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Evaluation of prediction models for the physical parameters in natural gas liquefaction processes

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ABSTRACT

The natural gas liquefaction process is a complicated and dynamic thermal system. Operation conditions change during the process of compression, throttling and heat transfer, which inevitably leads to changes of the thermodynamic property parameters and the phase state of the natural gas and refrigerants. Performing a simulation of the liquefaction process is one of the main methods to improve the economic efficiency of the process and to reduce the production cost; such a simulation can be conducted using a process simulation software package, such as Aspen HYSYS, Aspen Plus, SIMSCI PRO-II and Honeywell UniSim Design. Accurate prediction of the thermodynamic properties of natural gas and refrigerants with the change of working conditions, such as density, specific heat capacity, enthalpy, and entropy, is of significant importance for the simulation of natural gas liquefaction processes. There are many types of property methods embedded in simulation software. Because each property method achieves good performance in a certain range of working conditions, it is crucial to choose the proper method to conduct the simulation. The Soave–Redlich–Kwong equation, the Peng–Robinson equation and the Lee–Kesler–Plocker equation are the main calculation models for physical parameters in natural gas liquefaction processes. According to a literature review, the GERG-2008 equation shows high precision in calculating the thermodynamic properties and phase equilibrium of natural gas and similar mixtures in a wide range of temperature and pressure. Based on accurate experimental data, a comprehensive comparison and analysis among these equations is conducted in this paper. The GERG-2008 equation is recommended as the basis for the calculation of the physical parameters in natural gas liquefaction processes.

1. Introduction

An expanding population and economic growth are the main causes for the growth of global energy consumption (Kumar et al., 2011). According to the U.S. Energy Information Administration (2013), the world total energy consumption demand will increase from 5.528×10^{17} kJ in 2010 to 6.646×10^{17} kJ in 2020 and to 8.651×10^{17} kJ in 2040, with a 30-year increase of 56 percent. With the increasing environmental problems raised by the traditional petrochemical resources and the strong demand of alternative energy for global economic developments, natural gas continues to be favored, with the characteristics of abundant resources and robust production (Yuan et al., 2014). Although the energy market condition varies in different areas of the world, natural gas will represent prosperity and development because of its flexibility and environmental benefits.

Liquefied natural gas (LNG) is a safe and economic means of bringing natural gas to a potential market (Khan et al., 2014) for marginal oil and gas recovery. Design and optimization of natural gas liquefaction processes are target issues of academic research and engineering practice, in which process simulation software is an important tool. With the development of computer technology, many types of software can be used to simulate the liquefaction process, including Aspen HYSYS, Aspen Plus, SIMSCI PRO-II and Honeywell UniSim Design. During the process of compression,

Abbreviations: LNG, liquefied natural gas; SRK, Soave–Redlich–Kwong; PR, Peng–Robinson; LKP, Lee–Kesler–Plocker; RK, Redlich–Kwong; AD, absolute deviation; AAD, average absolute deviation; RD, relative deviation; ARD, average relative deviation.

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throttling and heat transfer, the phase state of natural gas and refrigerants varies, resulting in the change of the physical properties, an accurate prediction of which is essential for process simulation and optimization. Extensive theoretical and experimental research studies have been performed to determine the physical properties of natural gas, among which the equations of state are preferred. There are many types of equation-of-state based property methods embedded in simulation software, and different thermodynamic methods have been employed to conduct the liquefaction processes. Because each property method achieves good performance over a certain range of working conditions, it is crucial to choose the proper method to simulate the process, that is, it is necessary to compare the prediction accuracy of different models for determining the physical parameters.

In the simulation and optimization of natural gas liquefaction processes, the Soave–Redlich–Kwong (SRK) equation, the Peng–Robinson (PR) equation and the Lee–Kesler–Plocker (LKP) equation are the main models for predicting the physical parameters. To determine the refrigerant compositions and operation conditions of a mixed refrigerant cascade cycle, a synthesis problem was posed by a nonconvex nonlinear program (Vaidyaraman and Maranas, 2002) in which the SRK equation of state was used for calculations within the modules. Aspelund et al. (2007) described a new methodology for process synthesis, combining the traditional pinch analysis with exergy calculations via Aspen HYSYS using the SRK equation of state. Using the PR equation of state for physical properties calculations, Nogal et al. (2008) presented a new approach for the optimal design of mixed refrigerant cycles. Shirazi and Mowla (2010) conducted a research study on the selection and development of gas peak shaving processes for the lowest energy consumption based on the use of the PR equation to calculate the thermodynamic properties. To optimize the verified APCI LNG plant model, Alabdulkarem et al. (2011) explored the use of Aspen HYSYS as the thermodynamic model and Matlab as the optimizer by selecting the PR equation of state to model the property of substances. A comparison of the natural gas liquefaction processes with different precooling cycles was performed by Castillo et al. (2013) using Aspen HYSYS with the Peng–Robinson thermodynamic fluid package. Using the PR equation to calculate the thermodynamic properties, Khan and Lee (2013) optimized a single mixed refrigerant natural gas liquefaction process using Honeywell UniSim Design and the particle swarm paradigm. With the PR equation of state calculating the thermodynamic properties, a typical single mixed refrigerant with low energy consumption was analyzed to determine the optimum operating conditions (Moein et al., 2015). Cao et al. (2006) designed and simulated two typical types of small-scale natural gas liquefaction processes using Aspen HYSYS, in which the PR equation and the LKP equation were selected for the fluid package. Yuan et al. (2015) proposed a novel process to condense the double-stage pre-cooled and compressed BOG at LNG Terminals, where the SRK equation was used to calculate the phase equilibrium and the LKP equation was used to calculate the enthalpy and entropy.

Although these equations are widely used to simulate natural gas liquefaction processes, they still have some defects in predicting the liquid density and phase equilibrium (Kunz and Wagner, 2012). Thus, modifications of the liquid molar volume prediction have been made to improve the accuracy of the liquid density prediction. Dauber and Span (2012) conducted a study of the comparison of the liquid density and the isobaric heat capacity based on the standard SRK equation, the standard PR equation, the standard LKP equation and the GERG-2008 equation, in which significant deviations at low temperature were observed. The author made a great improvement on applying the GERG-2008 equation to the simulation of the liquefaction processes. However, the natural gas liquefaction process is a complicated thermal system that is also affected by other important physical properties, such as the gas density, enthalpy, as well as phase equilibrium parameters. Based on accurate experimental data and the Aspen Plus software (Aspen Technology, 2013), a comprehensive comparison and analysis among the modified SRK equation, the modified PR equation, the modified LKP equation and the GERG-2008 equation is conducted to predict the gas and saturated liquid densities, specific heat capacities, enthalpies, dew points and phase equilibrium parameters to provide a reference for the selection of property methods in the simulation of the liquefaction processes.

2. Equations of state

The equation of state for real gas, which was proposed by van der Waals in 1873, acted as the basic form of the cubic equations of state. Redlich and Kwong (RK) (1949) proposed a modified cubic equation of state with two individual coefficients, which provided satisfactory results above the critical temperature for any pressure. Reid introduced binary interaction parameters into the RK equation for calculating mixtures. To compensate for the defects of the RK equation in predicting the saturated pressure of pure substances and multi-component phase equilibrium, Soave (1972) improved the RK equation by introducing a third parameter with an acentric factor to obtain the Soave–Redlich–Kwong (SRK) equation. Peng and Robinson (1976) developed a new two-constant equation of state, which showed great advantages in the prediction of liquid phase densities.

The corresponding state principle is one of the methods for calculating the thermodynamic properties. Taking the critical point as the reference point, van der Waals proposed the principle of corresponding states in 1880, where a general form of the equation of state can be obtained by expressing the temperature, pressure and volume as a monotonic function of the respective critical parameter. Pitzer et al. (1955) introduced the acentric factor to improve the prediction accuracy of calculating the volume and the thermodynamic parameters. Leach et al. (1968) introduced the molecular shape factors into the pseudo-critical equations, which greatly improved the calculation of the vapor–liquid equilibrium for nonpolar hydrocarbon mixtures. Based on the 3-parameter corresponding states principle, Lee and Kesler (1975) developed an analytical correlation to facilitate processing using a computer, which was applied to mixtures for thermodynamic properties calculation (Plocker et al., 1978) to obtain the Lee–Kesler–Plocker equation.

As the expanded version of the GERG-2004 equation, the GERG-2008 equation (Kunz and Wagner, 2012) shows a good prediction performance over the gas phase, liquid phase, supercritical region, and vapor–liquid equilibrium states for 21 mixtures, including methane, nitrogen, carbon dioxide, ethane, propane, n-butane, isobutane, n-pentane, isopentane, n-hexane, n-heptane, n-octane, n-nonane, n-decane, hydrogen, oxygen, carbon monoxide, water, hydrogen sulfide, helium, and argon. The Aspen Plus software provides a variety of thermodynamic calculation methods for phase equilibrium, enthalpy, entropy, density, and other parameters, which is employed as the platform for accuracy analysis of the prediction models. For the SRK equation, the PR equation and the LKP equation, the binary parameters of Knapp et al. (1982) are used.

2.1. Soave–Redlich–Kwong equation of state

The Soave–Redlich–Kwong equation of state shows good performance in simulations of hydrocarbon processing, including gas treatment and refinery and petrochemical processes; the equation of state is given below: