Interfacial Tension and Phase Behaviour of Methane + Propane + Heptane Mixtures

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Predictive models for the interfacial tension and Equations of State (EOS) for calculating phase equilibrium in natural gas mixtures need to be validated by accurate experimental data. In this work, a differential capillary rise apparatus including a high-pressure visual sapphire equilibrium cell was used to measure mixture interfacial tension and VLE of binary mixtures of methane and propane and ternary mixtures of (methane + propane + n-heptane) along isotherms from 203.15 K to 363.15 K at pressure up to 24 MPa. Measurements were conducted by injecting metered amounts of methane at constant temperature into known amounts of propane (and propane + heptane) within the sapphire cell, raising the mixture’s pressure along the mixture’s bubble-point curve at constant temperature until the critical point was reached. The equilibrium phase compositions were measured using a gas chromatograph and interfacial tension was determined by measuring liquid heights in the capillaries and using the differential capillary rise method. Critical points were determined from observation of critical opalescence and/or by using a critical scaling method based on the measured VLE data. The measured interfacial tensions were compared with predictions of various models, including the Parachor method and Linear Gradient Theory (LGT). The Parachor method was able to describe the data within their uncertainty whereas LGT did not perform as well with systems containing heptane. The measured VLE data and critical points were compared with different EOS including the GERG-2008 and advanced Peng Robinson models. Those equations of states were found to have similar deviations from the data at conditions far from the critical point, while at conditions close to the critical point, the models deviated in quite different ways from the data.