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Assessment of predictive methods for the heat transfer coefficient during flow boiling of R50 and R170 mixtures

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Abstract. Liquefied natural gas (LNG) is a binary mixture of methane (R50) and ethane (R170). Proper evaluation of the heat transfer coefficient during flow boiling of such a mixture is very important to design LNG facilities. Pure fluid correlations have been tested with and without the adoption of corrections to account for the lowered performance due to mass transfer resistances arising both in the nucleate boiling and in the convective boiling regimes. In total, the prediction of 13 correlations has been compared with a new data set from the literature. Results show that the correction related to nucleate boiling has the greatest impact though, in general, the adoption of the corrections leads to under predict the data.

1. Introduction

Liquefied natural gas (LNG) is a binary mixture of methane (R50) and ethane (R170). The boiling characteristic of LNG have high importance not only for storage and transport, such as in vaporization section, but also for liquefaction, since R50 and R170 are the fundamental components in mixed-refrigerants for Joule-Thomson refrigeration cycle, widely used in LNG industry. The study of boiling heat transfer of R50/R170 binary mixtures is then very important to properly design LNG facilities, which requires reliable methods to compute the heat transfer coefficient.

Boiling heat transfer of multi-component mixtures has been extensively studied in the past and most of the approaches model the degradation of the heat transfer coefficient due to the arising mass transfer resistance.

Part of the models are developed for specific fluid mixtures under specific flow patterns. As significant examples, the Mishra [1] correlation is developed for R12/R22 mixtures in annular flow. Jung et al. [2], Granryd [3], Takamatsu et al. [4] work with R22/R114 and/or R12/R152a sharing at least part of the database. Accordingly, generalization of the results is hard to be achieved.

A more general approach consists of the correction of pure fluid correlations to account for performance degradation both in the nucleate boiling dominant and in the convective boiling dominant regime. Among the most recent contributions, starting from the Liu and Winterton [5] correlation, Zou et al. [6] modified the part describing the boiling effect, whereas Guo et al. [7] refitted both the convective enhancement factor and the boiling suppression factor using data for an R134a/R245fa mixture. On the other hand, Shah [8] proposed a general approach starting from any pure fluid correlation and adopting the Thome and Shakhir [9] correction for pool boiling of zeotropic mixtures to update the prediction in the boiling dominant regime, and the Bell and Ghaly [10] method to correct the prediction in the convective dominant regime. This approach was successful in predicting an 878 points database from 21 independent studies, except for LNG data taken from only one source [11].

One of the most recent approaches is provided by Zhang et al. [12] giving a physics-based correlation, in which a suitable formulation is derived according to the value of the ratio between the temperature glide and the saturation temperature. The correlation has been tested against 2091 data from 22 independent research groups, showing a mean absolute percentage deviation of 29.0%. However, validation does not seem to include explicitly LNG mixtures.

Data for LNG evaporation are relatively scarce [8]. Recently, Gong et al. [13] have published new measurements for R50/R170 mixtures at various composition, pressure, heat and mass flux. They reported 100 experimental points that can be used to check the so-called general prediction methods mentioned above. Accordingly, this paper presents a comparison between 13 correlation approaches applied to the prediction of the Gong et al. [13] LNG database as reported in the published work.

2. Correlations for the heat transfer coefficient during flow boiling in binary mixtures

As for a single-component system, a superheat is required to form a bubble of radius r. At equilibrium, from the Clausius-Clapeyron equation, it arises:

$$T_g - T_{SAT} = \frac{2\sigma}{(\partial p_{SAT}/\partial T)r} \tag{1}$$

For binary mixtures, the surface tension (σ), the bubble radius (r), and the slope of the vapour pressure curve ($\partial p_{SAT}/\partial T$) depend on the composition. An extension of Eq. 1 is provided by Stein [14]:

$$\frac{\partial p_{SAT}}{\partial T} = \left(\frac{\partial p}{\partial T}\right)_{\tilde{x}} + \frac{p}{\tilde{R}T}\left(\frac{\partial \tilde{x}}{\partial T}\right)_p (K-1)\tilde{x}\left(\frac{\partial^2 g}{\partial \tilde{x}^2}\right)_{T,p} \tag{2}$$

where K is the equilibrium constant and g the Gibbs free energy. It can be shown [15] that the second term in the right hand side is always negative, hence ($\partial p_{SAT}/\partial T$) will be always lower than the value for a pure fluid system. However, it is remarkable that this change is lower than the changes in both the surface tension and the active nucleation site radius. The maximum size of nucleation site is determined by the penetration of liquid into pre-existing surface cracks, a phenomenon influenced both by contact angle and surface tension changes. Shock [16] showed that for ethanol/water mixtures these changes can be very high and with a small addition of ethanol the superheating required drastically increases. There is a large difference between activation and deactivation superheating, which are affected by composition. Shakir and Thome [8] showed in a study about heterogeneous nucleation that Eq. 1 cannot predict the experimental variation between activation and deactivation of nucleation sites, which is considerably influenced by mass diffusion between the liquid and bubble.

2.1. The correction model

A model derived from an existing pure fluid correlation by applying corrections in order to take into account mixture effects is referred to as a correction model. Corrections apply to both nucleate boing and convective boiling regimes.

2.1.1. Nucleate boiling

Nucleate boiling heat transfer is strongly related to bubble dynamics and is the dominant mechanism at low qualities and high heat fluxes. Considering a binary mixture, at the liquid-vapor interface, the concentration of the less volatile component in the liquid is higher than in the bulk liquid due to the evaporation of the more volatile one. Hence, a higher bubble point is found, causing a decrease of the effective superheating. Moreover, the more volatile component from bulk liquid has to diffuse through the depleted region surrounding the bubble to ensure bubble growth, resulting in an additional mass transfer resistance. To account for these deviations compared to the pure fluid system, different corrections have been developed for pool boiling. Schlünder [17] developed a boiling model including mass diffusion shell resistance in an *n*-component mixture, derived from film layer analysis, which was able to predict the heat transfer coefficient over a wide range of reduced pressures:

$$F_c = \frac{h}{h_{id}} = \left\{ 1 + \frac{h_{id}}{q} \sum_{j=1}^{n-1} \left(T_{SAT,n} - T_{SAT,j} \right) \cdot \left(\tilde{y}_i - \tilde{x}_i \right) \cdot \left[1 - exp\left(-\frac{Bq}{\rho_l H_{lg} \beta_l} \right) \right] \right\}^{-1} \qquad (3)$$

According to Shah [8] the ideal heat transfer coefficient is conveniently calculated from the Cooper correlation [18]:

$$h_{Cooper} = 55 p_r^{0.12} \left(-log\, p_r \right)^{-0.55} M^{-0.5} q^{2/3} \qquad (4)$$

Moreover, taking the scaling factor B as unity implicitly assumes that all the heat transferred to the bubble interface is converted into latent heat. The liquid phase mass transfer coefficient β_l is set to the recommended value of 3×10^{-4} m/s. The Schlünder's model considers each component at equilibrium, but $\tilde{x}_i$ and $\tilde{y}_i$ are evaluated at their bulk values, which are not representative of the vapor-bubble interface. Moreover, at high and low reduced pressure some component may be under the freezing point or above the critical point, causing an impossible determination of saturation temperatures. Finally, Schlünder assumed for the sake of simplicity a fixed slope of the bubble-point curve for all composition, which does not describe well phase equilibria.

Thome and Shakir (TS) [9] refined the model to include the glide temperature and overcome the above mentioned issues. Their formulation reads:

$$F_c = \frac{h}{h_{id}} = \left\{ 1 + \frac{h_{id}}{q} \cdot \left(T_{dew} - T_{bub} \right) \cdot \left[1 - exp\left(-\frac{Bq}{\rho_l H_{lg} \beta_l} \right) \right] \right\}^{-1} \qquad (5)$$

It is stated that these assumptions hold for mixtures with temperature glide up to 30 K.

2.1.2. Convective boiling

Convective boiling is governed by evaporation mechanism that takes place at liquid-vapour interface and is dominant at low heat fluxes and high qualities. Once nucleation is suppressed, the mass transfer resistance effect is present in both vapour and liquid films but with a smaller influence [19]. The main heat transfer degradation is here caused by the additional sensible heat transfer to the liquid and vapour phases due to increase of bubble point temperature that occurs along the flow direction. Furthermore, physical mixture properties are smaller compared to the ideal values. Finally, the temperature glide affects heat transfer in the sense that the higher the glide, the higher the mass resistance such that a full suppression of nucleation may happen.

The correction for convective boiling follows the method proposed by Bell and Ghaly (BG) [10], based on a study on condensation. In their approach, a vapour-side thermal resistance was included in the overall thermal resistance:

$$\frac{1}{h} = \frac{1}{h_c} + \frac{Y}{h_g} \qquad (6)$$

$$Y = x c_{p,g} \frac{\partial T_{dew}}{\partial H} \qquad (7)$$

The single-phase vapour convective heat transfer coefficient is estimated through the Dittus-Boelter correlation [20] assuming the vapour flowing alone (i.e. with the superficial velocity):

$$h_g = 0.023 Re_g^{0.8} Pr_g^{0.4} k_g / D \qquad (8)$$

Since in both multicomponent convective boiling and multicomponent condensation sensible heat transfer and mass transfer resistance are present, an analogy is found. The method can be applied to boiling by replacing the condensation heat transfer coefficient h_c with the convective boiling heat transfer coefficient, and T_{dew} with T_{bub}. Finally, it was shown that:

$$\frac{\partial T_{dew}}{\partial H} \approx \frac{\partial T_{bub}}{\partial H} \approx \frac{T_{dew} - T_{bub}}{H_{lg}} \qquad (9)$$

2.1.3. Pure fluid correlations

The two corrections reported above are applied to pure fluid correlations. Four of them, which are well-known in the literature, have been selected for this work: the Gungor and Winterton's issued in 1986 (GW86) [21], the Gungor and Winterton's issued in 1987 (GW87) [22], the Liu and Winterton's (LW) [5], the Shah's (SH) [8] correlations have been evaluated with both corrections (TSBG), with only one correction (TS) and without corrections at all. The equations are summarized in Table 1. When needed, the Dittus-Boelter correlation [20] has been adopted to compute the convective heat transfer coefficient for the liquid-alone flow (see Eq. 8 replacing the properties of the liquid).

Denomination	Correlation	Additional information
GW86 [21]	$h = Sh_{Cooper} + Fh_l$	$F = 1 + 24000 Bo^{1.16} X_{tt}^{-0.86}$ $S = \left(1 + 1.15 \cdot 10^{-6} F^2 Re_l^{1.17}\right)^{-1}$
GW86-TS	$h = F_c Sh_{Cooper} + Fh_l$	F and S as above
GW86-TSBG	$h = F_c Sh_{Cooper} + \left[(Fh_l)^{-1} + Y/h_g\right]^{-1}$	F and S as above
GW87 [22]	$h = Fh_l$	$F = 1 + 3000 Bo^{0.86} + 1.12(1/x - 1)^{-0.75}\left(\rho_l/\rho_g\right)^{0.41}$
GW87-TS	$h = F_{mix} h_l$	$F_{mix} = 1 + 3000(F_c Bo)^{0.86} + 1.12(1/x - 1)^{-0.75}\left(\rho_l/\rho_g\right)^{0.41}$
GW87-TSBG	$h = \left[(F_{mix} h_l)^{-1} + Y/h_g\right]^{-1}$	F_{mix} as above
LW [5]	$h = \left[(Fh_l)^2 + (Sh_{Cooper})^2\right]^{1/2}$	$F = \left[1 + x Pr_l\left(\rho_l/\rho_g - 1\right)\right]^{0.35}$ $S = \left(1 + 0.055 F^{0.1} Re_l^{0.16}\right)^{-1}$
LW-TS	$h = \left[(Fh_l)^2 + (F_c Sh_{Cooper})^2\right]^{1/2}$	F and S as above
LW-TSBG	$h = \left[(1/Fh_l + Y/h_g)^{-2} + (F_c Sh_{Cooper})^2\right]^{1/2}$	F and S as above
SH [8]	$h = max[h_{cb}; h_{nb}; h_{bs}]$ $h_{cb} = \left(J^{0.8}/1.8h_l\right)^{-1}$ $h_{nb} = h_l\left(230 Bo^{0.5}\right)$ $h_{bs} = 0.064 h_{nb} \, exp\left(aJ^n\right)$	$\begin{cases} J = Co \text{ if } Fr_l > 0.04 \\ J = 0.38 Fr_l^{-0.3} \text{ if } Fr_l < 0.04 \end{cases}$ $\begin{cases} a = 2.47; n = -0.15 \text{ if } J < 0.1 \\ a = 2.47; n = -0.1 \text{ if } J > 0.1 \end{cases}$
SH-TS	$h = max[h_{cb,mix}; h_{nb,mix}; h_{bs,mix}]$ $h_{cb,mix} = \left(J^{0.8}/1.8h_l\right)^{-1}$ $h_{nb,mix} = h_l F_c\left(230 Bo^{0.5}\right)$ $h_{bs} = 0.064 h_{nb,mix} \, exp\left(aJ^n\right)$	J, a and n as above
SH-TSBG	$h = max[h_{cb,mix}; h_{nb,mix}; h_{bs,mix}]$ $h_{cb,mix} = \left(J^{0.8}/1.8h_l + Y/h_g\right)^{-1}$ $h_{nb,mix} = h_l F_c\left(230 Bo^{0.5}\right)$ $h_{bs} = 0.064 h_{nb,mix} \, exp\left(aJ^n\right)$	J, a and n as above

Table 1. Summary of the correction models.

2.2. A recent correlation for binary mixtures

Recently, Zhang et al. [12] introduced a new correlation specifically designed for binary mixtures. The model considers that the temperature glide plays a major role since a large value leads to a great suppression of nucleate boiling, hence to a dominant contribution of convective boiling. Besides, the saturation temperature affects heat transfer since its increase determines a lowered surface tension, a higher slope of vapour pressure curve thus increasing the size of active nucleation sites. Hence, at higher saturation temperatures the suppression of nucleate boiling is weaker.

It is then introduced the dimensionless temperature glide $T^* = (T_{dew} - T_{bub})/T_{bub}$. The approach considers a region ($T^* < 0.06$) in which both convective boiling and nucleate boiling coexist, and a region where only convective boiling is present ($T^* > 0.06$). Accordingly, the heat transfer coefficient is formulated as:

$$h_{TP} = \left[(0.7 \cdot F_c h_{Cooper})^2 + h_{Mishra}^2 \right]^{0.5} \quad \text{for} \quad T^* < 0.06 \tag{10}$$

Where

$$h_{Mishra} = 21.75 \left(\frac{1}{X_{tt}} \right)^{0.29} Bo^{0.23} h_l \tag{11}$$

$$h_{TP} = \left[1 + 3000 (F_c Bo)^{0.86} + 1.12 \left(\frac{x}{1-x} \right)^{0.75} \left(\frac{\rho_l}{\rho_g} \right)^{0.41} \right] h_l \quad \text{for} \quad T^* > 0.06 \tag{12}$$

3. Data analysis

LNG flow boiling data are relatively scarce. Shah [8] based on the currently available database from Chen and Shi [11] concluded that unexpectedly mixture effects were not observed, thus correlations with no correction were able to achieve the best prediction.

Recently, Gong et al. [13] provided a new experimental dataset (in total 100 data point reported in the paper) for flow boiling of R50-R170 mixtures in 50 mm long, 6 mm inner diameter copper tubes, varying composition, pressure, heat and mass flux. The operating conditions range as follows: pressure, 0.3-0.7 MPa; heat flux, 5-62 kW m^{-2}; mass flux, 150-250 kg m^{-2} s^{-1}; component R50/R170 (mole fraction), 0.76/0.24, 0.43/0.57, 0.31/0.69, 0.15/0.85. Fluid properties have been computed through REFPROP [23]. As a representative sample, the phase diagram at 0.3 MPa is reported in Figure 1.
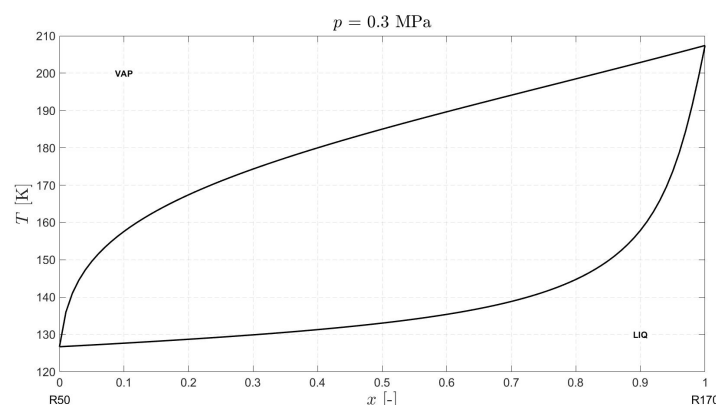

Figure 1. Phase diagram for R50-R170 mixtures.

All the 13 correlations listed in Table 1 have been tested against this database by evaluating the mean absolute relative deviation (MARD), the mean relative deviation (MRD), the percentage of data points within ±15% deviation (PCT15), the percentage of data points within ±30% deviation (PCT30) and the percentage of data points within ±50% deviation (PCT50):

$$MARD = \frac{1}{n}\sum_{i=1}^{n}\frac{\left|h_{i,\text{model}} - h_{i,\text{exp}}\right|}{h_{i,\text{model}}} \tag{13}$$

$$MRD = \frac{1}{n}\sum_{i=1}^{n}\frac{h_{i,\text{model}} - h_{i,\text{exp}}}{h_{i,\text{model}}} \tag{14}$$

It is worth noting that all the experimental conditions are characterized by $T^{*}>0.22$, which would imply only convective boiling according to Zhang et al. [12] (see Eq. 12). However, bubble point temperatures for R50-R170 mixtures are very low, hence assuming $T^{*}=0.06$ to discriminate between boiling dominant and convective dominant evaporation might be misleading. Accordingly, the threshold value of T^{*} has been set to ensure the best correlation performance. Reference is then made to the "modified" Zhang et al. model.

4. Results

The statistical parameters are summarized in Table 2. Figure 2 reports the predicted heat transfer coefficients against the experimental values for the three best predictors. The following considerations arise from the comparisons.

Model	MARD [%]	MRD [%]	PCT15 [%]	PCT30 [%]	PCT50 [%]
GW86 [21]	94.0	93.3	2.0	10.9	22.8
GW86-TS	42.6	38.3	25.7	38.6	59.4
GW86-TSBG	41.8	37.3	24.8	39.6	60.4
LW [5]	57.2	55.4	13.9	26.7	44.6
LW-TS	30.1	-24.9	12.9	52.5	88.1
LW-TSBG	42.0	-42.0	8.9	19.8	69.3
GW87 [22]	**28.0**	**23.8**	**34.7**	**52.5**	**85.2**
GW87-TS	29.1	-27.0	25.7	38.6	59.4
GW87-TSBG	47.7	-47.7	1.0	13.9	48.5
SH [8]	**18.8**	**4.7**	**37.6**	**80.2**	**100.0**
SH-TS	39.4	-38.3	11.9	22.8	75.3
SH-TSBG	53.8	-53.8	1.0	11.9	27.7
Zhang et al. [12]	**21.4**	**-19.5**	**36.6**	**73.3**	**99.0**

Table 2. Statistical indicators of prediction performance. The three best predictors are highlighted.
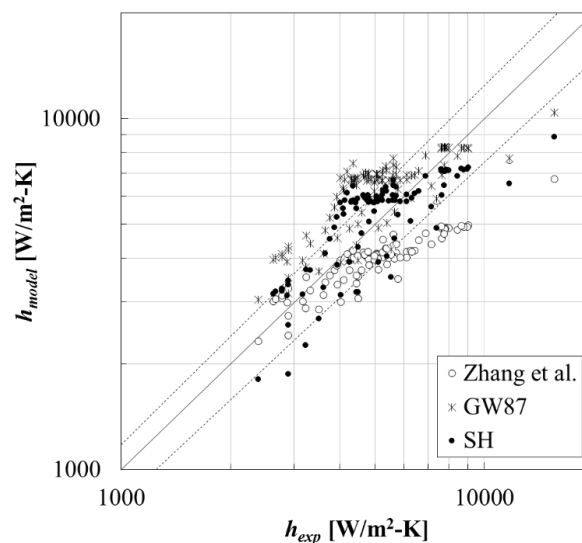

Figure 2. Parity plot for the three best predictors.

The GW87 [22], the SH [8] and the modified Zhang et al. [12] correlations are the best ones from the MARD point of view. The GW87 [22] underperforms slightly considering the PCT30 and PCT50 values compared to the others. For the Zhang et al. correlation [12], a threshold value T^*=0.39 has been determined as the one leading to the best performance for all the statistical parameters, which suggests that nucleation is a very important factor in determining the heat transfer performance. In any case, the SH [8], showing the best statistical indicators, and the GW87 [22] are pure fluid correlations, which confirms that mixture effects for R50 and R170 are possibly overestimated using either the Thome and Shakir [9] or the Bell and Ghali [10] corrections.

The Thome-Shakir correction [9] can either improve the heat transfer coefficient prediction as in GW86 [21] and LW [5] correlations or worsen it as in GW87 [22] and SH [8] correlations. Such a correction is mostly related to bubble nucleation, hence it can be inferred that nucleation plays a significant role in the data set, which confirms the above reported consideration about the effectiveness of the Zhang et al. [12] model. It can be noted from the MRD values that, as expected, the correction always lowers the prediction, leading in most cases to underestimate the data. However, it has to be stated that the validity range of the correction is up to 30 K glide, while for the dataset the glide ranges between 40 K and 55 K.

The TSBG correction generally worsens the prediction since the Bell and Ghali [10] model introduces further reduction in the heat transfer coefficient. Accordingly, strong data underestimation is observed. The only exception is seen with the GW86 [21] model since it over predicts so strongly the data that it benefits from the double correction.

5. Concluding remarks

A new data set including R50-R170 mixture flow boiling data has been analysed. 13 correlations have been tested to derive general considerations. The Shah [8] and the modified Zhang et al. [12] models showed the best MARD, respectively 18.8% and 21.4%.

There is evidence that on this data set the correction method by Thome and Shakhir [9] provides a general improvement compared with the basic pure fluid correlation. However, it may lead to significant overestimation of the heat transfer coefficient, which might be due to a glide temperature range higher than the applicable range.

The Bell and Ghali correction [10] always lowers considerably the prediction. If it is applied together with the Thome and Shakhir's [9], the results strongly under predict the data.

Nucleate boiling appears as a key mechanism in the heat transfer performance in LNG mixtures, hence extensive experimental study is still needed to extend the database useful to validate the models.

6. Acknowledgements

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