Dielectric Permittivity Measurements of Alternative Refrigerants and their Mixtures

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A growing need for low-global-warming-potential (GWP) refrigerants that comply with new EU standards has motivated the study of "4th generation" refrigerants, including fluorinated hydrocarbons known as hydrofluoroolefins (HFOs), to replace existing refrigerants such as R134a. Although the outlook for HFOs is promising in terms of low environmental impact, extensive characterization of their thermophysical properties is required for them to be effectively implemented in existing systems and to optimize new designs. This work has focused on measuring the dielectric permittivity of two HFOs, R1234ze and R1234yf, and their equimolar mixture using a microwave re-entrant cavity resonator at pressures up to 11 MPa and temperatures from (248 to 365) K. For the pure refrigerants, the molecular polarizability, dipole moment and second dielectric virial coefficient were determined from the measured dielectric permittivity, densities from the reference equations of state, and the Kirkwood expressions. The measured effective dipole moments were compared with ab initio calculations by Laesecke et al. for the isolated molecules, and are consistent within the experimental uncertainty of the vapor phase measurements. In the liquid phase, the experimental results show that the effective dipole moments are significantly lower than predicted for the isolated molecules, with Kirkwood g factors below one indicating a tendency for antiparallel dipole-dipole alignments for both refrigerants in the liquid phase. Measurements of the dielectric permittivity for the liquid binary mixture were compared with values predicted using polarizability correlations for the pure components and the polarizability mixing rule of Harvey and Prausnitz. The predicted mixture polarizabilities differed from those calculated from the measured permittivities and EOS densities by between 3 and 8 %, which is significantly larger than the estimated uncertainty of the mixture EOS. This indicates that while the mixing rule of Harvey and Prausnitz has been demonstrated to work well for gaseous mixtures including those with a polar compound, the assumption of ideal mixing breaks down in liquid mixtures of polar compounds.