SURFACE TENSION OF SOLUTIONS OF LIQUEFIED GASES: EXPERIMENT, THEORY, AND SIMULATION

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The differential capillary method has been used to measure the capillary constant a^2 and determine the surface tension σ of binary solutions of liquefied gases with complete (CH_4-N_2, O_2-N_2) and partial (CH_4-He, C_2H_6-He, C_3H_8-He, CH_4-H_2, C_2H_6-H_2) solubility of the components. Experiments were conducted in the temperature range from the triple point to temperatures close to the critical point of the solvent at pressures from that of saturation of the pure solvent to 4 MPa. The error of the data obtained was 0.5-2.0 percent, where the upper value refers to the vicinity of the critical point. The temperature, baric and concentration dependences of a^2 and σ are presented in analytical form.

Experimental data are interpreted in the framework of the finite thickness layer method and the van der Waals capillarity theory. The results of molecular dynamics simulation have been used to determine the excess free energy of a homogeneous fluid. Equations of state have been set up for solutions of the liquefied gases under investigation with the use of a one-liquid model. The influence coefficient in the functional of the free energy of an inhomogeneous system has been determined from data on the surface tension at a plane interface and an equation of state describing stable and metastable solution regions. The adsorption and the effective thickness of the surface layer have been calculated, the positions of the dividing surfaces in the interfacial layer have been determined.

The possibility of describing the surface properties of gas-saturated solutions in the framework of the generalized law of corresponding states is considered. It is shown that the saturation of hydrocarbons of the methane series with helium leads to changes in the character of the temperature dependence of the solution surface tension. If for solutions of helium in methane the derivative $(d\sigma/dp)_T$ is negative in the whole temperature range from the triple point to temperatures close to the critical point, for a propane-helium system the derivative $(d\sigma/dp)_T$ changes its sign from negative to positive even at a temperature close to that of the solvent normal boiling.

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