ACKNOWLEDGMENT

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# Viscosity of Ternary Liquid Mixtures

R. KALIDAS and G. S. LADDHA
Alagappa Chettiar College of Technology, University of Madras, Guindy, Madras 25, India

**The three-body ternary model proposed by Chandramouli and Laddha to predict viscosities of ternary liquid solutions has been used to analyze the experimental data for acetone–methanol–ethylene glycol system. The experimental data are well correlated.**

THE IMPORTANCE of accurate viscosity data to the chemical engineer cannot be overemphasized. Viscosities of liquid mixtures and the variation of viscosity with temperature are required for solution of many problems concerning heat transfer, mass transfer, and fluid flow. Few investigations have been reported for prediction of viscosities of liquid mixtures from the properties of the pure components (*2, 7, 9*). In most of the liquid mixtures there is no linear relationship between viscosity and composition of the liquid mixture. The equation given by McAllister (*5*) on the basis of Eyring's theory of absolute reaction rates (*3*) correlates the viscosities of binary liquid mixtures with the viscosities of pure components satisfactorily. Chandramouli and Laddha (*1*) have proposed a theoretical equation which is an extension of McAllister's three-body model for binary liquid mixtures to ternary liquid mixtures. The equation fits the published data of Ramanujam and Laddha (*8*) for the system ethanol-water-ethylene glycol.

The prediction of viscosities of these binary and ternary liquid mixtures-acetone–methanol, methanol–ethylene glycol, acetone–ethylene glycol, and acetone–methanol–ethylene glycol—was undertaken at 30° C. McAllister's equation was used to correlate the data of binary mixtures. The equation proposed by Chandramouli and Laddha for ternary liquid systems correlate the data satisfactorily for the acetone–methanol–ethylene glycol ternary system.

## McALLISTER'S BINARY MODEL

McAllister's theory of viscosity of liquid mixtures is based on Eyring's theory of absolute reaction rates. Assuming that for liquid mixtures the free energies of activation for viscosity are additive on a mole fraction basis, McAllister proposed the following equation for three-bodied interactions.

$$\log \nu = X_1^3 \log \nu_1 + 3X_1^2X_2 \log \nu_{12} + 3X_1X_2^2 \log \nu_{21} + X_2^3 \log \nu_2 - \log\left[X_1 + X_2 \frac{M_2}{M_1}\right] + 3X_1^2X_2 \log\left[\frac{2+M_2/M_1}{3}\right] + 3X_1X_2^2 \log\left[\frac{1+2M_2/M_1}{3}\right] + X_2^3 \log\left(\frac{M_2}{M_1}\right) \quad (1)$$

Equation 1 contains only two undetermined constants, $\nu_{12}$ and $\nu_{21}$ at each temperature level and considers the possibility of having a maximum, a minimum, neither, or both for $\nu$, the viscosity of the binary liquid mixture.

## TERNARY MODEL

In a study of the viscosity of a ternary mixture, molecules of types 1, 2, and 3 and their different interactions are considered. These are represented in Figure 1. Only three-bodied interactions in a single plane were considered

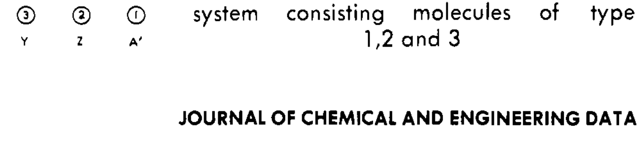

Figure 1. Interaction model for ternary system consisting molecules of type 1,2 and 3

and, arbitrarily, the three types of molecules did not differ in size (radius) by more than a factor of 1.5 with respect to each other. The three-body model shown in Figure 1 describes the situation for the acetone–methanol–ethylene glycol system studied in this investigation. The ratio of molar volumes for this system is 1.3695 to 1.00 to 1.8165 resulting in a ratio of the radii of approximately 1.11 to 1.00 to 1.22 (Table I).

Each type of interaction would correspond to a characteristic free energy of activation $\Delta G^*$. In the case of pure component 1, the type of interaction A (Figure 1) would correspond to $\Delta G_1^*$. Similarly, for pure component 2 (Figure 1, B) and for pure component 3 (Figure 1, C) the free energies of activation would correspond to $\Delta G_2^*$ and $\Delta G_3^*$, respectively.

Two molecules of type 1 and one molecule of type 2 take part in the interaction of type D, E, and F. The free energy of activation for type D is referred to as $\Delta G_{121}^*$. The free energy of activation for types E and F are identical and may be referred to as $\Delta G_{112}^*$.

The energy of activation for various types of interactions is designated in the above way. Table II consists of the various types of interactions and their corresponding energies of activation. Types V, W, X, Y, Z, and A′, involving the interactions of one molecule each of 1, 2, and 3, can be reasonably expected to have identical activation energies denoted by $\Delta G_{123}^*$. The occurrences of the various types of interactions between the molecules comprising the ternary liquid system are listed in Table II. The free energy of activation of the mixture is given by the following expression:

$$\Delta G^* = X_1^3\Delta G_1^* + X_2^3\Delta G_2^* + X_3^3\Delta G_3^* + X_1^2X_2\Delta G_{121}^* + 2X_1^2X_2\Delta G_{112}^*$$
$$+ X_1^2X_3\Delta G_{131}^* + 2X_1^2X_3\Delta G_{113}^* + X_2^2X_1\Delta G_{212}^* + 2X_2^2X_1\Delta G_{221}^*$$
$$+ X_2^2X_3\Delta G_{232}^* + 2X_2^2X_3\Delta G_{223}^* + X_3^2X_1\Delta G_{313}^* + 2X_3^2X_1\Delta G_{331}^*$$
$$+ X_3^2X_2\Delta G_{323}^* + 2X_3^2X_2\Delta G_{332}^* + 6X_1X_2X_3\Delta G_{123}^* \qquad (2)$$

Owing to the difficulty in differentiating between $\Delta G_{121}^*$ and $\Delta G_{112}^*$, and similar terms, the following additional assumptions are made:

$$\Delta G_{12}^* = (\Delta G_{121}^* + 2\Delta G_{112}^*)/3 \qquad (3)$$

$$\Delta G_{13}^* = (\Delta G_{131}^* + 2\Delta G_{113}^*)/3 \qquad (4)$$

$$\Delta G_{21}^* = (\Delta G_{212}^* + 2\Delta G_{221}^*)/3 \qquad (5)$$

$$\Delta G_{23}^* = (\Delta G_{232}^* + 2\Delta G_{223}^*)/3 \qquad (6)$$

$$\Delta G_{31}^* = (\Delta G_{313}^* + 2\Delta G_{331}^*)/3 \qquad (7)$$

$$\Delta G_{32}^* = (\Delta G_{323}^* + 2\Delta G_{332}^*)/3 \qquad (8)$$

Equation 2 is simplified to

$$\Delta G^* = X_1^3\Delta G_1^* + X_2^3\Delta G_2^* + X_3^3\Delta G_3^* + 3X_1^2X_2\Delta G_{12}^*$$
$$+3X_1^2X_3\Delta G_{13}^* + 3X_2^2X_1\Delta G_{21}^* + 3X_2^2X_3\Delta G_{23}^*$$
$$+ 3X_3^2X_1\Delta G_{31}^* + 3X_3^2X_2\Delta G_{32}^* + 6X_1X_2X_3\Delta G_{123}^* \qquad (9)$$

Each type of energy of activation can be related to its corresponding kinematic viscosity by

$$\nu = (hN/M)\, e^{\Delta G^*/RT} \qquad (10)$$

Table II lists the various types of energies of activation considered here and the corresponding kinematic viscosities.

For interactions involving two molecules of type 1 and one molecule of type 2, the average molecular weight may be computed as indicated below:

$$M_{12} = (2M_1 + M_2)/3 \qquad (11)$$

Similarly,

$$M_{13} = (2M_1 + M_3)/3 \qquad (12)$$

$$M_{21} = (2M_2 + M_1)/3 \qquad (13)$$

$$M_{23} = (2M_2 + M_3)/3 \qquad (14)$$

$$M_{31} = (2M_3 + M_1)/3 \qquad (15)$$

$$M_{32} = (2M_3 + M_2)/3 \qquad (16)$$

For the interaction involving one molecule of each of type 1, 2, and 3

$$M_{123} = (M_1 + M_2 + M_3)/3 \qquad (17)$$

and

$$M_{\text{av.}} = (X_1M_1 + X_2M_2 + X_3M_3) \qquad (18)$$

Substituting the value of $\Delta G^*$ from Equation 9 into Equation 10, taking logarithms, eliminating the free energies of activation (Table III), and substituting for the various molecular weights in terms of the molecular weights

Table I. Size Ratios of Systems Studied

| System | Volumetric Size Ratio (Molal) | Volumetric Size Ratio (Radii) |
|---|---|---|
| Acetone–methanol | 1/1.8165 | 1/1.22 |
| Methanol–ethylene glycol | 1/1.3695 | 1/1.015 |
| Acetone–ethylene glycol | 1/1.326 | 1/1.0985 |
| Acetone–methanol–ethylene glycol | 1.3695 : 1.00 : 1.8165 | 1.11 : 1.00 : 1.22 |

Table II. Free Energy of Activation and Fraction of Total Occurrences for All the Types of Interactions

| No. | Type of Interaction | Free Energy of Activation | Fraction of Total Occurrences |
|---|---|---|---|
| 1 | 1-1-1 | $\Delta G_1^*$ | $X_1^3$ |
| 2 | 2-2-2 | $\Delta G_2^*$ | $X_2^3$ |
| 3 | 3-3-3 | $\Delta G_3^*$ | $X_3^3$ |
| 4 | 1-2-1 | $\Delta G_{121}^*$ | $X_1^2X_2$ |
| 5 | 1-1-2<br>2-1-1 | $\Delta G_{112}^*$ | $2X_1^2X_2$ |
| 6 | 1-3-1 | $\Delta G_{131}^*$ | $X_1^2X_3$ |
| 7 | 1-1-3<br>3-1-1 | $\Delta G_{113}^*$ | $2X_1^2X_3$ |
| 8 | 2-1-2 | $\Delta G_{212}^*$ | $X_2^2X_1$ |
| 9 | 2-2-1<br>1-2-2 | $\Delta G_{221}^*$ | $2X_2^2X_1$ |
| 10 | 2-3-2 | $\Delta G_{232}^*$ | $X_2^2X_3$ |
| 11 | 2-2-3<br>3-2-2 | $\Delta G_{223}^*$ | $2X_2^2X_3$ |
| 12 | 3-1-3 | $\Delta G_{313}^*$ | $X_3^2X_1$ |
| 13 | 3-3-1<br>1-3-3 | $\Delta G_{331}^*$ | $2X_3^2X_1$ |
| 14 | 3-2-3 | $\Delta G_{323}^*$ | $X_3^2X_2$ |
| 15 | 3-3-2<br>2-3-3 | $\Delta G_{332}^*$ | $2X_3^2X_2$ |
| 16 | 1-2-3<br>1-3-2<br>2-3-1<br>2-1-3<br>3-1-2<br>3-2-1 | $\Delta G_{123}^*$ | $6X_1X_2X_3$ |

of pure components, the following expression results, upon simplification and rearrangement, in the kinematic viscosity of the mixture:

$$\begin{aligned}\log \nu = {} & X_1^3 \log \nu_1 + X_2^3 \log \nu_2 + X_3^3 \log \nu_3 + 3X_1^2 X_2 \log \nu_{12} \\ & + 3X_1^2X_3 \log \nu_{13} + 3X_2^2X_1 \log \nu_{21} + 3X_2^2X_3 \log \nu_{23} + 3X_3^2X_1 \log \nu_{31} \\ & + 3X_3^2X_2 \log \nu_{32} + 6X_1X_2X_3 \log \nu_{123} - \log (X_1M_1 + X_2M_2 + X_3M_3) \\ & + X_1^3 \log M_1 + X_2^3 \log M_2 + X_3^3 \log M_3 + 3X_1^2X_2 \log (2M_1 + M_2)/3 \\ & + 3X_1^2X_3 \log (2M_1 + M_3)/3 + 3X_2^2X_1 \log (2M_2 + M_1)/3 \\ & + 3X_2^2X_3 \log (2M_2 + M_3)/3 + 3X_3^2X_1 \log (2M_3 + M_1)/3 \\ & + 3X_3^2X_2 \log (2M_3 + M_2)/3 + 6X_1X_2X_3 \log (M_1 + M_2 + M_3)/3 \quad (19)\end{aligned}$$

The equation consists of seven constants comprising of six binary constants and one ternary constant. The three binary systems of components 1 and 2, 2 and 3, and 3 and 1 give the constants $\nu_{12}$ and $\nu_{21}$, $\nu_{23}$ and $\nu_{32}$, and $\nu_{31}$ and $\nu_{13}$, respectively.

Equation 19 contains only one undetermined ternary constant, $\nu_{123}$, for each temperature level.

Table III. Free Energies of Activation and Kinematic Viscosity

| No. | Function of Free Energy of Activation | Kinematic Viscosity, Cs. |
|---|---|---|
| 1 | $(hN/\mathrm{Mav.})\, e^{\Delta G^*/RT}$ | $\nu_{\mathrm{mixture}}$ |
| 2 | $(hN/M_1)\, e^{\Delta G_1^*/RT}$ | $\nu_1$ |
| 3 | $(hN/M_2)\, e^{\Delta G_2^*/RT}$ | $\nu_2$ |
| 4 | $(hN/M_3)\, e^{\Delta G_3^*/RT}$ | $\nu_3$ |
| 5 | $(hN/M_{12})\, e^{\Delta G_{12}^*/RT}$ | $\nu_{12}$ |
| 6 | $(hN/M_{13})\, e^{\Delta G_{13}^*/RT}$ | $\nu_{13}$ |
| 7 | $(hN/M_{21})\, e^{\Delta G_{21}^*/RT}$ | $\nu_{21}$ |
| 8 | $(hN/M_{23})\, e^{\Delta G_{23}^*/RT}$ | $\nu_{23}$ |
| 9 | $(hN/M_{31})\, e^{\Delta G_{31}^*/RT}$ | $\nu_{31}$ |
| 10 | $(hN/M_{32})\, e^{\Delta G_{32}^*/RT}$ | $\nu_{32}$ |
| 11 | $(hN/M_{123})\, e^{\Delta G_{123}^*/RT}$ | $\nu_{123}$ |

## EXPERIMENTAL

Chemically pure acetone, methanol, and ethylene glycol were further purified according to methods recommended by Weissberger and Proskawer (*10*). A set of U-tube viscometers supplied by Technico, conforming to British Standard Specification No. 188 (*4*) were used in the determination of viscosities. The precautions recommended by Mikhail and Kimel (*6*) were followed. The viscosity measurements were made in a constant temperature bath controlled by an electronic unit capable of keeping the temperature constant to within ± 0.05° C.

## RESULTS AND DISCUSSION

Equation 1 was tested with the experimentally obtained data for the viscosity of binary mixtures of acetone–methanol, methanol–ethylene glycol, and acetone–ethylene glycol systems at 30° C. presented in Tables IV, V, and VI. The two binary constants for each of the three binary systems were determined by analysis of respective experimental data by the method of least squares. With these constants, $\nu$ was calculated at each composition. The deviation of the experimental data was calculated as:

$$(\nu \text{ exptl.} - \nu \text{ calcd.})/\nu \text{ calcd.} \times 100 = \text{per cent deviation, } d$$

Table IV. Kinematic Viscosities of Acetone–Methanol Mixtures

| Mole Fraction of Acetone | Kinematic Viscosities, Cs. Exptl. | Calcd. |
|---|---|---|
| 0.0000 | 0.6704 | 0.6704 |
| 0.0450 | 0.6267 | 0.6235 |
| 0.0851 | 0.5982 | 0.6039 |
| 0.1472 | 0.5627 | 0.5649 |
| 0.2077 | 0.5300 | 0.5340 |
| 0.3187 | 0.4862 | 0.4884 |
| 0.3539 | 0.4774 | 0.4789 |
| 0.3950 | 0.4671 | 0.4696 |
| 0.4643 | 0.4532 | 0.4502 |
| 0.5636 | 0.4345 | 0.4301 |
| 0.6658 | 0.4166 | 0.4145 |
| 0.7564 | 0.4056 | 0.4025 |
| 0.7837 | 0.4021 | 0.3992 |
| 0.8606 | 0.3929 | 0.3902 |
| 1.0000 | 0.3728 | 0.3728 |

Table V. Kinematic Viscosities of Methanol–Ethylene Glycol Mixtures

| Mole Fraction of Methanol | Kinematic Viscosities, Cs. Exptl. | Calcd. |
|---|---|---|
| 0.0000 | 12.2470 | 12.2470 |
| 0.0419 | 11.1200 | 11.1300 |
| 0.1045 | 9.8280 | 9.5740 |
| 0.1615 | 8.4730 | 8.3010 |
| 0.1890 | 7.7118 | 7.7290 |
| 0.2058 | 7.3980 | 7.3962 |
| 0.3345 | 5.1297 | 5.1980 |
| 0.4661 | 3.5266 | 3.5470 |
| 0.5893 | 2.4580 | 2.4390 |
| 0.6567 | 1.9790 | 1.9770 |
| 0.7534 | 1.4960 | 1.4600 |
| 0.8371 | 1.1446 | 1.1200 |
| 0.9213 | 0.8485 | 0.8581 |
| 1.0000 | 0.6704 | 0.6704 |

Table VI. Kinematic Viscosities of Acetone–Ethylene Glycol Mixtures

| Mole Fraction Acetone | Kinematic Viscosities, Cs. Exptl. | Calcd. |
|---|---|---|
| 0.0000 | 12.2470 | 12.2470 |
| 0.0640 | 9.3260 | 9.2170 |
| 0.1405 | 6.7700 | 6.5830 |
| 0.2421 | 4.2120 | 4.2520 |
| 0.3628 | 2.5385 | 2.5800 |
| 0.4521 | 1.7792 | 1.8170 |
| 0.5407 | 1.2925 | 1.3074 |
| 0.6655 | 0.8518 | 0.8574 |
| 0.7672 | 0.6338 | 0.6328 |
| 0.8745 | 0.4810 | 0.4806 |
| 1.0000 | 0.3728 | 0.3728 |

The equation for the average deviation is

$$\text{Av. dev.} = (d^2/N)^{1/2}$$

The values of binary constants, the maximum deviation, and the average deviation for the three binary systems studied are listed in Table VII.

Equation 19 was tested using the experimental data for acetone–methanol–ethylene glycol mixture at 30° C. presented in Table VIII and values of the binary constants given in Table VII. The only ternary constant $\nu_{123}$ was determined by the method of least squares. With this value of $\nu_{123}$ the kinematic viscosity of the ternary mixture was calculated for each data point. Equation 19 fits the data with a maximum deviation of ±1.81% and an average deviation of ±0.95%. For the system at 30° C. $\nu_{123}$ has a value of 0.8476 centistokes.

The accuracy with which Equation 19 fits the data for the ternary system acetone–methanol–ethylene glycol supports the proposed model for the estimation of viscosity of ternary liquid system.

Table VII. Binary Constants and Per Cent Deviations of Experimental Data

| | |
|---|---|
| Acetone-methanol system | |
| | $\nu_{12}$ = 0.4269 Cs. |
| | $\nu_{21}$ = 0.4428 Cs. |
| Per cent av. dev. | = ±0.60 |
| Per cent max. dev. | = ±0.97 |
| Methanol-ethylene glycol system | |
| | $\nu_{23}$ = 1.9710 Cs. |
| | $\nu_{32}$ = 5.8750 Cs. |
| Per cent av. dev. | = ±1.51 |
| Per cent max. dev. | = ±2.58 |
| Ethylene glycol-acetone system | |
| | $\nu_{31}$ = 2.7750 Cs. |
| | $\nu_{13}$ = 0.6604 Cs. |
| Per cent av. dev. | = ±1.45 |
| Per cent max. dev. | = ±2.76 |

## NOMENCLATURE

$\Delta G^*$ = molal free energy of activation for viscosity cal./gram mole
$R$ = gas constant, 1.987 cal./gram mole ° K.
$T$ = absolute temperature, ° K.
$N$ = Avogadro number, $6.023 \times 10^{23}$ molecules/gram mole
$h$ = Planck constant, $6.6240 \times 10^{-27}$ erg. - sec./molecule
$M$ = molecular weight, grams/gram mole
$\nu$ = kinematic viscosity, stoke or centistoke

### Subscripts

1 = acetone
2 = methanol
3 = ethylene glycol

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Table VIII. Kinematic Viscosities of Acetone–Methanol–Ethylene Glycol Mixtures

| Mole Fraction of Components | | | Kinematic Viscosities, Cs. | |
|---|---|---|---|---|
| Acetone | Methanol | Ethylene glycol | Exptl. | Calcd. |
| 0.0817 | 0.7878 | 0.1305 | 0.9043 | 0.8941 |
| 0.1902 | 0.6789 | 0.1309 | 0.7807 | 0.7802 |
| 0.3124 | 0.5570 | 0.1306 | 0.6745 | 0.6873 |
| 0.6033 | 0.2659 | 0.1308 | 0.5472 | 0.5571 |
| 0.0740 | 0.6948 | 0.2312 | 1.2453 | 1.2270 |
| 0.1699 | 0.5989 | 0.2312 | 1.0702 | 1.0670 |
| 0.2744 | 0.4949 | 0.2307 | 0.9411 | 0.9387 |
| 0.4021 | 0.3668 | 0.2311 | 0.8159 | 0.8213 |
| 0.5368 | 0.2321 | 0.2311 | 0.7355 | 0.7336 |
| 0.6782 | 0.0908 | 0.2310 | 0.6595 | 0.6655 |
| 0.0674 | 0.6221 | 0.3105 | 1.5772 | 1.5810 |
| 0.1523 | 0.5371 | 0.3106 | 1.4019 | 1.3790 |
| 0.2449 | 0.4440 | 0.3111 | 1.2129 | 1.2140 |
| 0.3578 | 0.3315 | 0.3107 | 1.0643 | 1.0590 |
| 0.4809 | 0.2090 | 0.3101 | 0.9278 | 0.9365 |
| 0.0571 | 0.5679 | 0.3750 | 1.9383 | 1.9580 |
| 0.1371 | 0.4879 | 0.3751 | 1.7315 | 1.7070 |
| 0.2209 | 0.4035 | 0.3756 | 1.5066 | 1.5060 |
| 0.3260 | 0.2969 | 0.3771 | 1.3301 | 1.3170 |
| 0.4318 | 0.1923 | 0.3759 | 1.1556 | 1.1660 |
| 0.5409 | 0.0836 | 0.3756 | 1.0491 | 1.0500 |
| 0.2939 | 0.2615 | 0.4446 | 1.6728 | 1.6540 |
| 0.3818 | 0.1738 | 0.4444 | 1.4637 | 1.4810 |
| 0.4850 | 0.0697 | 0.4453 | 1.3200 | 1.3290 |

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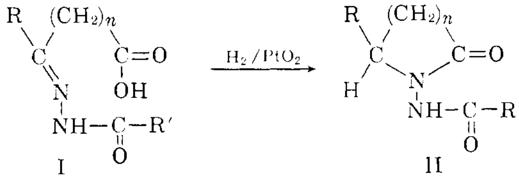
## CORRECTION:

In the article, "Reductive Cyclization of Hydrazones," [J. Chem. Eng. Data 8, 601 (1963)], the product of the reaction in Table I is incorrect. The corrected table is reproduced below.

Table I. Properties of Hydrazones



| | I Hydrazone | | | | II Hydrazine | | Carbon | | Hydrogen | | Nitrogen | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | M.P.[a] ° C. | R | R′ | n | M.P.[a] ° C. | Formula | Calcd. | Found | Calcd. | Found | Calcd. | Found |
| A | 188-189[b] | $CH_3$ | $NH_2$ | 2 | 181-183 | $C_6H_{11}N_3O_2$ | 45.85 | 45.92 | 7.05 | 7.11 | 26.74 | 26.79 |
| B | 143-144 | $CH_3$ | $CH_2$ | 2 | 54-56 | $C_7H_{12}N_2O_2$ | 53.83 | 53.59 | 7.74 | 7.84 | 17.94 | 17.66 |
| C | 111-113 | $CH_3$ | $OC_2H_5$ | 2 | 59-60 | $C_8H_{14}N_2O_3$ | 51.60 | 51.68 | 7.58 | 7.49 | 15.04 | 15.02 |
| D | –[c] | $CH_3$ | $OC_2H_5$ | 3 | 48-50[d] | $C_9H_{16}N_2O_3$ | 53.98 | 54.08 | 8.06 | 8.07 | 13.99 | 14.16 |
| E | 153-155 | $C_6H_5$ | $OC_2H_5$ | 2 | –[e] | $C_{13}H_{16}N_2O_3$ | 62.89 | 63.15 | 6.50 | 6.56 | 11.28 | 11.45 |

[a] Melting points taken on Hoover-Thomas apparatus and are corrected. [b] Reported (5) m.p. 191–2°. [c] Not isolated. [d] B.P. 105–7°/0.1 mm. [e] B.P. 192–5°/1.0 mm.; after standing for 1 year it did not crystallize.