Evaluation of Two Different Liquefaction Procedures for the Measurement of Thermophysical Properties of Cryogenic Liquid Mixtures

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The experimental determination of accurate thermophysical properties (e.g., density or vapor pressure) of fluid mixtures requires exact knowledge of the sample composition. When considering cryogenic liquid mixtures, e.g., liquefied natural gas (LNG), often reference gas mixtures of well-known composition are being liquefied. In order to achieve small uncertainties for the properties measured of a liquefied sample, the composition of the reference gas mixture must be preserved during the liquefaction. For this purpose, the procedure of a supercritical liquefaction in conjunction with a special vapor-liquid-equilibrium (VLE) cell avoids a change in composition and, thus, provides a metrologically traceable composition of the liquefied sample. Since this is a demanding high-pressure application, we investigated the possibility of low-pressure condensation for liquefaction. This technique not only imposes fewer requirements on the apparatus but also allows to study mixtures that cannot easily be liquefied from the supercritical state, e.g., due to the course of the vapor-liquid phase boundary.

Based on experimental investigations and VLE calculations, preservation of the composition of the liquid sample produced during condensation was evaluated. For this purpose, various reference gas mixtures were liquefied both, supercritically and through condensation, utilizing a reference single-sinker magnetic-suspension densimeter for cryogenic liquid mixtures. By comparing the measured densities, it can be determined whether the composition changes significantly when condensation for liquefaction is applied. We observed that, under certain circumstances, no detectable change in composition occurs; thus, this technique provides liquefied samples in a relatively easy manner. However, it was also found that this technique is very sensitive and that even small flaws in sample handling and apparatus operation can cause a non-negligible distortion of the liquid sample, both during the liquefaction and during preservation of the liquefied sample under study.