



## Measurement and correlation of the ( $p$ , $\rho$ , $T$ ) relation of ethane II. Saturated-liquid and saturated-vapour densities and vapour pressures along the entire coexistence curve

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Comprehensive and accurate measurements of the saturated-liquid and saturated-vapour densities together with the vapour pressure of pure ethane were carried out from the temperature  $T = 91$  K (triple-point temperature  $T_t = 90.368$  K) to 0.022 K below the critical temperature. Typical values of the total uncertainties of the measurements are:  $\pm 0.01\%$  for the vapour pressures,  $\pm 0.015\%$  for the saturated-liquid densities, and  $\pm 0.02\%$  for the saturated-vapour densities. The critical parameters ( $T_c = 305.322$  K;  $\rho_c = 206.18 \text{ kg} \cdot \text{m}^{-3}$ ,  $p_c = 4.8722$  MPa) and the isothermal compressibilities in the critical region close to the phase boundary have also been determined from these measurements. Comparisons with experimental results of other workers are presented. Using the new data of this work, new correlation equations for the vapour pressure, the saturated-liquid density, and the saturated-vapour density have been developed. © 2002 Elsevier Science Ltd. All rights reserved.

KEYWORDS: ethane; vapour pressures; saturation densities; critical constants; two-sinker densimeter

### 1. Introduction

This paper is the second part of a project which deals with the ( $p$ ,  $\rho$ ,  $T$ ) behaviour of pure ethane. The first part,<sup>(1)</sup> in which the reasons for the new comprehensive measurements are given, describes the experimental results in the homogeneous gas and liquid region including a large part of the critical region in the temperature range from 95 K to 340 K at pressures up to 12 MPa. This second paper reports measurements of the saturated-liquid and saturated-vapour densities and of the vapour pressure of ethane along the entire coexistence curve from  $T = 91$  K (triple-point temperature  $T_t = 90.368$  K) up to slightly

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below the critical temperature  $T_c = 305.322$  K. The aim of this work is to provide a comprehensive and reliable set of measurements which accurately describe the coexistence curve and allow the development of new correlation equations of the thermal saturation properties.

The experimental results described in reference 1 and in this work cover the essential part of the  $(p, \rho, T)$  surface of ethane. Using these new results and the accurate measurements of other workers, a new equation of state of ethane is being developed within our group by Bücker and Wagner. This equation will describe the entire thermodynamic surface of ethane within the experimental uncertainty of the most accurate values of the various thermodynamic properties, and will therefore serve as a reference equation of state.

## 2. Experimental

The design of the densimeter for accurate  $(p, \rho, T)$  measurements in the entire fluid region of pure substances has been described in a previous paper.<sup>(2)</sup> The apparatus covers a density range from  $1 \text{ kg} \cdot \text{m}^{-3}$  to  $2000 \text{ kg} \cdot \text{m}^{-3}$  at temperatures from 60 K to 340 K and pressures up to 12 MPa. The method used for density measurements (“Two-Sinker-Method”) was briefly outlined in the first part of this work.<sup>(1)</sup> Nevertheless, in order to understand the measurements reported in this paper, the operating procedure for determining saturated-vapour and saturated-liquid densities is briefly explained in this section.

As in the case of methane,<sup>(2,3)</sup> carbon dioxide,<sup>(4)</sup> argon,<sup>(5)</sup> ethylene,<sup>(6)</sup> nitrogen,<sup>(7)</sup> and sulphur hexafluoride,<sup>(8)</sup> the saturated-vapour densities of ethane along the largest part of the coexistence curve were determined as follows. In the upper part of the measuring cell (see the schematic drawing in reference 2), a specially designed small reference cell was integrated. It was filled with ethane in such a way that the fluid was in (vapour + liquid) phase equilibrium. The measuring cell was filled with gas at pressures closely below the vapour pressure. By means of a differential-pressure indicator (uncertainty  $< \pm 10$  Pa) the pressure difference between the measuring cell and the reference cell could be measured accurately. With this arrangement, three or more  $(p, \rho, T)$  values were measured along the corresponding isotherms at pressures closely below the vapour pressure  $p_\sigma$  [i.e., pressure difference  $(p - p_\sigma) = -50$  hPa to  $-10$  hPa]. Outside the critical region, the saturated-vapour densities were determined by extrapolating the measured values to the vapour pressure by means of a linear equation fitted to the  $(p, \rho, T)$  values. In the critical region, however, a linear extrapolation is no longer suitable for the curved isotherms. Therefore, for  $T \geq 303.5$  K, several densities were measured on the corresponding isotherms at pressure differences  $(p - p_\sigma)$  between the pressure  $p$  in the measuring cell and the vapour pressure  $p_\sigma$  in the reference cell ranging from about  $-43$  hPa to  $-0.6$  hPa. The saturated-vapour densities were then determined by extrapolating these isothermal  $(p, \rho, T)$  values to  $(p - p_\sigma) = 0$  using the equation:

$$|p - N_1| = N_2 \cdot |\rho - N_3|^d, \quad (1)$$

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