THE PROBLEM

The rapid growth of the natural gas industry and greater emphasis on petrochemical processes has increased interest in the separation of natural gases into component streams of high purity. Of the separation processes studied, those which would be carried out at low temperatures seem the most promising;[17] however, low-temperature phase equilibrium and thermodynamic data necessary for the engineering design of separation plants are incomplete.[15]

To help supply the needed data, the Institute of Gas Technology began, in November 1947, as a part of its basic research program, study of the liquid-vapor phase equilibria, pressure-volume-temperature relationships, and determination of the thermodynamic properties of selected mixtures of natural gas components at low temperatures.

Since the inception of this program, data on liquid-vapor phase equilibria and P-V-T relationships of the methane-nitrogen and methane-ethane binary systems have been obtained and published.[6, 9] The thermodynamic properties of nitrogen[7] and two mixtures of methane and nitrogen[8] have been calculated by use of a modification of the Benedict-Webb-Rubin equation of state. No phase equilibrium data were available for the ethane-nitrogen system, the remaining binary combination of the three important components of natural gas: methane, ethane and nitrogen. Recently Reamer *et al.*[18] published data on the gas-phase P-V-T relationships of ethane-nitrogen mixtures covering temperatures from 40° to 460°F and pressures up to 10,000 psia. These are now the only data available in the literature.

The problem is, therefore, to measure the liquid-vapor phase behavior and the gas-phase pressure-volume-temperature relationships of specific mixtures of ethane and nitrogen over a wide range of concentrations, pressures, and temperatures and to present this information in a form useful for engineering calculations.

THE INSTITUTE OF GAS TECHNOLOGY PHASE EQUILIBRIA LABORATORY

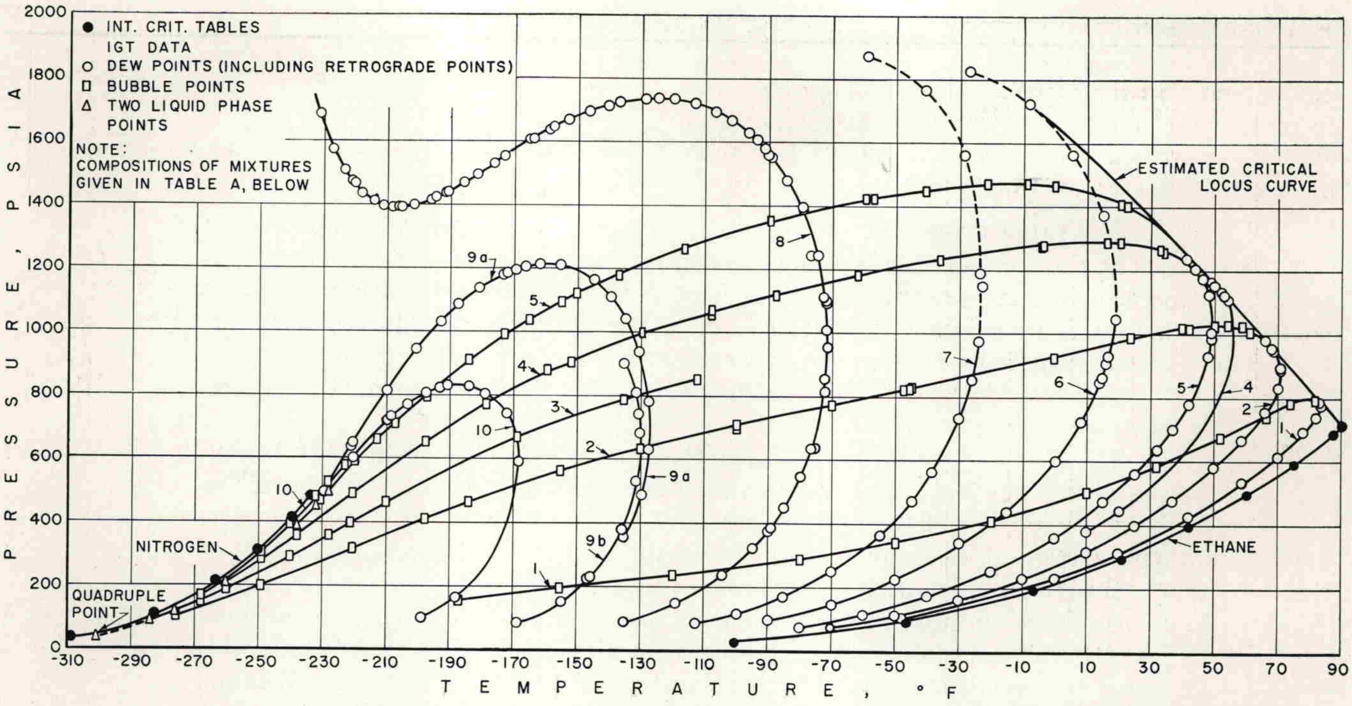

Fig. A.—DEW AND BUBBLE POINT DIAGRAMS FOR THE ETHANE-NITROGEN SYSTEM

SUMMARY OF RESULTS

I. Liquid-Vapor Phase Equilibrium Data

A. Dew and Bubble Points

The dew and bubble point pressure-temperature diagrams, or phase boundary curves, of six mixtures of ethane and nitrogen, dew point curves of three additional mixtures, and the bubble point curve of one other mixture were determined. These constitute the primary phase equilibria data. The compositions of the mixtures investigated are given in Table A. The experimental dew and bubble point data are presented in Table I of **Tabulated Data** and shown graphically in Fig. A.

Each of the curves for ethane-nitrogen mixtures 1, 2, 4, 5, 9 and 10, Fig. A, forms a loop. The lower segment of each loop represents the conditions of pressure and temperature at which the mixture in the vapor phase begins to condense upon cooling or compression and is thus the *dew point curve*. The upper segment of the loop, or *bubble point curve*, represents either the completion of condensation of the vapor, or the boiling point of the liquid. These curve segments join at the critical point of the mixture where the properties of the saturated liquid and vapor phase become indistinguishable.

All of the mixtures tested exhibit retrograde[14] phenomena. The 15% ethane-85% nitrogen mixture has a double retrograde[23] region, and does not have a normal bubble point curve, as certain ethane-nitrogen mixtures are not completely miscible under liquid conditions. Mixtures containing less than 31.7% nitrogen show partial miscibility in the liquid region through certain low-temperature intervals by the formation of two distinct liquid phases, one containing about 70% and the other about 5% ethane. However, the 2% ethane-98% nitrogen mixture shows normal dew and bubble point behavior. Due to the large retrograde region

Table A.—CALCULATED COMPOSITIONS OF MIXTURES INVESTIGATED FOR DEW AND BUBBLE POINT DETERMINATIONS

Mixture No.	Fig. A Curve No.	Ethane Mole %	Nitrogen Mole %
A	1	95.02	4.98
B	2	84.99	15.01
C	3	79.98	20.02
D_1	4	75.07	24.93
D_2	4	75.00	25.00
E_1	5	68.31	31.69
E_2	5	68.28	31.72
F	6	49.82	50.18
G	7	30.00	70.00
H_1	8	15.03	84.97
H_2	8	14.94	85.06
J_1	9a	5.10	94.90
J_2	9b	4.95	95.05
K	10	1.98	98.02