# The Selection of International Standards for the Thermodynamic Properties of Difluoromethane and Pentafluoroethane[1]

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**ABSTRACT**

The Montreal Protocol stipulates that chlorinated hydrofluorocarbons (HCFCs) are to be phased out and replaced by more environmentally acceptable refrigerants. The fluorinated hydrocarbons (HFCs); 1,1,1,2-tetrafluoroethane (R134a), pentafluoroethane (R125) and difluoromethane (R32) and their mixtures are likely replacements. One of the goals of the Annex 18 of the International Energy Agency is to establish standard formulations that best represent the thermodynamic properties of difluoromethane and pentafluoroethane. This paper describes the role of IUPAC Thermodynamic Tables Project Centre in this process. Equations of state for difluoromethane and for pentafluoroethane from independent researchers in Germany , Japan and the United States were reviewed. They represented the most accurate formulations available in December 1996.

The two groups of equations were compared with the experimental data for their respective fluids. The method of assessment included analysis of the experimental data, statistical comparisons of values calculated from the property formulations to experimental values and property plots of the thermodynamic surface. Selected comparisons of all the equations will be shown.

**KEY WORDS:** environmentally acceptable refrigerants; equation of state; international standards; thermodynamic properties

## 1. INTRODUCTION

Annex 18 of the International Energy Agency (IEA) Heat Pump Programme was established to determine international standards for the thermophysical properties of environmentally acceptable refrigerants. Following submission of several proposals, the Annex was approved in 1990, with the United States as operating agent. Other member countries that participate in the Annex are Austria, Canada, Germany Japan, Norway, Sweden, and the United Kingdom.

One goal of the Annex is to determine property formulations that can be considered as international standards for the environmentally acceptable refrigerants. To achieve this , two surveys were conducted by the Annex to assess the current research in the areas of experimental measurement and correlation. A report on the first survey was published in 1991 [1]. An update to this survey was undertaken in late 1992.

Given the importance of 1,1,-dichloro-2,2,2-trifluoroethane (R123) and 1,1,2,2-tetrafluoroethane (R134a), a large quantity of property data had become available in a short time, and consequently a number of equations of state. An evaluation of submitted existing equations of state for both fluids were carried out by independent groups of evaluators in the United States and the United Kingdom. Following the presentation of the final recommendations of the evaluators [2,3], the selected equations of Tillner-Roth and Baehr [4] for R134a and Younglove and McLinden [5] for R123 were published. These equations formed the basis of the international properties bulletins published by the International Institute of Refrigeration.

In 1996, it was agreed that equations of state for the two alternative refrigerants, R32 and R125 should be developed and evaluated. The equations were submitted by June 1995. and the data to be used in the evaluation were deposited in the electronic database at the University of Stuttgart by December 1994. The majority of the data, both published and unpublished, were supplied from a compilation prepared by Outcalt and McLinden from the National Institute of Science and Technology (NIST), Bolder, Colorado, USA. An independent group, the IUPAC Thermodynamic Tables Project Centre, agreed to undertake the evaluations. Four equations of state for R32 and three equations of state for R125 were submitted for evaluation. This paper summarises the procedure used in the selection of the equations of state. Thermodynamic properties, calculated using candidate equations of state compared with experimental data, are summarised.

## 2. ANALYSIS OF EXPERIMENTAL DATA AND EQUATIONS OF STATE

A summary of the experimental data for R32 and R125 is given in Table 1. The sources reporting these data were reviewed and the estimated uncertainties in the measurements were used to establish the quality of the data. The equations of state submitted for evaluation together with the authors' estimated range of applicability are listed in Table 2. The procedure used is similar to that previously described for R134a and R123 [10,2,3]. All experimental data, for each type of property were compared with values calculated from each candidate equation of state, except where the author failed to provide the required functionality. The recommendations of the evaluators to

which equation of state gave the best representation of the highest quality experimental data are based on a report [11]. These contained detailed graphical comparisons for each equation of state with each set of experimental data. In addition, for each equation of state, graphs showing isobars on isobaric heat capacity ($C_p$), isochoric heat capacity ($C_v$), isenthalpic Joule - Thomson coefficient ($(\partial T / \partial p)_H$) and speed of sound ($\omega$) - temperature plots were given. Examples of these plots are given in Figs. 1 and 2 for the equations of state in this study. Plots of this type allow the thermodynamic surface computed from each equation of state to be examined for discontinuities or anomalous behaviour. Statistical comparisons were also made using the absolute average deviation , the bias, the standard deviation, and the root mean square deviation. The results of these statistical comparisons are summarised in Table 3 for R32 and Table 4 for R125.

## 3. SELECTED EQUATIONS OF STATE

Using these techniques, the equations of state recommended by the evaluators as international standards were those of Tillner-Roth [6] for R32 and Ely [10] for R125.

The equation of state for R32 developed by Tillner-Roth [6] is explicit in the dimensionless Helmholtz function and is given as

$$A / RT = A^{ig}(\delta,\tau) / RT + A^{r}(\delta,\tau) / RT \qquad (1)$$

where $\delta = \rho / \rho$ and $\tau = T / T$ and $\rho$ and $T$ are reducing parameters for the equation of state that are close to the critical values. $A^{ig} / RT$ is the ideal gas contribution to the dimensionless Helmholtz function and $A^{r} / RT$ is the residual part. The former is fitted as a simple third degree polynomial in $T_r$ and the latter by

$$A^r / RT = \sum_{i=1}^{8} n_i \omega^{r_i} \tau^{s_i} + \sum_{i=9}^{21} n_i \omega^{r_i} \tau^{s_i} \exp\left(-\omega^{t_i}\right) \tag{2}$$

where all the coefficients, $n_i$, are obtained by fitting to experimental data. All thermodynamic properties can be obtained from this type of equation by differentiation.

Ely [9] also selected a reduced Helmholtz function equation of state of novel form to represent the thermodynamic properties of R125. The ideal gas contribution was represented by an empirically fitted truncated Einstein equation given by

$$C_p^0 / R = 4 + \sum_{i=1}^{3} c_i \left(\Theta_i / T\right)^2 \exp\left(\Theta_i / T\right) / \left(\exp\left(\Theta_i / T\right) - 1\right)^2 \tag{3}$$

where $C_p^0$ is the isobaric ideal gas heat capacity and $c_i$ and $\Theta_i$ are constants derived through fitting.

The reduced residual Helmholtz function was represented by

$$A^r / RT = \sum_{i=1}^{27} n_i \omega^{r_i} \tau^{(s_i + 0..25 u_i)} \exp\left(-\omega^{t_i} - \kappa_{r_i,0}\right) \tag{4}$$

where $\delta = \rho / \rho^c$ and $\tau = T^c / T$, and $\kappa_{r_i,0}$ is the kroneker delta function, defined as $\kappa_{\alpha,\beta} = 1$ if $\alpha = \beta$ otherwise zero.

Details of the development of these equations are to be published.

## 4. CONCLUSIONS

An impartial and extensive evaluation of the equations of state for R32 and R125 has resulted in recommendations by the evaluators to be made to the Annex. For R32, the choice hinged on the better representation of the isochoric liquid phase density, the isochoric heat capacities and the gas phase speed of sound by the Tillner-Roth equation of state. For R125, the evaluators considered that the equation of Ely gave by far the

best representation of the thermodynamic properties. Only the minor issue of the unusual behaviour of the isobaric heat capacity isobars at high pressures (> 50 MPa) and low temperatures (< 200 K) need be considered. However, as this behaviour potentially occurs below the melting curve, which has yet to be fully established at these higher pressures, the evaluators consider this a minor issue.

**ACKNOWLEDGEMENTS**

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of Chemical Engineering and Chemical Technology, Imperial College of Science, Technology and Medicine, London, UK, (1996).

**Table 1** Experimental Data for R32 and R125

| Data type | Points | Range of data | | |
|---|---|---|---|---|
| | | $p$/MPa | $T$/K | $\rho$ / $molm^{-3}$ |
| Difluoromethane (R32) | | | | |
| Saturated vapour pressure | 278 | | 149 - 352 | |
| Saturated liquid density | 107 | | 138 - 352 | |
| Saturated vapour density | 67 | | 219 - 352 | |
| Saturated liquid heat capacity | 95 | | 141 - 343 | |
| $p$-$\rho$-$T$ | 1305 | 0.017 - 72 | 139 - 474 | 6 - 27794 |
| Second virial coefficient | 3 | | 290 - 310 | |
| $C_v$-$\rho$-$T$ | 74 | | 152 - 342 | 17400 - 26800 |
| $C_p$-$p$-$T$ | 13 | 2.1 - 3.0 | 275 - 315 | |
| $\omega$-$p$-$T$ | 99 | 0.019 - 10.4 | 273 - 343 | |
| Pentafluoroethane (R125) | | | | |
| Saturated vapour pressure | 293 | | 195 - 340 | |
| Saturated liquid density | 25 | | 173 - 340 | |
| Saturated vapour density | 8 | | 326 - 340 | |
| Saturated liquid heat capacity | 85 | | 175 - 278 | |
| $p$-$\rho$-$T$ | 967 | 0.018 - 68 | 178 - 449 | 6 - 14159 |
| Second virial coefficient | 23 | | 240 - 400 | |
| $C_v$-$\rho$-$T$ | 97 | | 200 - 342 | 10533 - 13532 |
| $C_p$-$p$-$T$ | 5 | 0.13 | 233 - 334 | |
| $\omega$-$p$-$T$ | 179 | 0.038 - 16.5 | 240 - 400 | |

**Table 2** Equations of State used in this study and their calculated critical values

| Ref | Abbrev. | $p_{max}$/ MPa | $T_{min}$/K to $T_{max}$/K | $T_c$/ K | $p_c$/ MPa | $\rho_c$ / $molm^{-3}$ |
|---|---|---|---|---|---|---|
| | | | Difluoromethane (R32) | | | |
| Tillner-Roth [6] | **TR** | [a] | [a] | 351.351 | 5.793 | 8207.8 |
| Piao and Noguchi [7] | **PN** | 80 | 150 - 500 | [b] | [b] | [b] |
| Outcalt and McLinden [8] | **OM** | 60 | 160 - 500 | [b] | [b] | [b] |
| Ely [9] | **Ely** | [a] | [a] | 351.35 | 5.795 | 7956.2 |
| | | | Pentafluoroethane (R125) | | | |
| Piao and Noguchi [7] | **PN** | 80 | 150 - 500 | [b] | [b] | [b] |
| Outcalt and McLinden [8] | **OM** | 60 | 174 - 500 | [b] | [b] | [b] |
| Ely [9] | **Ely** | [a] | [a] | 339.33 | 3.629 | 4759.9 |

[a] information not supplied by author

[b] information not supplied by author nor calculateable from their supplied programs

**Table 3** Statistical Comparison of Properties calculated using candidate equations of state with experimental data for R32

| | EOS | Points | AAD | Bias | Std Dev | RMS | Max Dev | Bad |
|---|---|---|---|---|---|---|---|---|
| Ideal gas | **Ely** | 82 | 0.06027 | 0.05641 | 0.05240 | 0.07678 | 0.1300 | 1 |
| heat | **OM** | 82 | 0.2525 | 0.1817 | 1.152 | 1.159 | 7.129 | 61 |
| capacity | **TR** | 82 | 0.1977 | 0.05987 | 0.7832 | 0.7807 | 4.801 | 61 |
| Saturated | **Ely** | 278 | 0.1071 | 0.07189 | 0.1493 | 0.1655 | 0.6311 | 0 |
| Vapour | **OM** | 278 | 0.1412 | -0.02083 | 0.2188 | 0.2194 | 1.517 | 0 |
| pressure | **PN** | 278 | 0.1200 | -0.02709 | 0.1833 | 0.1850 | 0.8627 | 0 |
| | **TR** | 277 | 0.08714 | 0.05060 | 0.1865 | 0.1929 | 1.107 | 0 |
| Saturated | **Ely** | 107 | 0.3819 | -0.3082 | 0.6545 | 0.7206 | -4.358 | 2 |
| Liquid | **OM** | 107 | 0.4349 | -0.4106 | 0.5800 | 0.7084 | -3.763 | 2 |
| Density | **PN** | 107 | 0.5374 | -0.2577 | 1.293 | 1.313 | 9.029 | 1 |
| | **TR** | 107 | 0.4891 | -0.2789 | 1.237 | 1.262 | -9.373 | 0 |
| Saturated | **Ely** | 67 | 0.9782 | 0.9696 | 1.203 | 1.538 | 5.840 | 0 |
| Vapour | **OM** | 67 | 1.015 | 0.9911 | 1.159 | 1.518 | 5.339 | 0 |
| Density | **PN** | 67 | 1.511 | 1.511 | 1.621 | 2.207 | 7.390 | 2 |
| | **TR** | 67 | 0.7139 | 0.3464 | 1.138 | 1.181 | 4.681 | 0 |
| Saturated | **Ely** | 95 | 0.7399 | -0.004211 | 1.346 | 1.339 | -7.851 | 0 |
| liquid | **OM** | 95 | 1.087 | -0.7581 | 1.472 | 1.649 | -9.429 | 0 |
| heat | **PN** | 95 | 1.583 | -1.427 | 2.016 | 2.461 | -9.367 | 2 |
| capacity | **TR** | 95 | 0.4946 | -0.2939 | 1.094 | 1.127 | -7.670 | 1 |
| *PVT* | **Ely** | 560 | 0.07755 | -0.03549 | 0.1361 | 0.1405 | 1.124 | 4 |
| (liquid | **OM** | 560 | 0.07164 | -0.02973 | 0.1647 | 0.1672 | -1.536 | 4 |
| phase) | **PN** | 560 | 0.1092 | 0.02947 | 0.2924 | 0.2936 | 5.097 | 0 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| | **TR** | 560 | 0.09293 | -0.01292 | 0.2041 | 0.2044 | 2.389 | 4 |
| *PVT* (gas phase) | **Ely** | 745 | 0.2876 | -0.06335 | 0.8370 | 0.8388 | -6.382 | 2 |
| | **OM** | 745 | 0.3544 | -0.07457 | 0.9497 | 0.9520 | 6.342 | 2 |
| | **PN** | 745 | 0.3530 | -0.04943 | 0.8923 | 0.8930 | 6.384 | 2 |
| | **TR** | 745 | 0.2607 | -0.1002 | 0.7994 | 0.8051 | 6.219 | 2 |
| $C_v$-$\rho$-$T$ (liquid phase) | **Ely** | 74 | 0.4361 | 0.2061 | 0.5130 | 0.5496 | 1.476 | 0 |
| | **OM** | 74 | 0.4557 | 0.2704 | 0.5490 | 0.6086 | 1.359 | 0 |
| | **PN** | 74 | 1.158 | 0.1225 | 1.418 | 1.414 | 3.193 | 0 |
| | **TR** | 74 | 0.3427 | 0.1720 | 0.3952 | 0.4285 | 1.168 | 0 |
| $C_p$-$p$-$T$ | **Ely** | 13 | 0.6172 | -0.6172 | 0.2573 | 0.6648 | -1.032 | 0 |
| | **OM** | 13 | 0.9478 | -0.9478 | 0.4362 | 1.036 | -1.644 | 0 |
| | **PN** | 13 | 1.016 | -1.016 | 0.6396 | 1.187 | -2.056 | 0 |
| | **TR** | 13 | 0.9490 | -0.9490 | 0.3015 | 0.9922 | -1.416 | 0 |
| $\omega$-$p$-$T$ (liquid phase) | **Ely** | 30 | 0.2337 | 0.2093 | 0.1558 | 0.2594 | 0.6914 | 0 |
| | **OM** | 30 | 1.013 | -1.013 | 0.5315 | 1.140 | -2.585 | 0 |
| | **PN** | 30 | 0.5708 | 0.4596 | 0.8695 | 0.9706 | 2.403 | 0 |
| | **TR** | 30 | 0.3462 | -0.3462 | 0.1699 | 0.3844 | -0.6964 | 0 |
| $\omega$-$p$-$T$ (gas phase) | **Ely** | 66 | 0.03481 | -0.03481 | 0.00448 | 0.03509 | -0.04669 | 0 |
| | **OM** | 66 | 0.05513 | -0.05513 | 0.01470 | 0.05703 | -0.08149 | 0 |
| | **PN** | 66 | 0.02364 | -0.02364 | 0.01348 | 0.02716 | -0.06676 | 0 |
| | **TR** | 66 | 0.00502 | 0.000324 | 0.00682 | 0.006776 | 0.02104 | 0 |

**Table 4** Statistical Comparison of Properties calculated using candidate equations of state with experimental data for R125

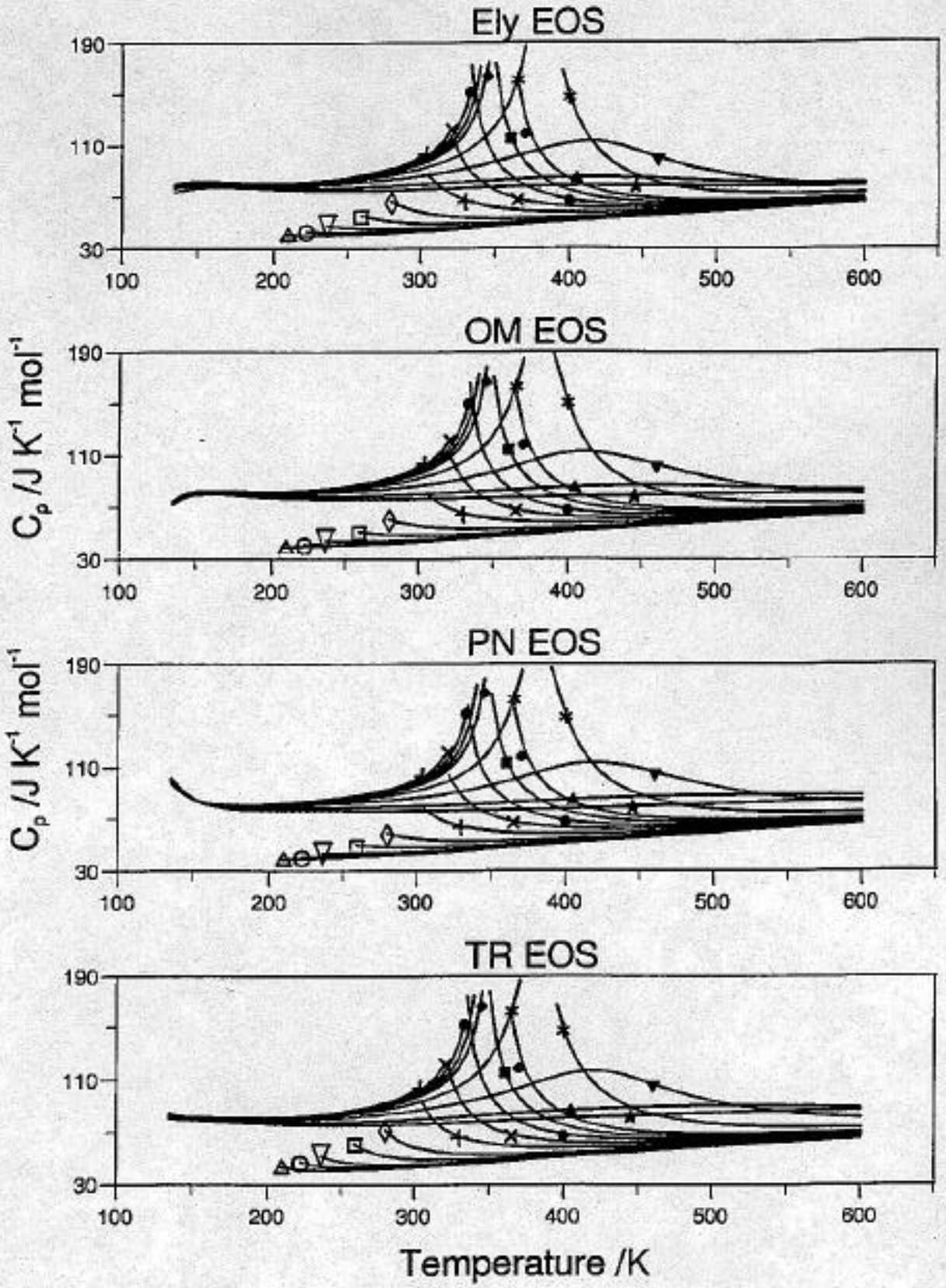
| | EOS | Points | AAD | Bias | Std Dev | RMS | Max Dev | Bad |
|---|---|---|---|---|---|---|---|---|
| Ideal gas | **Ely** | 27 | 0.4276 | 0.1156 | 0.6035 | 0.6034 | 1.432 | 1 |
| heat | **OM** | 27 | 1.665 | -0.2973 | 2.660 | 2.627 | 8.630 | 2 |
| capacity | | | | | | | | |
| Saturated | **Ely** | 293 | 0.1429 | 0.05808 | 0.2843 | 0.2879 | 1.859 | 0 |
| Vapour | **OM** | 293 | 0.1981 | 0.07366 | 0.2971 | 0.3056 | 1.751 | 0 |
| pressure | **PN** | 293 | 0.3022 | 0.2542 | 0.4685 | 0.5323 | 2.762 | 0 |
| Saturated | **Ely** | 25 | 0.4570 | 0.05244 | 1.150 | 1.128 | -4.671 | 2 |
| Liquid | **OM** | 25 | 0.4004 | 0.07580 | 1.001 | 0.9842 | -3.498 | 2 |
| Density | **PN** | 25 | 0.7865 | 0.3282 | 2.044 | 2.030 | 9.204 | 0 |
| Saturated | **Ely** | 8 | 1.647 | 1.647 | 1.194 | 1.990 | 4.222 | 1 |
| Vapour | **OM** | 8 | 2.122 | 2.122 | 2.913 | 3.454 | 9.043 | 0 |
| Density | **PN** | 8 | 2.515 | 2.169 | 2.484 | 3.179 | 6.942 | 1 |
| Saturated | **Ely** | 85 | 0.2023 | -0.02433 | 0.2546 | 0.2543 | 0.6590 | 0 |
| liquid | **OM** | 85 | 0.3454 | -0.1573 | 0.4077 | 0.4348 | 1.147 | 0 |
| heat | **PN** | 85 | 0.4765 | 0.1160 | 0.5633 | 0.5718 | -1.384 | 0 |
| capacity | | | | | | | | |
| *PVT* | **Ely** | 494 | 0.2739 | -0.04851 | 0.7295 | 0.7304 | 9.979 | 8 |
| (liquid | **OM** | 494 | 0.2550 | -0.02538 | 0.7008 | 0.7006 | 9.288 | 8 |
| phase) | **PN** | 494 | 0.2874 | -0.04353 | 0.6564 | 0.6572 | 5.743 | 8 |
| *PVT* | **Ely** | 473 | 0.09235 | 0.03228 | 0.4874 | 0.4879 | 9.779 | 0 |
| (gas | **OM** | 473 | 0.1670 | -0.05098 | 0.5288 | 0.5307 | 9.770 | 0 |
| phase) | **PN** | 473 | 0.1513 | 0.008023 | 0.6353 | 0.6347 | 9.735 | 0 |

| | | | | | | | | |
|---|---|---|---|---|---|---|---|---|
| *B* | **Ely** | 23 | 1.036 | -0.1580 | 1.242 | 1.225 | 2.728 | 1 |
| | **OM** | 23 | 2.991 | -2.425 | 3.442 | 4.149 | -9.893 | 2 |
| | | | | | | | | |
| $C_v$-$\rho$-$T$ | **Ely** | 97 | 0.5836 | 0.4764 | 0.4985 | 0.6877 | 1.859 | 0 |
| (liquid | **OM** | 97 | 0.4250 | 0.1717 | 0.5216 | 0.5466 | 1.639 | 0 |
| phase) | **PN** | 97 | 0.4513 | 0.02251 | 0.5752 | 0.5726 | -1.689 | 0 |
| | | | | | | | | |
| $C_p$-$p$-$T$ | **Ely** | 5 | 1.449 | -0.9926 | 2.292 | 2.278 | -4.916 | 0 |
| (liquid | **OM** | 5 | 1.236 | -0.8003 | 1.870 | 1.854 | -3.939 | 0 |
| phase) | **PN** | 5 | 1.171 | -0.2194 | 1.967 | 1.773 | -3.477 | 0 |
| | | | | | | | | |
| $\omega$-$p$-$T$ | **Ely** | 30 | 0.1042 | -0.01938 | 0.1314 | 0.1306 | 0.3616 | 0 |
| (liquid | **OM** | 30 | 0.3246 | -0.3246 | 0.1607 | 0.3610 | -0.7495 | 0 |
| phase) | **PN** | 30 | 0.2457 | -0.2228 | 0.2089 | 0.3030 | -0.7264 | 0 |
| | | | | | | | | |
| $\omega$-$p$-$T$ | **Ely** | 149 | 0.01420 | 0.001333 | 0.01745 | 0.01744 | 0.04825 | 0 |
| (gas | **OM** | 149 | 0.1146 | 0.1053 | 0.1569 | 0.1886 | 0.6098 | 0 |
| phase) | **PN** | 149 | 0.04361 | 0.003594 | 0.06110 | 0.06100 | 0.2349 | 0 |

## Figure Captions

**Figure 1** Isobaric heat capacity ($C_P$) property plot calculated from all four EOSs for R32. Isobars: (△) 0.05 MPa; (○) 0.1 MPa; (▽) 0.2 MPa; (□) 0.5 MPa; (◇) 1.0 MPa; (+) 2.0 MPa; (×) 3.0 MPa; (•) 4.0 MPa; (■) 5.0 MPa; (◆) 6.0 MPa; (*) 10.0 MPa; (▼) 20.0 MPa; (▲) 50.0 MPa; (⋆) 100.0 MPa.

**Figure 2** Isobaric heat capacity ($C_P$) property plot calculated from the all three EOSs for R125. Isobars: (△) 0.05 MPa; (○) 0.1 MPa; (▽) 0.2 MPa; (□) 0.5 MPa; (◇) 1.0 MPa; (+) 2.0 MPa; (×) 3.0 MPa; (•) 4.0 MPa; (■) 5.0 MPa; (◆) 6.0 MPa; (*) 10.0 MPa; (▼) 20.0 MPa; (▲) 50.0 MPa; (⋆) 100.0 MPa.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PN EOS
TR EOS
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110
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100
200
300
400
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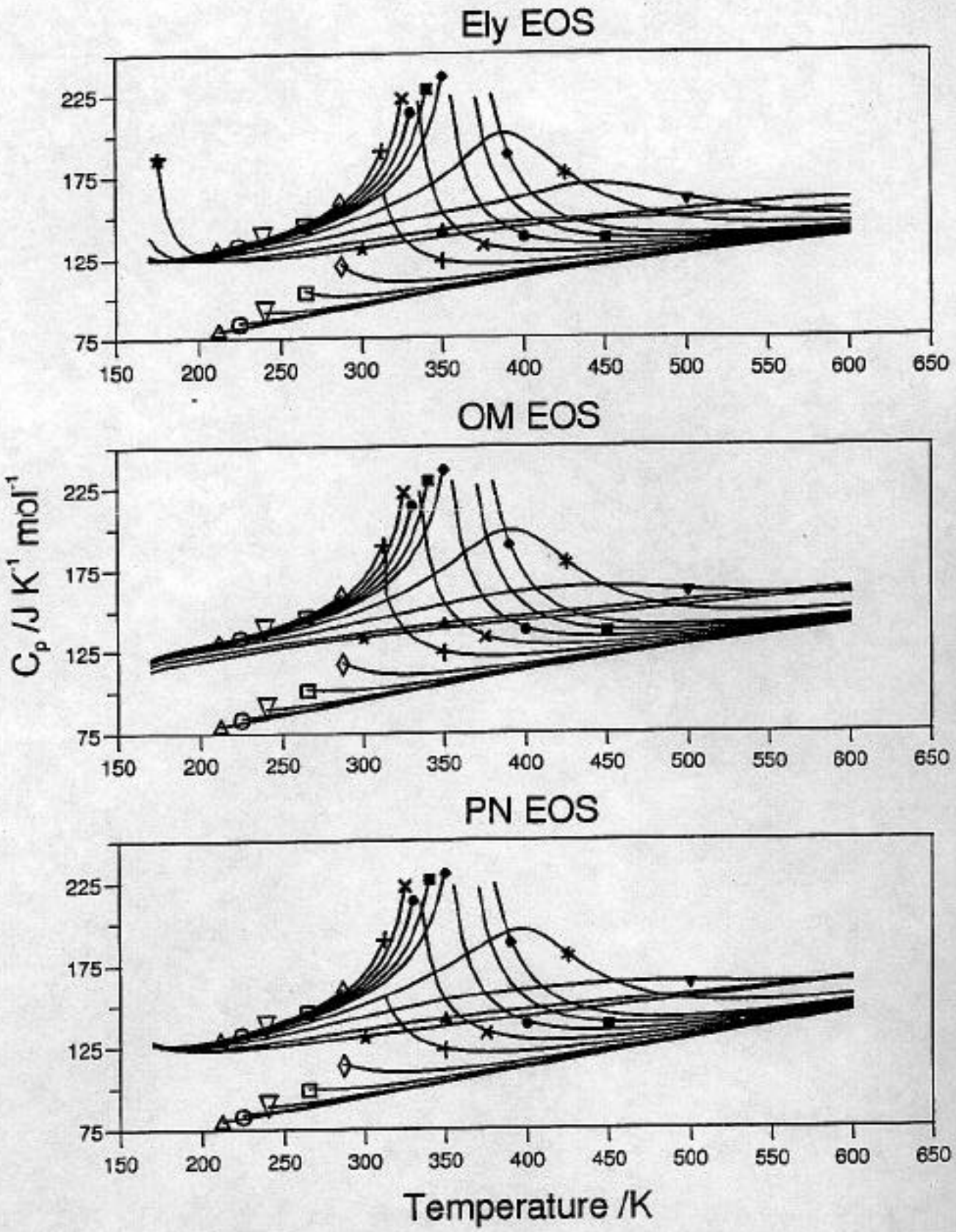
600
Temperature /K


Ely EOS
OM EOS
PN EOS
$C_p$ /J K$^{-1}$ mol$^{-1}$
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400
450
500
550
600
650