

# Mixing rules for the specific heat capacities of several HFC-mixtures

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The specific heat capacity at constant pressure ( $c_p$ ) of some relevant HFCs as replacements for R12, R502 and R22 was measured. The liquids investigated are binary or ternary mixtures of R134a, R152a, R125, R32 and R143a. Empirical functional relations in polynomial forms between the temperature, specific heat capacity and concentration are established and the coefficients of the polynomial correlations are presented. These equations can be used to calculate the  $c_p$ -values for the mixtures investigated over the whole concentration range and the predicted properties generally agree with the source data to  $\pm 0.1\%$  for the pure substances. The accuracy of the measurements is better than  $<1\%$  for the pure fluids and  $<1.5\%$  for the mixtures. Differences between 1 and 2% can occur only at temperatures  $>40^\circ\text{C}$  and  $<-50^\circ\text{C}$ .  
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(Keywords: refrigerant; mixture; specific heat; calculation; R134a; R152a; R125; R32; R143a; measurement; comparison)

## Règles de mélange pour les chaleurs spécifiques de plusieurs mélanges de HFC

*On a mesuré la chaleur spécifique à pression constante de quelques HFC représentatifs comme produits de remplacement des R12, R502 et R22. Les liquides étudiés sont des mélanges binaires ou tertiaires de R134a, R152a, R125, R32 et R143a. On établit les relations empiriques fonctionnelles sous forme de polynôme entre température, chaleur spécifique et concentration. On peut utiliser ces équations pour calculer les valeurs  $c_p$  pour les mélanges étudiés sur la gamme entière de concentrations. Les propriétés prévues sont généralement en accord avec les données mesurées, avec une marge d'environ plus ou moins 0,1% pour les corps purs. La précision des mesures est supérieure à 1% pour les corps purs et à 1,5% pour les mélanges. Des différences de 1 à 2% maximum n'apparaissent qu'à des températures supérieures à 40 deg C et inférieures à -50 deg C.*

(Mot clés: frigorigène; mélange; chaleur massique; calcul; R134a; R152a; R125; R32; R143a; mesure; comparaison)

### Introduction

Research in the fields of air-conditioning and refrigeration applications has focused on the replacement of the refrigerants R12, R22 and R502; R22 is widely used in all areas of air-conditioning and refrigeration and R502 is mainly employed in supermarket and transport refrigeration systems.

In order to reduce environmental pollution the phasing out of H-CFC22 will be completed in Germany by 1999, and even earlier deadlines are under discussion. The use of R502, which contains CFC115, has been completely prohibited for new applications since 1 January 1995. As per the decision of the German government, R12 has to be retrofitted within a period of 30 months, which started December 1995.

In view of the above-mentioned ecological problems, the alternative refrigerants should be non-flammable,

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### Nomenclature

|                 |                                             |
|-----------------|---------------------------------------------|
| $a, b, c, d, e$ | Coefficients                                |
| $c_B$           | Excess heat capacity                        |
| $c_p$           | Specific heat capacity at constant pressure |
| $p$             | Pressure                                    |
| $r$             | Deviation                                   |
| $T$             | Temperature                                 |
| $x$             | Concentration                               |
| $X$             | Mole fraction                               |

### Subscripts

|           |                              |
|-----------|------------------------------|
| $R$       | Reduced                      |
| $me\beta$ | Determined from measurements |
| $ber.$    | Calculated                   |
| $n$       | Variable                     |

|        |          |
|--------|----------|
| $c$    | Critical |
| $ges.$ | Total    |
| $B$    | Binary   |

### Definition of the deviations

$$\text{Deviation : } r = \frac{c_{p,me\beta} - c_{p,ber}}{c_{p,me\beta}} \times 100\%$$

$$\text{Mean deviation : } r_m = \frac{1}{n} \sum_1^n |r|$$

$$\text{Standard deviation : } r_s = \sqrt{\frac{1}{n} \sum_1^n \left( r - \frac{1}{n} \sum_1^n r \right)^2}$$

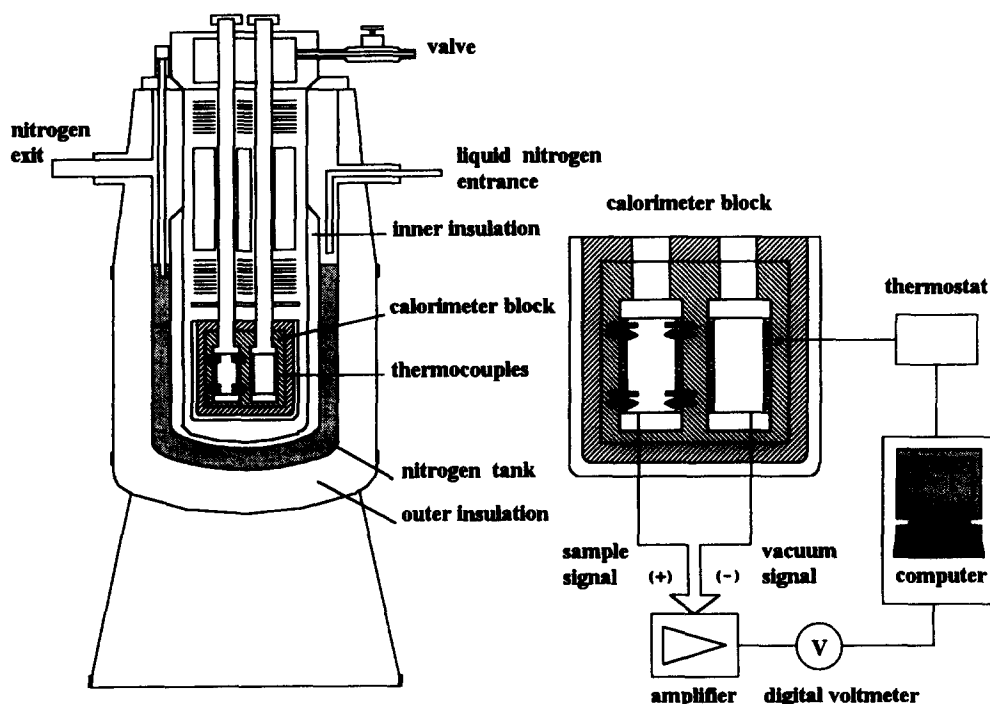


Figure 1 Layout of heat capacity measurement equipment

Figure 1 Disposition du matériel de mesure de la chaleur massique

non-toxic, have a zero ozone depletion potential (ODP), a zero global warming potential (GWP) and, also should be efficient. The problem is to find a replacement candidate which meets all these necessary qualities.

If non-flammable and non-toxic refrigerants with a thermodynamic behaviour equal to R22 or R502 are required GWP is inevitable. Long-term safety replacements with a low GWP are necessary, therefore, to keep ecological damage to a minimum.

Investigations have been carried out on the following alternative blends with a zero ODP and a relatively low GWP:

- blends of R134a/R152a with concentrations of 20, 40, 60 and 80 wt% R134a;
- blends of R143a/R125 with concentrations of 25, 50 (R507) and 75 wt% R143a;
- blends of R134a/R125 with concentrations of 25, 50 and 75 wt% R134a;

- blends of R134a/R32 with concentrations 20, 40, 60 and 80 wt% R134a;
- blends of R32/R125 with the concentrations 25, 50 (R410A) and 75 wt% R32;
- commercially available ternary blends such as R407A, R407C and R404A.

### Experimental implementation for heat capacity measurement

The heat capacity of the liquid at a constant pressure was determined using a Calvet-microcalorimeter which works according to the scanning method<sup>1</sup>. The calorimeter can be used over a temperature range of  $-196$  to  $200^{\circ}\text{C}$  with a maximum resolution of  $1\ \mu\text{V}$ . The layout of the heat capacity measurement equipment is shown in *Figure 1*.

By means of a control system, the calorimeter block is heated with a constant rate of temperature increase. Due to the temperature difference between the calorimeter block and the sample, heat flows through the thermopile into the cell. The resulting voltage in the thermocouples is registered by the evaluating system and is a direct measure of the transferred heat flow.

The cells in the calorimeter block are filled with the refrigerant using a membrane container, which remains connected to the measuring device during

the whole measurement. In this way an evaporation of the refrigerant can be prevented. The slight pressure increase of a maximum of  $0.05\ \text{bar}$  could be neglected in the temperature range investigated.

In our case, the temperature varied between  $-70$  and  $50^{\circ}\text{C}$  at a constant pressure of  $30\ \text{bar}$ . Because of the vapour pressure the  $c_p$ -values could be determined only up to  $40^{\circ}\text{C}$  for R32 and up to  $45^{\circ}\text{C}$  for R125 and R143a. A more detailed description of the measuring device and the evaluation of the measurements is given in Ref. 2.

Using the device described, it was possible to obtain  $c_p$ -values not only at certain temperatures but over the whole temperature range. The uncertainties are within  $\pm 1\%$  for the pure fluids over the temperature ranges of  $-50^{\circ}\text{C} < T < 40^{\circ}\text{C}$  and within  $\pm 2\%$  for temperatures between  $-70^{\circ}\text{C} < T < -50^{\circ}\text{C}$  and  $40^{\circ}\text{C} < T < 50^{\circ}\text{C}$ . The uncertainties for the mixtures increase slightly by *ca*  $0.5\%^2$ .

### Determination of the heat capacity at constant pressure and the mixing effects

#### Pure liquids

The specific heat capacity of the ideal gases together with reliable  $p$ ,  $v$ ,  $T$ ,  $x$ -data is sufficient to completely describe the thermodynamic behaviour of fluids. But even if the precise properties of the heat capacity of

**Table 1** Coefficients to Equation (1) ( $c_p$  at 30 bar), range of correlation:  $203.15\ \text{K} < T < 323.15\ \text{K}$

Tableau 1 Coefficients de l'équation (1) ( $c_p$  à 30 bar), gamme de corrélation :  $203,15\ \text{K} < T < 323,15\ \text{K}$

| R152a–R134a $x$ in (wt%) |           |             |             |             |             |           |
|--------------------------|-----------|-------------|-------------|-------------|-------------|-----------|
|                          | R152a     | 80.18–19.82 | 60.17–39.83 | 40.09–59.91 | 25.42–74.58 | R134a     |
| $a$                      | 2.323139  | 2.378378    | 2.396295    | 2.025721    | 1.878176    | 1.523057  |
| $b$                      | 0.036902  | 0.044411    | 0.044898    | 0.058063    | 0.077326    | 0.046390  |
| $c$                      | -4.091243 | -4.674676   | -4.796301   | -3.914176   | -3.947407   | -2.136808 |
| $d$                      | 5.846006  | 6.672435    | 6.618012    | 6.003144    | 6.547084    | 3.341614  |
| $e$                      | -2.260359 | -2.711136   | -2.663541   | -2.830098   | -3.520957   | -1.525919 |
| $*T_c\ (\text{K})$       | 386.41    | 384.73      | 382.73      | 380.37      | 377.55      | 374.18    |

\* Pure substances from Ref. 3 mixtures calculated from mole quantity balance

**Table 2** Coefficients to Equations (1) ( $c_p$  at 30 bar) and (5) ( $c_B$  – excess heat capacity), range of correlation:  $203.15\ \text{K} < T < 313.15\ \text{K}$

Tableau 2 Coefficients de l'équation (1) ( $c_p$  à 30 bar) et 5 ( $c_B$  – excès de chaleur massique), gamme de corrélations :  $203,15\ \text{K} < T < 313,15\ \text{K}$

| R134a–R32 $x$ in (wt%) |           |             |             |             |             |             |
|------------------------|-----------|-------------|-------------|-------------|-------------|-------------|
|                        | R32       | 20.82–79.18 | 39.83–60.17 | 59.45–40.55 | 74.84–25.16 | R134a $c_B$ |
| $a$                    | 2.280202  | 2.652152    | 4.775369    | 5.202856    | 2.206065    | 1.523057    |
| $b$                    | 0.094172  | 0.087267    | 0.113717    | 0.108494    | 0.090787    | 0.046390    |
| $c$                    | -3.700349 | -5.903840   | -15.776099  | -18.188789  | -5.237787   | -2.136808   |
| $d$                    | 4.886391  | 8.427172    | 23.123988   | 26.993743   | 8.279623    | 3.341614    |
| $e$                    | -2.306336 | -4.088435   | -11.594404  | -13.548120  | -4.499773   | -1.525919   |
| $*T_c\ (\text{K})$     | 351.23    | 354.36      | 357.75      | 361.98      | 365.95      | 374.18      |

\* According to Ref. 11

**Table 3** Coefficients to Equation (1) ( $c_p$  at 30 bar) and (5) ( $c_B$  – excess heat capacity), range of correlation: 203.15 K <  $T$  < 318.15 K

Tableau 3 Coefficients de l'équation (1) ( $c_p$  à 30 bar) et 5 ( $c_B$  – excès de chaleur massique), gamme de corrélations: 203,15 K <  $T$  < 318,15 K

|             | R125      | R134a–R125 x in (wt%) |             |             | R134a     | $c_B$    |
|-------------|-----------|-----------------------|-------------|-------------|-----------|----------|
|             |           | 24.95–75.05           | 50.24–49.76 | 74.86–25.14 |           |          |
| <i>a</i>    | 2.629323  | 3.432058              | 3.256015    | 3.246083    | 1.523057  | 4.5710   |
| <i>b</i>    | 0.059411  | 0.079559              | 0.096777    | 0.086733    | 0.046390  | –0.0645  |
| <i>c</i>    | –8.235309 | –12.069612            | –11.337748  | –10.812275  | –2.136808 | –19.3656 |
| <i>d</i>    | 13.022110 | 19.110761             | 18.416096   | 17.172272   | 3.341614  | 27.4950  |
| <i>e</i>    | –6.735505 | –10.028976            | –10.032563  | –9.092536   | –1.525919 | –12.4308 |
| * $T_c$ (K) | 339.43    | 349.09                | 358.16      | 366.30      | 374.18    |          |

\* According to Ref. 4

**Table 4** Coefficients to Equation (1) ( $c_p$  at 30 bar), range of correlation: 203.15 K <  $T$  < 313.15 K

Tableau 4 Coefficients de l'équation (1) ( $c_p$  à 30 bar) et 5 ( $c_B$  – excès de chaleur massique), gamme de corrélations: 203,15 K <  $T$  < 313,15 K

|             | R125      | R143a–R125 x in (wt%) |             |             | R143a     |
|-------------|-----------|-----------------------|-------------|-------------|-----------|
|             |           | 25.52–74.48           | 50.18–49.82 | 74.58–25.42 |           |
| <i>a</i>    | 2.629323  | 3.015531              | 3.761587    | 3.898078    | 1.886436  |
| <i>b</i>    | 0.059411  | 0.070922              | 0.077957    | 0.078873    | 0.076849  |
| <i>c</i>    | –8.235309 | –9.916324             | –12.920331  | –13.227531  | –4.658835 |
| <i>d</i>    | 13.022110 | 15.709629             | 19.957944   | 20.207903   | 8.468966  |
| <i>e</i>    | –6.735505 | –8.187892             | –10.212785  | –10.209283  | –4.844599 |
| * $T_c$ (K) | 339.43    | 341.95                | 343.64      | 344.85      | 345.88    |

\* According to Ref. 4

**Table 5** Coefficients to Equations (1) ( $c_p$  at 30 bar) and (5) ( $c_B$  – excess heat capacity)

Tableau 5 Coefficients de l'équations (1) ( $c_p$  à 30 bar) et 5 ( $c_B$  – excès de chaleur massique)

|             | R125      | R32–R125 x in (wt%) |             |             | R32       | $c_B$     |
|-------------|-----------|---------------------|-------------|-------------|-----------|-----------|
|             |           | 24.95–75.05         | 49.96–50.04 | 75.12–24.88 |           |           |
| <i>a</i>    | 2.629323  | 7.818461            | 7.478618    | 6.903290    | 2.280202  | 25.5642   |
| <i>b</i>    | 0.059411  | 0.124595            | 0.141150    | 0.129854    | 0.094172  | 0.2978    |
| <i>c</i>    | –8.235309 | –30.698336          | –28.851252  | –24.928073  | –3.700349 | –114.8689 |
| <i>d</i>    | 13.022110 | 45.824560           | 43.283526   | 36.336742   | 4.886391  | 170.0580  |
| <i>e</i>    | –6.735505 | –23.126908          | –22.089202  | –18.052248  | –2.306336 | –85.7313  |
| * $T_c$ (K) | 339.43    | 341.40              | 344.56      | 347.96      | 351.23    |           |

\* According to Ref. 12

ideal gases are available, there are nevertheless great difficulties in exactly calculating the enthalpy of liquids. Therefore, based on the measurements, simple functional relations between the specific heat capacity at constant pressure ( $c_p$ ) and the temperature have been established in order to enable the calculation of the enthalpy of the liquid phase.

The coefficients of the following equation were adapted to the specific heat capacity data  $c_p$  (kJ kg<sup>–1</sup> K<sup>–1</sup>)

$$c_p(T) = a + b(1 - T_R)^{-1} + cT_R + dT_R^2 + eT_R^3 \quad (1)$$

with

$$T_R = T/T_c$$

and

$$T \text{ in (K)}$$

Over the range of correlation as given in Tables 1 to 5 the maximum deviation from the source data is 0.3%. The mean deviation is <0.1%.

Tables 1 to 5 show the coefficients of Equation (1) and the range of the correlation for each of the fluids.

The results for R134a, R152a and R32 are in good agreement with the literature data, the mean

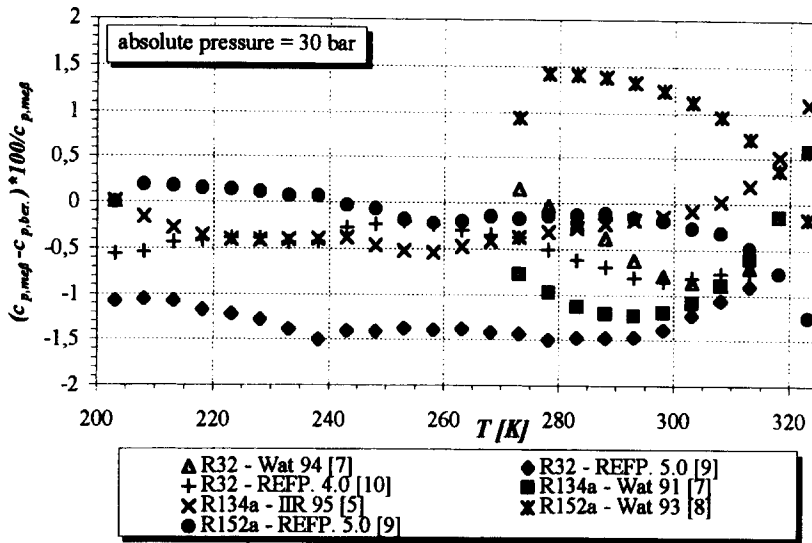


Figure 2 Deviations of literature data from the measured  $c_p$ -values at 30 bar

Figure 2 *Déviations des valeur mesurées  $c_p$  à 30 bar par rapport aux données de la bibliographic*

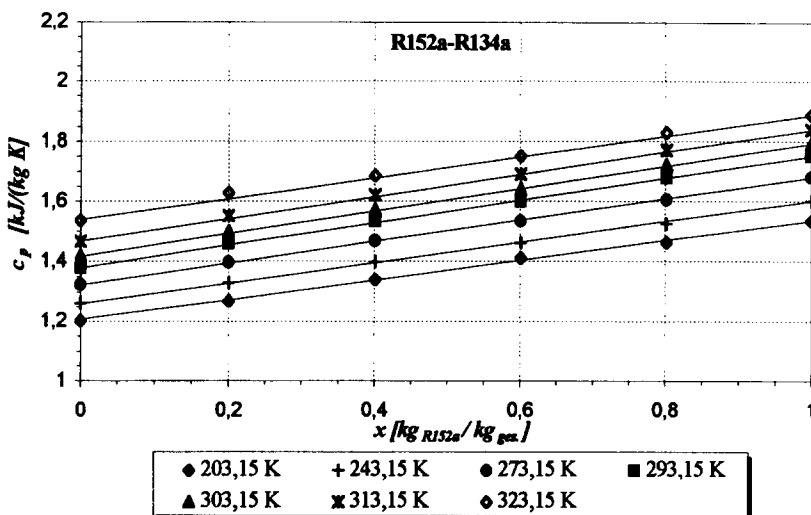


Figure 3 Comparison of the measured specific heat capacity at 30 bar (symbols) with the calculated  $c_p$ -data [lines – Equation (2)] as a function of the concentration

Figure 3 *Comparaison de la chaleur massique spécifique mesurée à 30 bar (symboles) avec les données calculées  $c_p$  [lignes – Equation (2)] en fonction de la concentration*

deviations are  $<1\%^{5-10}$  cf Figure 2. A more detailed discussion of the deviations from the literature data is given in Ref. 2.

#### Mixing rules for the calculation of specific heat capacities of mixtures from the data on pure substances

In the simplest case, the specific heat capacity of the mixture can be directly calculated from the quantities of pure substances:

$$c_p = x_1 c_{p1} + x_2 c_{p2} + \dots + x_n c_{pn} \quad (2)$$

where  $(\text{kg} \cdot \text{kg}_{\text{ges}}^{-1})$  is the concentration of the

components 1–n and  $c_p (\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1})$  is the specific heat capacity at constant pressure.

The calculation from Equation (2) can be applied to mixtures in which the molecules of the components do not interact. However, if any positive or negative deviations from the ideal behaviour occur, the  $c_p$  for binary mixtures can be calculated by a quantity balance combination of the pure substance equations and an additional function  $c_B$  as follows:

$$c_p(T, x) = x c_{p1}(T) + (1 - x) c_{p2}(T) + x(1 - x) c_{B12}(T) \quad (3)$$

Corresponding to Equation (3), the  $c_p$  of the ternary

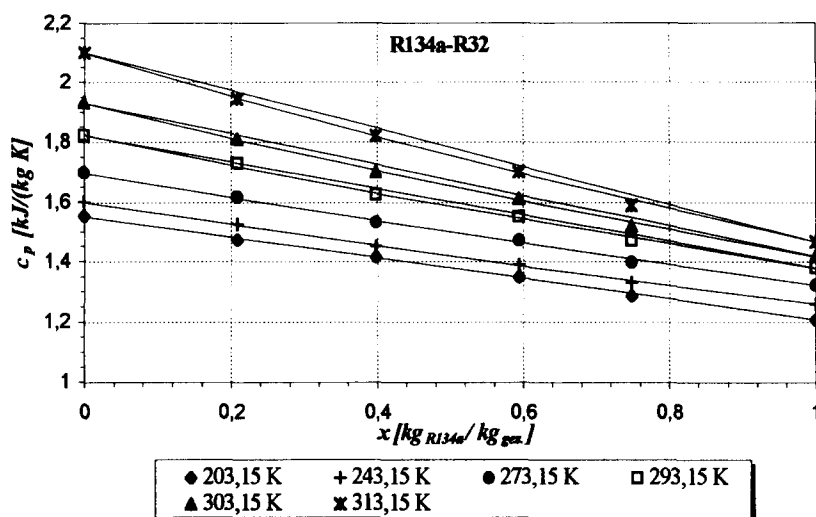


**Figure 4** Comparison of the measured specific heat capacity at 30 bar (symbols) with the calculated  $c_p$ -data [lines – Equations (2) and (3)] as a function of the concentration

*Figure 4 Comparaison de la chaleur massique spécifique mesurée à 30 bar (symboles) avec les données calculées  $c_p$  [lignes – Equations (2) et (3)] en fonction de la concentration*

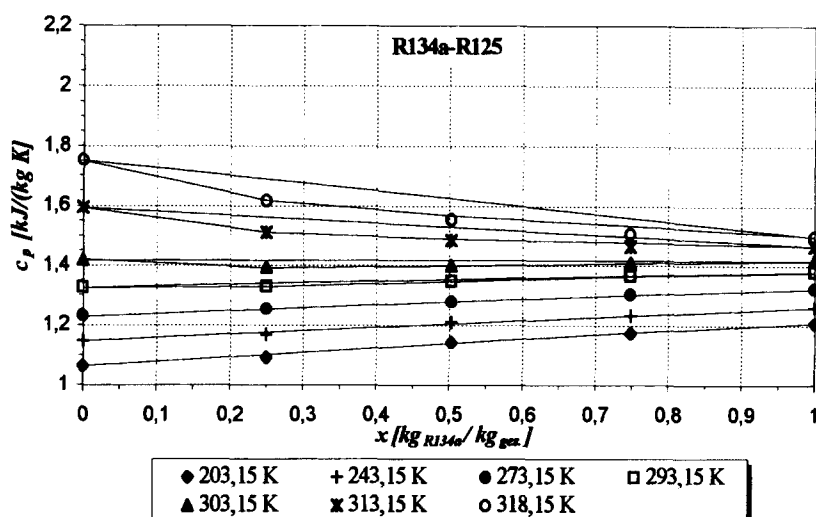


**Figure 5** Comparison of the measured specific heat capacity at 30 bar (symbols) with the calculated  $c_p$ -data [lines – Equations (2) and (3)] as a function of the concentration

*Figure 5 Comparaison de la chaleur massique spécifique mesurée à 30 bar (symboles) avec les données calculées  $c_p$  [lignes – Equations (2) et (3)] en fonction de la concentration*

mixtures can be calculated as follows:

$$c_p(T, x) = x_1 c_{p1}(T) + x_2 c_{p2}(T) + x_3 c_{p3}(T) \\ + x_1 x_2 c_{B12}(T) + x_1 x_3 c_{B13}(T) \\ + x_2 x_3 c_{B23}(T) \quad (4)$$

The additional function  $c_B(T)$  describes the excess heat capacity of binary mixtures. Equations (3) and (4) are determined from the respective binary subsystems by adapting the coefficients ( $a$ – $e$ ) of Equation (5) to the measurements.

$$c_B(T) = a + b(1 - T_R)^{-1} + cT_R + dT_R^2 + eT_R^3 \quad (5)$$

The coefficients  $a$ – $e$  have the unit are measured in  $\text{kJ} \cdot \text{kg}^{-1} \cdot \text{K}^{-1}$ . The  $T_c$ s of the binary mixtures investigated were calculated according to Refs 4, 11 and 12 (cf Table 1–5). The  $T_c$  of the binary subsystems forming the ternary mixtures can be calculated from a simple mole quantity balance of the  $T_c$ s of the pure substances. For this, the sum of the mole quantities of each subsystem has to be assumed to be 100% and the new ‘pseudo-concentration’  $X^*$  of component 1 in the binary subsystem is:

$$X_1^* = \frac{X_1}{X_1 + X_2}$$

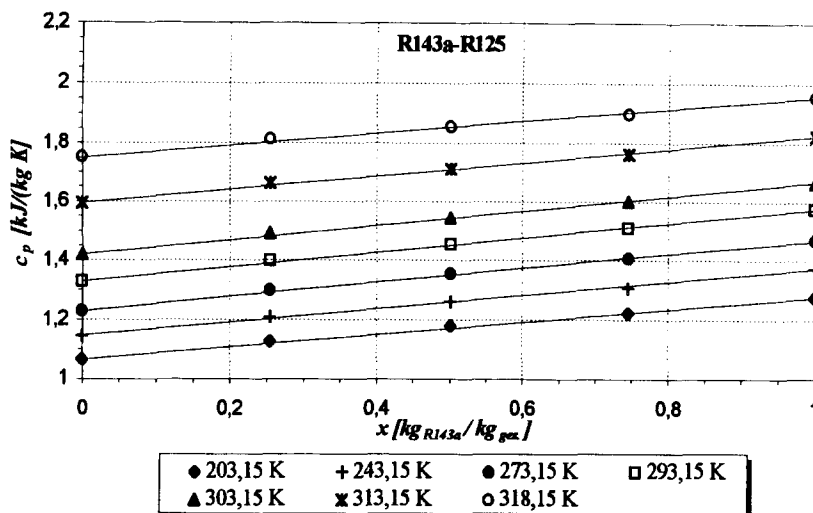


Figure 6 Comparison of the measured specific heat capacity at 30 bar (symbols) with the calculated  $c_p$ -data [lines – Equation (2)] as a function of the concentration

Figure 6 Comparaison de la chaleur massique spécifique mesurée à 30 bar (symboles) avec les données calculées  $c_p$  [lignes – Equation (2)] en fonction de la concentration

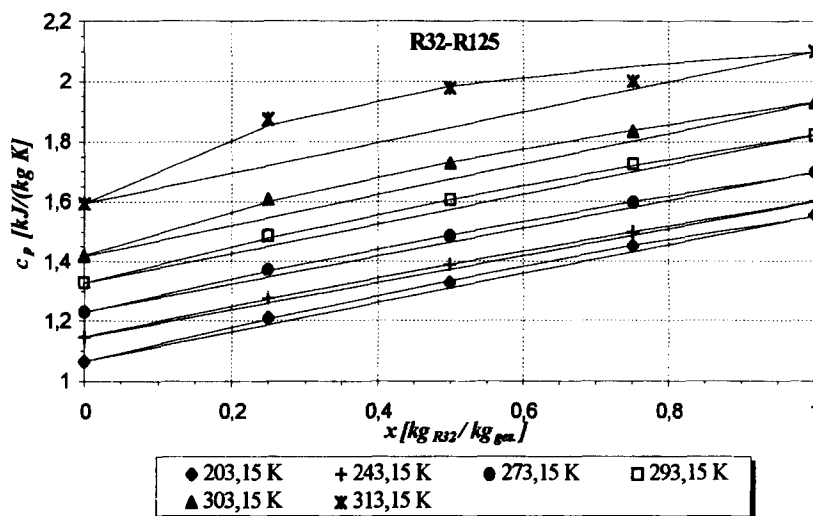


Figure 7 Comparison of the measured specific heat capacity at 30 bar (symbols) with the calculated  $c_p$ -data [lines – Equations (2) and (3)] as a function of the concentration

Figure 7 Comparaison de la chaleur massique spécifique mesurée à 30 bar (symboles) avec les données calculées  $c_p$  [lignes – Equations (2) et (3)] en fonction de la concentration

Tables 2, 3 and 5 show the coefficients of Equation (5) for the mixtures with significant deviations from the ideal behaviour. The calculated as compared to the actual  $c_{p,me\beta}$ -values for all the mixtures investigated are plotted in Figures 3–8.

Table 6 summarizes the results of the calculations on the  $c_p$ -data of mixtures from the pure substance data. The mean deviations from the measurements are  $<0.7\%$  for all calculations. The asymmetry of the mixture effect of R134a–R125 (cf Figure 5) and R32–R125 (cf Figure 7) could be reproduced very well using only the critical temperatures according to Refs.

4 and 12. Some mixtures (cf Figure 3 R134a, Figure 6 R143a–R125) are very close to an ideal behaviour, so that these values were calculated with the quantity balance according to Equation (2), the deviations also showed no uniform tendency with respect to the concentration or the temperature.

The mixture effects of the ternary mixtures can only be slightly better reproduced by Equation (4) than by the ideal behaviour calculation (cf Figure 8). It can be assumed that the sum of the occurring positive (cf Figure 6 R32–R125) and negative (cf Figure 4 R134a–R32, Figure 5 R134a–R125) deviations from the ideal mixture

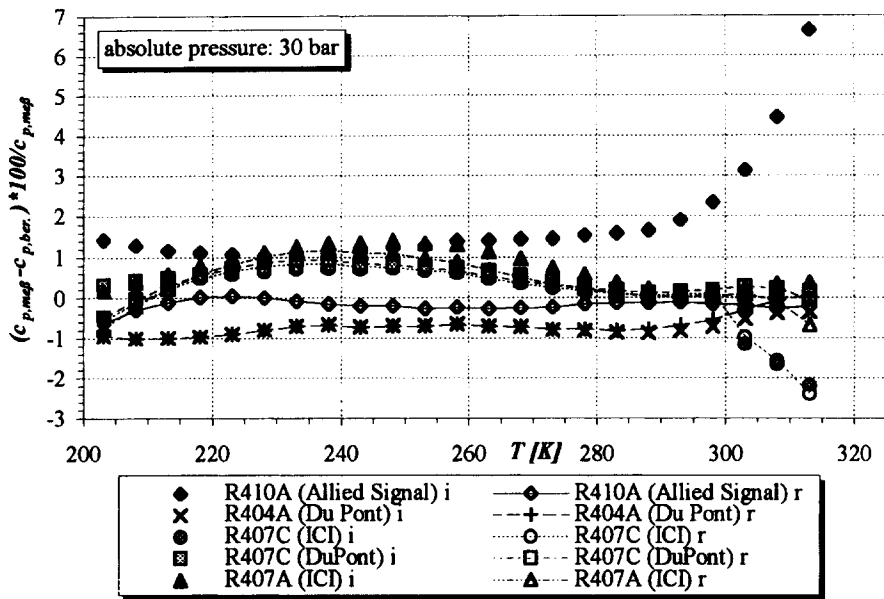


**Figure 8** Deviations of the calculated [Equation (4) for  $r$  and Equation (2) for  $i$ ] from the measured  $c_{p,me\beta}$ -data – commercially available refrigerants  
**Figure 8** Déviations des données  $c_{p,me\beta}$  calculées [Equation (4) pour  $r$  et Equation (2) pour  $i$ ] par rapport aux données mesurées – frigorigènes commercialisés

**Table 6** Deviations as  $(c_{p,me\beta} - c_p) \times 100 / c_{p,me\beta}$

**Tableau 6** Déviation pour  $(c_{p,me\beta} - c_p) \times 100 / c_{p,me\beta}$

| Deviations (%)  | Standard | Mean | Max. | $c_p$ calc. with Equation | Max. temp (K) |
|-----------------|----------|------|------|---------------------------|---------------|
| R134a/R152a     | 0.2      | 0.3  | 1.2  | 2                         | 323.15        |
| R134a/R32       | 0.2      | 0.4  | 1.2  | 4                         | 313.15        |
| R134a/R125      | 0.4      | 0.3  | 1.8  | 3                         | 318.15        |
| R143a/R125      | 0.2      | 0.3  | 0.9  | 2                         | 318.15        |
| R32/R125        | 0.3      | 0.3  | 2.6  | 3                         | 313.15        |
| R410A           | 0.1      | 0.2  | 0.6  | 4                         | 313.15        |
| R407C (Du Pont) | 0.4      | 0.7  | 0.4  | 4                         | 313.15        |
| R407C (ICI)     | 0.8      | 0.7  | 2.6  | 4                         | 313.15        |
| R407A           | 0.6      | 0.6  | 1.2  | 4                         | 313.15        |
| R404A           | 0.3      | 0.7  | 1    | 4                         | 313.15        |
| R507            | 0.2      | 0.2  | 0.7  | 4                         | 318.15        |

behaviour of the binary subsystems are relatively small for the ternary mixture concentrations investigated.

### Discussion and conclusions

The aim of part of this project was to determine the specific heat capacity of HFC-mixtures over a temperature range of  $-70^\circ\text{C} < T < 50^\circ\text{C}$ . The specific heat capacity of the pure refrigerants R134a, R152a, R125, R32 and R143a (min. purity 99.8%), as well as of several binary mixtures of these refrigerants, was determined. In addition, the commercially available ternary binary refrigerant mixtures R407A, R407C, R404A and R410A were examined. The mixture compositions of the refrigerants available from the companies ICI, Du Pont, Solvay and Allied Signal were not tested in these investigations.

The uncertainty of the measurements for the specific heat capacity, over a temperature range of  $-50$  to  $40^\circ\text{C}$ , is  $<1\%$  for the pure fluids and  $<1.5\%$  for the mixtures. Over the temperature ranges of  $-70^\circ\text{C}$  to  $-50^\circ\text{C}$  and  $40$ – $50^\circ\text{C}$  the measuring uncertainty is  $<2\%$ .

The specific heat capacities at constant pressure obtained from the measurements can be determined by an empirical correlation (1) with five substance-specific coefficients (cf *Tables 1–5*). The mean relative deviations are  $<0.1\%$  and clearly below the measuring uncertainty.

Furthermore, correlation equations which reproduce the mixture effects of the binary systems could be established. By means of these correlations, it is possible to calculate the specific heat capacity of binary and ternary mixtures from the data on pure fluids, as far as they have been investigated

in the present work. The following results were obtained:

- the data on the mixtures R134a–R152a and R143a–R125 can be calculated with a simple quantity balance from Equation (2);
- the mixture effects which arise from the mixtures R134a–R125, R32–R125 and R134a–R32 are successfully approximated by Equation (3);
- the mixture effects of the ternary mixtures (R407A R407C and R404A) are slightly better represented by Equation (4) than by using a quantity balance [Equation (2)].

More precise calculations on the basis of pure substance data are not possible due to the measuring uncertainty.

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