryogenic and internal heat exchangers. The exergetic assessment shows that the compressors represent the lion’s share of the total system irreversibilities, followed by the cryogenic box. These findings suggest therefore further investment in more performant compressors and larger heat exchangers, although this may not be practicable for small-scale applications. Finally, the comparison between the test and simulation results shows good agreement for all tested variables, from temperatures to heat loads, with the exception of two temperatures. The first discrepancy may be imputed to the inaccuracies of the flow meter at the test conditions, whilst the second may result from an incorrect estimate of the compressor isentropic efficiency. All in all, those results support the use of these models for further development of the liquefaction facility.

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