Complexes between Gaseous Gallium Chloride and Cobalt Chloride

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In gaseous gallium chloride, Co\(^{3+}\) is found as CoGa\(_4\)Cl\(_8\) with the following thermodynamic functions of formation at 073 K:

\[ \Delta H^\circ = 43.9 \pm 0.7 \text{ kJ}, \quad \Delta S^\circ = 36.9 \pm 1.1 \text{ J K}^{-1} \]

The structure of CoGa\(_4\)Cl\(_8\) is discussed, and a coordination of CoCl\(_4\) to the GaCl\(_4\) molecule, without breaking the double bond in GaCl\(_4\), is suggested. An octahedral coordination of Co\(^{3+}\) is considered unlikely.

As an extension of a general study of gas complexes between AlCl\(_3\) and divalent 3d-metal chlorides \(^1\)\(^-\)\(^5\) it was decided to investigate the complexing properties of GaCl\(_3\) with respect to CoCl\(_4\).

Gaseous GaCl\(_3\) and AlCl\(_3\) have very similar structural and thermodynamic properties, both being subject to the equilibrium

\[ \text{A}_2\text{Cl}_4(g) = 2\text{ACl}_3(g), \quad \text{A} = \text{Al}, \text{ Ga} \]  \hspace{1cm} (1)

which is shifted to the right with increasing temperature. However, in spite of their similarities it is of interest to determine how the heavier Ga\(^{3+}\) ion compares to Al\(^{3+}\) for the general complexing reaction:

\[ \text{MCl}_4(s, g) + n_2\text{A}_2\text{Cl}_4(g) = \text{MA}_{2n}\text{Cl}_{4n+1}(g) \] \hspace{1cm} (2)

with CoCl\(_4\) as the metal chloride in the present study. Although GaCl\(_3\) is more difficult to handle than AlCl\(_3\), the experiments were expected to benefit from the shift to the left of the side reaction:

\[ 2\text{A}_2\text{Cl}_4(g) + 3\text{SiO}_2(s) = 2\text{A}_2\text{O}_3(s) + 3\text{SiCl}_4(g) \] \hspace{1cm} (3)

when substituting Ga\(_4\)Cl\(_8\) for Al\(_4\)Cl\(_8\).

The chosen experimental method was absorption spectroscopic determination of the pressure of the gas complex in closed quartz cells according to eqn. (2). This method has been described in detail previously.\(^1\)\(^4\)

The molar absorptivity of a known amount of MCl\(_3\) in the gas phase is first determined by the equation:

\[ \varepsilon_{\text{Co-Ga-Cl}} = A \cdot V / n \ell \] \hspace{1cm} (4)

whereafter the pressure of the gas complex in the presence of MCl\(_3\)(s) is determined by

\[ P_{\text{Co-Ga-Cl}} = \frac{A \cdot R \cdot T}{\varepsilon_{\text{Co-Ga-Cl}}} \] \hspace{1cm} (5)

\[ \varepsilon_{\text{Co-Ga-Cl}}: \text{molar absorptivity of the gas complex at wavelength } \ell, \text{ } A: \text{optical density at wavelength } \ell, \text{ } V: \text{volume, } n: \text{mol of gas complex, } \ell: \text{optical path length, } P: \text{pressure of gas complex, } R: \text{gas constant and } T: \text{mean cell temperature.} \]

The pressure of Ga\(_4\)Cl\(_8\) as well as the temperature were varied so that the stoichiometry and the thermodynamic functions for the reaction corresponding to eqn. (2) could be obtained.

**EXPERIMENTAL**

Anhydrous GaCl\(_3\) was obtained in ampoules as 99.99 % (Schuchardt, München, DBR) and no further purification was performed. Anhydrous CoCl\(_4\) was prepared from CoCl\(_4\)-6H\(_2\)O "pure" (Koch-Light Labs., Colubrook, England) by heating under vacuum at 120 °C for 4 h,
followed by purging with HCl for 2 h at 450 °C, and then repeated sublimation.

The experimental procedure has been described previously.1 Equilibrium was found to be established within 30 min, at which time the spectrum was recorded.

RESULTS

Spectrophotometrically the gas complex formation between CoCl₄(s) and GaCl₃ was detected above 280 °C by the appearance of two composite absorption bands at about 15.6 and 7.1 kK (Fig. 1). By analyzing the spectrum in detail, absorption maxima and shoulders were observed at the following wave numbers [expressed in kK with the molar absorptivities at 400 °C given in parenthesis (Exp. 2, Table 1): 14.9(85), 15.6(150), 16.8(140), 17.6(sh), 18.5(sh), 7.1(30), 5.9(—)].

Due to the composite nature of the two main absorption regions, individual spectral half-widths could not be given, but would probably not exceed 2 kK.

The molar absorptivity for the strongest peak was determined in three experiments specified in Table 1. The molar absorptivity decreased with increasing temperature and did not reveal any dependence on the pressure of Ga₃Cl₆ above 400 °C. Between 300 and 400 °C a small decrease in molar absorptivity with increasing Ga₃Cl₆ pressure may be present, similar to what is observed by Dell'Anna and Emmenegger* for the CoCl₃–Al₆Cl₁₆ gas complex. Statistically, however, the pressure dependence was not significant on a 90% confidence level, and all the molar absorptivity data were fitted to the following pressure-independent equation with a 2.5% standard deviation:

$$
\varepsilon^{14}_{\text{Co-Ga-Cl}} = -36.4 + 0.685T - 6.017 \times 10^{-4}T^2
$$

Eqn. (6) was used for determining the vapour pressure of the complex in the experiments with excess CoCl₄ (Table 1, Exp. 4–9).

Using a non-linear regression analysis program by Hertzberg,1 previously described by Øye and Gruen,4 the experiments were best described by the following complexing equilibrium:

$$
\text{CoCl}_4(s) + \text{Ga}_3\text{Cl}_6(g) = \text{CoGa}_3\text{Cl}_6(g)^* \tag{7}
$$

$$
P_{\text{CoGaCl}_6} = P_{\text{GaCl}_3} \exp \left( \frac{1.936 - 2295}{T_{\text{min}}} \right) \tag{8}
$$

corresponding to $\Delta H^\circ = 43.9 \pm 0.7$ kJ and $\Delta S^\circ = 36.9 \pm 1.1 \text{ J K}^{-1}$ at 673 K. The standard deviation in the estimate was 9.5%. The pressure of the dimer, $P_{\text{CoCl}_4}$ was calculated by the ideal-gas law using the weighed-in amount of GaCl₃ and correcting for the amount consumed in the complexing and dissociation reaction:4

$$
\text{Ga}_3\text{Cl}_6(g) = 2\text{GaCl}_3(g), \log_{10}K(\text{atm}) = -4595/T + 7.066 \tag{9}
$$

DISCUSSION

The expected absence of the side reaction due to attack of the quartz cuvette [eqn. (3)] by using GaCl₃ instead of AlCl₃ was experimentally verified. A loaded cell was left at 425 °C for 5 days without any detectable spectral changes. Even with this excellent stability and good reproducibility shown by repeated experiments, the fit to the model equations was not any better than the results with AlCl₃.1 The reason may be that some GaCl₃ after all did evaporate at the filling of the cell.

Assuming constant temperature and a one-species model, the stoichiometric constant according to eqn. (2) may be calculated by using

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*The method does not unambiguously exclude the presence of polynuclear Co-species as Co₄Ga₃Cl₁₆ but they are considered unlikely.

Table 1. Co—Ga—Cl gas complexes. Molar absorptivity at 15.6 kK (Exp. 1–3) and calculated vapour pressure in presence of CoCl₄(s) (Exp. 4–9).

<table>
<thead>
<tr>
<th>Exp. No.</th>
<th>Specification</th>
<th>Tmin (K)</th>
<th>Tbar (K)</th>
<th>Optical density A</th>
<th>Molar absorptivity ( \epsilon_{\text{Co-Ga-Cl}} ) (l/mol cm) (atm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>( P^\circ \text{GaCl} = 1.276 \times 10^{-5} T )</td>
<td>671</td>
<td>677</td>
<td>0.275</td>
<td>188.8</td>
</tr>
<tr>
<td></td>
<td>( l = 14.2 ) cm</td>
<td>721.5</td>
<td>728.5</td>
<td>0.255</td>
<td>147.2</td>
</tr>
<tr>
<td></td>
<td>( V = 76.73 ) cm³</td>
<td>770</td>
<td>778</td>
<td>0.230</td>
<td>132.8</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 1.215 ) mg</td>
<td>808</td>
<td>817</td>
<td>0.210</td>
<td>121.3</td>
</tr>
<tr>
<td>2</td>
<td>( P^\circ \text{GaCl} = 3.417 \times 10^{-5} T )</td>
<td>571</td>
<td>575</td>
<td>0.580</td>
<td>162.3</td>
</tr>
<tr>
<td></td>
<td>( l = 14.2 ) cm</td>
<td>620.5</td>
<td>628.5</td>
<td>0.555</td>
<td>160.9</td>
</tr>
<tr>
<td></td>
<td>( V = 76.73 ) cm³</td>
<td>719.5</td>
<td>728.5</td>
<td>0.490</td>
<td>142.0</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 2.42 ) mg</td>
<td>774</td>
<td>782</td>
<td>0.445</td>
<td>129.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>812</td>
<td>821</td>
<td>0.420</td>
<td>121.8</td>
</tr>
<tr>
<td>3</td>
<td>( P^\circ \text{GaCl} = 7.34 \times 10^{-5} T )</td>
<td>570</td>
<td>571</td>
<td>0.520</td>
<td>183.6</td>
</tr>
<tr>
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<td>( l = 5.0 ) cm</td>
<td>565</td>
<td>564</td>
<td>0.510</td>
<td>150.6</td>
</tr>
<tr>
<td></td>
<td>( V = 14.22 ) cm³</td>
<td>708</td>
<td>709</td>
<td>0.480</td>
<td>141.7</td>
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<tr>
<td></td>
<td>( w_{\text{CoCl}} = 1.26 ) mg</td>
<td>762</td>
<td>763</td>
<td>0.450</td>
<td>132.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>817.5</td>
<td>818.5</td>
<td>0.415</td>
<td>122.6</td>
</tr>
<tr>
<td>4</td>
<td>( P^\circ \text{GaCl} = 0.303 \times 10^{-5} T )</td>
<td>492.5</td>
<td>495.5</td>
<td>0.017</td>
<td>155.3</td>
</tr>
<tr>
<td></td>
<td>( l = 15.0 ) cm</td>
<td>540.5</td>
<td>544.5</td>
<td>0.032</td>
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</tr>
<tr>
<td></td>
<td>( V = 55.04 ) cm³</td>
<td>588.5</td>
<td>593.5</td>
<td>0.061</td>
<td>158.2</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 640.5 ) mg</td>
<td>660</td>
<td>667</td>
<td>0.063</td>
<td>155.0</td>
</tr>
<tr>
<td></td>
<td></td>
<td>692</td>
<td>699</td>
<td>0.059</td>
<td>148.4</td>
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<td></td>
<td>743</td>
<td>751</td>
<td>0.047</td>
<td>138.7</td>
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<tr>
<td></td>
<td></td>
<td>796</td>
<td>805</td>
<td>0.0355</td>
<td>125.1</td>
</tr>
<tr>
<td>5</td>
<td>( P^\circ \text{GaCl} = 0.71 \times 10^{-5} T )</td>
<td>522</td>
<td>525</td>
<td>0.065</td>
<td>157.4</td>
</tr>
<tr>
<td></td>
<td>( l = 15.0 ) cm</td>
<td>574.5</td>
<td>578.5</td>
<td>0.125</td>
<td>158.5</td>
</tr>
<tr>
<td></td>
<td>( V = 55.86 ) cm³</td>
<td>632.5</td>
<td>637.5</td>
<td>0.200</td>
<td>155.8</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 693.5 ) mg</td>
<td>692</td>
<td>699.5</td>
<td>0.230</td>
<td>148.3</td>
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<tr>
<td></td>
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<td>738</td>
<td>745</td>
<td>0.215</td>
<td>140.0</td>
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<tr>
<td></td>
<td></td>
<td>790</td>
<td>798</td>
<td>0.180</td>
<td>127.1</td>
</tr>
<tr>
<td>6</td>
<td>( P^\circ \text{GaCl} = 1.455 \times 10^{-5} T )</td>
<td>535</td>
<td>538</td>
<td>0.180</td>
<td>155.0</td>
</tr>
<tr>
<td></td>
<td>( l = 15.1 ) cm</td>
<td>583</td>
<td>587</td>
<td>0.325</td>
<td>158.4</td>
</tr>
<tr>
<td></td>
<td>( V = 44.06 ) cm³</td>
<td>628</td>
<td>633</td>
<td>0.405</td>
<td>156.1</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 677 ) mg</td>
<td>677</td>
<td>683</td>
<td>0.630</td>
<td>150.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>726</td>
<td>733</td>
<td>0.075</td>
<td>142.4</td>
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<td></td>
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<td>774.5</td>
<td>782.5</td>
<td>0.845</td>
<td>131.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>817</td>
<td>826</td>
<td>0.580</td>
<td>118.9</td>
</tr>
<tr>
<td>7</td>
<td>( P^\circ \text{GaCl} = 1.50 \times 10^{-5} T )</td>
<td>585</td>
<td>588</td>
<td>0.115</td>
<td>158.4</td>
</tr>
<tr>
<td></td>
<td>( l = 5.0 ) cm</td>
<td>643.5</td>
<td>644.5</td>
<td>0.190</td>
<td>155.1</td>
</tr>
<tr>
<td></td>
<td>( V = 15.12 ) cm³</td>
<td>700</td>
<td>701</td>
<td>0.250</td>
<td>148.1</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 810.5 ) mg</td>
<td>756</td>
<td>757</td>
<td>0.235</td>
<td>137.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>810.5</td>
<td>811.5</td>
<td>0.210</td>
<td>123.2</td>
</tr>
<tr>
<td>8</td>
<td>( P^\circ \text{GaCl} = 2.94 \times 10^{-5} T )</td>
<td>541</td>
<td>542</td>
<td>0.160</td>
<td>158.1</td>
</tr>
<tr>
<td></td>
<td>( l = 5.0 ) cm</td>
<td>591.5</td>
<td>592.5</td>
<td>0.290</td>
<td>158.2</td>
</tr>
<tr>
<td></td>
<td>( V = 14.84 ) cm³</td>
<td>646</td>
<td>647</td>
<td>0.470</td>
<td>154.9</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 699 ) mg</td>
<td>699</td>
<td>700</td>
<td>0.625</td>
<td>148.3</td>
</tr>
<tr>
<td></td>
<td></td>
<td>706</td>
<td>707</td>
<td>0.020</td>
<td>147.1</td>
</tr>
<tr>
<td></td>
<td></td>
<td>752</td>
<td>753</td>
<td>0.700</td>
<td>138.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td>808</td>
<td>809</td>
<td>0.890</td>
<td>124.0</td>
</tr>
<tr>
<td>9</td>
<td>( P^\circ \text{GaCl} = 7.42 \times 10^{-5} T )</td>
<td>564</td>
<td>568</td>
<td>(2.27) a</td>
<td>158.6</td>
</tr>
<tr>
<td></td>
<td>( l = 15.0 ) cm</td>
<td>604.5</td>
<td>609</td>
<td>(3.10)</td>
<td>157.6</td>
</tr>
<tr>
<td></td>
<td>( V = 5 ) cm³</td>
<td>643.5</td>
<td>649</td>
<td>(4.05)</td>
<td>154.7</td>
</tr>
<tr>
<td></td>
<td>( w_{\text{CoCl}} = 672 ) mg</td>
<td>672</td>
<td>678</td>
<td>(4.50)</td>
<td>151.4</td>
</tr>
</tbody>
</table>

a Uncertain values.
the experiments with excess CoCl₂ at different Ga₂Cl₄ pressures. Referred to exp. 8 (Table 1),
typical values between 0.96 and 1.08 are obtained at 394 °C, while slightly higher values
were found at the lowest temperatures. In spite of these deviations from the value n₁ = 1,
corresponding to CoGa₃Cl₄(g), a two-species model
[e.g. CoGa₃Cl₄(g) - CoGa₅Cl₁₁(g)] or
CoGa₅Cl₁₁(g) - CoGa₅Cl₁₄(g)] did not improve
the standard deviation relative to a one-species model with CoGa₃Cl₄(g) present. In conclusion,
CoGa₃Cl₄(g) is considered to be the main gas
species, but small amounts of CoGaCl₄(g),
CoGa₅Cl₁₁(g) or CoGa₅Cl₁₄(g) may be present.

Referring to Table 1, Exp. 4 and 5, it may
seem curious that P_{CoGaCl₄} is decreasing with
increasing temperature for the very highest
temperatures in spite of a positive ΔH° of the
reaction given by eqn. (7). This is, however,
consistent with the present model given by
eqns. (7) and (8) and is simply due to dissociation
of Ga₂Cl₄ [eqn. (9)].

The spectral data do not lend themselves to
any unambiguous conclusion as to the geo-
metric arrangement of chloride around the
cobalt. The molar absorptivity is between
the values one might expect for the octahedral
and tetrahedral geometries, however, closer to
those of octahedral geometry. The variation
with temperature, however, is similar to that
for a tetrahedral species,⁴,¹⁰ and thus the octa-

hedral coordination of Co²⁺ is considered highly
unlikely. However, the possibilities of other
geometries cannot be excluded.

The present experimental results for CoCl₂ -
GaCl₄ are parallel to some very recent results
on CoCl₂ - AlCl₃ gas complexes by Dell'Anna
and Emmenegger,⁶ Papatheodorou,¹¹ Thistle-
thewaite and Ciach,¹⁴ as well as a previous
study by Dewing.¹³ They all find the main gas
complex to be CoAl₂Cl₆ (as compared to
CoGa₂Cl₄), and the spectral results are all very
similar for the Al as well as the Ga containing
complex. The main peak is found around 16 kK
with a decrease in molar absorptivity with in-
creasing temperature.

Table 2 gives the thermodynamic functions
for gas complexation of CoCl₂(s) as well as the
dissociation of Al₂Cl₄(g) and Ga₂Cl₄(g). The
entropy of dissociation of Al₂Cl₄(g) and Ga₂Cl₄(g)
is seen to be the same. The entropy of complexa-
tion of CoCl₂(s) with either Al₂Cl₄(g) or
Ga₂Cl₄(g) is also seen to be equal, confirming
the assumption of a similar structure of the two
gas complexes. The remarkable feature of
Table 2, however, is that the enthalpy of
complexation is equal for the Al and the Ga
containing complexes, in spite of the fact that the
enthalpies of dissociation of Al₂Cl₄(g) and
Ga₂Cl₄ differ by 30 kJ. This suggests that the
Al₂Cl₄ and Ga₂Cl₄ molecules remain essentially
intact and do not dissociate, the CoCl₂ co-

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Al₂Cl₄(g) = 2AlCl₃(g)</td>
<td>14 673</td>
<td>118</td>
<td>135</td>
<td>0.008</td>
</tr>
<tr>
<td>Ga₂Cl₄(g) = 2GaCl₃(g)</td>
<td>8 673</td>
<td>88</td>
<td>135</td>
<td>1.7</td>
</tr>
<tr>
<td>CoCl₂(s) + Al₂Cl₄(g) = CoAl₂Cl₆(g)</td>
<td>13 750</td>
<td>53.5</td>
<td>51.9</td>
<td>11.4 x 10⁻⁴</td>
</tr>
<tr>
<td>6a 673</td>
<td>43.1 ± 2.2</td>
<td>39.3 ± 2.8</td>
<td>5.1 x 10⁻⁴</td>
<td></td>
</tr>
<tr>
<td>11 673</td>
<td>41.8 ± 0.8</td>
<td>41.0 ± 1.3</td>
<td>7.9 x 10⁻⁴</td>
<td></td>
</tr>
<tr>
<td>12 673</td>
<td>42.0 ± 0.7</td>
<td>45.0</td>
<td>12.3 x 10⁻⁴</td>
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</tr>
<tr>
<td>CoCl₂(s) + Ga₂Cl₄(g) = CoGa₂Cl₄(g)</td>
<td>This work 673</td>
<td>43.9 ± 0.7</td>
<td>36.9 ± 1.1</td>
<td>3.3 x 10⁻⁴</td>
</tr>
</tbody>
</table>

a Mean value of three methods.
ordinating at the end of the molecule, for instance approximately tetrahedrally in fashion II rather than I:

\[
\begin{align*}
\text{Cl} & \quad \text{Cl} \\
\text{Cl} & \quad \text{Co} \quad \text{Cl} \\
\text{Cl} & \quad \text{Cl} \\
\text{Cl} & \quad \text{Cl} \\
\text{Cl} & \quad \text{Ga} \\
\text{Cl} & \quad \text{Cl} \\
\text{Cl} & \quad \text{Ga} \\
\text{Cl} & \quad \text{Cl}
\end{align*}
\]  

(10)

Dell’Anna and Emmenegger 8 describe the Co coordination in CoAl4Cl4 as tetrahedral, whereas Papatheodorou 11 and Thistlethwaite and Ciach 18 describe it as mainly octahedral. Papatheodorou 11 explains the increase of molar absorptivity with decreasing temperature as due to small amounts of tetrahedrally coordinated Co^{3+} in equilibrium with the octahedral form, the tetrahedral coordination being more prevalent at lower temperatures.

We are at variance with this interpretation because it infers that the equilibrium

\[
\text{Co(tettr)Al}_4\text{Cl}_4(g) = \text{Co(oct)Al}_4\text{Cl}_4(g)
\]  

(11)

has \( \Delta H > 0 \) and, since \( K > 1 \), \( \Delta S > 0 \). The signs of the thermodynamic functions are both opposite to what is expected. Whether structure I or II is assumed for a tetrahedral structure, a tetrahedral-octahedral transformation will result in the formation of two additional Co – Cl bonds with an expected negative \( \Delta H \). The vibrational entropy cannot be anticipated to counteract the negative rotational entropy 11 for eqn. (11). The main contribution to the vibrational entropy is given by the low frequency torsional vibrations and they are expected to be lower for the more loosely bonded AlCl_4^- in the tetrahedral coordination, hence resulting in \( \Delta S < 0 \) for eqn. (11) both due to rotational and vibrational entropy.

It should, however, be stressed that the coordination equilibria in liquid and gaseous aluminium chloride still are intriguing. Co^{3+} and Ti^{2+} both have a similarly low octahedral site stabilization energy, but Ti^{4+} coordinates octahedrally in both the liquid and gaseous aluminium chloride, while octahedral coordination of Co^{3+} is only found in liquid aluminium chloride. 10

From a practical point of view it should be mentioned in conclusion that in spite of the similar thermodynamic properties of gas complex formation, GaCl_4 becomes less effective at higher temperatures due to its ease of dissociation relative to AlCl_4. The greater stability of GaCl_4 in the presence of SiO_2 may, however, make it more attractive for detailed optical studies.

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