# Supplementary information

**Contents**



## 1 Diffusion measurements

Table 1: Diffusion coefficients of the $(bpy)_3$ molecules of $Co^{2+}(bpy)_3(NTf_2)_2$ samples. Solvent is MPN.

| **Samples** / **T** (K) | $\mathbf{Co^{2+}(bpy)_3(NTf_2)_2}$ **D** ($\times 10^{-10}$ $m^2$ $s^{-1}$) | | |
|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** |
| 293 | 3.29 | 2.89 | 2.71 |
| 303 | 4.04 | 3.57 | 3.41 |
| 313 | 5.07 | 4.40 | 4.25 |
| 323 | 6.23 | 5.51 | 5.25 |
| 333 | 7.44 | 6.82 | 6.40 |
| 343 | 9.02 | 8.03 | 7.64 |
| 353 | 10.8 | 9.66 | 9.25 |

Table 2: Diffusion coefficients of the $(bpy)_3$ molecules of $Co^{3+}(bpy)_3(NTf_2)_3$ samples. Solvent is MPN.

| **Samples** / **T** (K) | $\mathbf{Co^{3+}(bpy)_3(NTf_2)_3}$ **D** ($\times 10^{-10}$ $m^2$ $s^{-1}$) | | |
|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** |
| 293 | 2.56 | 2.26 | 1.89 |
| 303 | 3.27 | 2.82 | 2.29 |
| 313 | 4.07 | 3.54 | 2.90 |
| 323 | 5.06 | 4.36 | 3.56 |
| 333 | 6.16 | 5.35 | 4.37 |
| 343 | 7.22 | 6.23 | 5.15 |
| 353 | 8.80 | 7.33 | 6.26 |

Table 3: Diffusion coefficients of the $(bpy)_3$ molecules of $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ samples. Solvent is MPN.

| Samples / T (K) | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ **D** ($\times 10^{-10}$ m$^2$ s$^{-1}$) | | | |
|---|---|---|---|---|
| | ***0.05 M*** | | ***0.10 M*** | |
| | ***$Co^{2+}(bpy)_3$*** | ***$Co^{3+}(bpy)_3$*** | ***$Co^{2+}(bpy)_3$*** | ***$Co^{2+}(bpy)_3$*** |
| 293 | 2.58 | 2.32 | 2.33 | 1.99 |
| 303 | 3.13 | 2.99 | 2.82 | 2.41 |
| 313 | 3.97 | 3.72 | 3.39 | 2.96 |
| 323 | 4.87 | 4.46 | 4.17 | 3.66 |
| 333 | 5.93 | 5.44 | 5.05 | 4.56 |
| 343 | 7.36 | 6.31 | 6.02 | 5.36 |
| 353 | 8.69 | 7.45 | 7.17 | 6.36 |

Table 4: Diffusion coefficients of the MPN molecules of all samples.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ **D** ($\times 10^{-9}$ m$^2$ s$^{-1}$) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ **D** ($\times 10^{-9}$ m$^2$ s$^{-1}$) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ **D** ($\times 10^{-9}$ m$^2$ s$^{-1}$) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 0.93 | 0.87 | 0.81 | 0.90 | 0.80 | 0.71 | 0.83 | 0.72 |
| 303 | 1.16 | 1.08 | 1.00 | 1.13 | 1.01 | 8.89 | 1.04 | 0.90 |
| 313 | 1.40 | 1.31 | 1.25 | 1.37 | 1.26 | 1.09 | 1.28 | 1.11 |
| 323 | 1.69 | 1.60 | 1.51 | 1.64 | 1.51 | 1.33 | 1.54 | 1.34 |
| 333 | 2.00 | 1.89 | 1.80 | 1.95 | 1.79 | 1.60 | 1.83 | 1.61 |
| 343 | 2.35 | 2.25 | 2.12 | 2.29 | 2.12 | 1.87 | 2.15 | 1.91 |
| 353 | 2.75 | 2.57 | 2.48 | 2.63 | 2.44 | 2.17 | 2.47 | 2.21 |

Table 5: Diffusion coefficients of the NTf2 molecules of all samples.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ **D** ($\times 10^{-10}$ m$^2$ s$^{-1}$) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ **D** ($\times 10^{-10}$ m$^2$ s$^{-1}$) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ **D** ($\times 10^{-10}$ m$^2$ s$^{-1}$) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 4.05 | 3.86 | 3.37 | 3.74 | 3.02 | 2.68 | 3.26 | 2.75 |
| 303 | 4.75 | 4.55 | 4.13 | 4.52 | 3.87 | 3.35 | 4.03 | 3.42 |
| 313 | 6.08 | 5.70 | 5.01 | 5.54 | 4.78 | 4.06 | 5.15 | 4.33 |
| 323 | 7.32 | 6.68 | 6.10 | 6.53 | 5.79 | 5.06 | 6.14 | 5.11 |
| 333 | 8.59 | 8.10 | 7.33 | 7.78 | 6.98 | 6.07 | 7.33 | 6.24 |
| 343 | 1.02 | 9.31 | 8.65 | 9.11 | 8.17 | 7.08 | 8.54 | 7.26 |
| 353 | 1.14 | 1.09 | 9.87 | 10.20 | 9.49 | 8.37 | 9.61 | 8.53 |

## 2 Viscosity measurements

Table 6: Viscosities of the liquid electrolytes.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ η (mPa s) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ η (mPa s) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ η (mPa s) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 1.31 | 1.47 | 1.62 | 1.39 | 1.66 | 1.92 | 1.57 | 2.20 |
| 303 | 1.11 | 1.24 | 1.36 | 1.17 | 1.40 | 1.60 | 1.32 | 1.82 |
| 313 | 0.95 | 1.07 | 1.16 | 1.02 | 1.20 | 1.36 | 1.13 | 1.54 |
| 323 | 0.83 | 0.93 | 1.01 | 0.89 | 1.04 | 1.17 | 0.99 | 1.32 |
| 333 | 0.74 | 0.82 | 0.89 | 0.79 | 0.92 | 1.03 | 0.87 | 1.16 |
| 343 | 0.66 | 0.74 | 0.79 | 0.71 | 0.82 | 0.91 | 0.78 | 1.04 |
| 353 | 0.59 | 0.66 | 0.70 | 0.63 | 0.74 | 0.81 | 0.70 | 0.94 |

## 3 Ionic conductivity measurements

Table 7: Ionic conductivities of the liquid electrolytes.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ σ ($\times 10^{-3}$ S cm$^{-1}$) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ σ ($\times 10^{-3}$ S cm$^{-1}$) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ σ ($\times 10^{-3}$ S cm$^{-1}$) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 2.26 | 3.78 | 4.64 | 2.90 | 4.54 | 5.40 | 4.05 | 5.61 |
| 303 | 2.61 | 4.37 | 5.43 | 3.33 | 5.27 | 6.31 | 4.71 | 6.57 |
| 313 | 2.96 | 4.96 | 6.22 | 3.75 | 5.96 | 7.18 | 5.34 | 7.58 |
| 323 | 3.30 | 5.54 | 6.85 | 4.15 | 6.60 | 8.06 | 5.96 | 8.59 |
| 333 | 3.62 | 6.17 | 7.67 | 4.52 | 7.23 | 8.88 | 6.57 | 9.52 |
| 343 | 4.00 | 6.75 | 8.35 | 4.88 | 7.83 | 9.80 | 7.14 | 10.40 |
| 353 | 4.30 | 7.32 | 9.05 | 5.21 | 8.38 | 10.49 | 7.70 | 11.34 |

## 4 Relaxation time measurements

Table 8: $^1$H NMR $T_1$ relaxation times of the MPN protons.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $T_1$ (ms) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $T_1$ (ms) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ $T_1$ (ms) | | Pure MPN $T_1$ (ms) |
|---|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | |
| 293 | 1120 | 751 | 590 | 3862 | 3205 | 2750 | 989 | 880 | 2843 |
| 303 | 1226 | 818 | 655 | 4621 | 3648 | 3201 | 1137 | 1022 | 3318 |
| 313 | 1513 | 950 | 719 | 5372 | 4265 | 3631 | 1260 | 1078 | 3770 |
| 323 | 1610 | 1010 | 785 | 6110 | 4879 | 4166 | 1384 | 1288 | 4282 |
| 333 | 1853 | 1149 | 869 | 7002 | 5924 | 4799 | 1533 | 1428 | 4910 |
| 343 | 1984 | 1254 | 964 | 7772 | 6778 | 5718 | 1716 | 1608 | 5520 |
| 353 | 2262 | 1386 | 1031 | 8608 | 7375 | 6409 | 1903 | 1657 | 6173 |

Table 9: $^{19}F$ NMR $T_1$ relaxations measured from the $NTf_2^-$ anions.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $T_1$ (ms) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $T_1$ (ms) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ $T_1$ (ms) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 675 | 484 | 423 | 2535 | 2287 | 2056 | 774 | 587 |
| 303 | 766 | 543 | 470 | 3137 | 2818 | 2304 | 867 | 677 |
| 313 | 834 | 603 | 510 | 3499 | 3175 | 2687 | 962 | 741 |
| 323 | 916 | 655 | 542 | 4015 | 3486 | 2974 | 1051 | 809 |
| 333 | 966 | 704 | 588 | 4229 | 3748 | 3232 | 1145 | 897 |
| 343 | 1055 | 753 | 639 | 4418 | 4066 | 3544 | 1239 | 960 |
| 353 | 1115 | 799 | 680 | 4801 | 4396 | 3648 | 1335 | 1040 |

Table 10: $^1H$ $T_1$ values of the $(bpy)_3$ protons of the 0.05 M $Co^{3+}(bpy)_3(NTf_2)_3$ and 0.05 M $Co^{2+}(bpy)_3(NTf_2)_2$ samples.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $T_1$ (ms) | | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $T_1$ (ms) | | | |
|---|---|---|---|---|---|---|---|---|
| | ***$H_1$*** | ***$H_2$*** | ***$H_3$*** | ***$H_4$*** | ***$H_1$*** | ***$H_2$*** | ***$H_3$*** | ***$H_4$*** |
| 293 | 16.84 | 38.08 | 25.79 | 1.38 | 880.20 | 1366.00 | 1316.01 | 1325.01 |
| 303 | 21.95 | 49.37 | 29.61 | 1.67 | 925.08 | 1470.01 | 1407.99 | 1426.00 |
| 313 | 25.27 | 48.16 | 32.83 | 1.85 | 989.85 | 1610.02 | 1498.99 | 1527.99 |
| 323 | 28.61 | 56.92 | 36.42 | 1.99 | 1044.00 | 1716.01 | 1585.99 | 1611.02 |
| 333 | 31.09 | 62.05 | 40.53 | 2.24 | 1094.99 | 1822.01 | 1680.00 | 1708.99 |
| 343 | 35.92 | 61.31 | 44.61 | 2.58 | 1145.99 | 1915.01 | 1783.98 | 1823.01 |
| 353 | 39.89 | 72.31 | 48.46 | 2.82 | 1269.99 | 2109.02 | 2009.00 | 2037.00 |

Table 11: $^{19}F$ NMR $T_2$ relaxations measured from the $NTf_2^-$ anions

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $T_2$ (ms) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $T_2$ (ms) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ $T_2$ (ms) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 487 | 306 | 303 | 1783 | 1650 | 1255 | 527 | 314 |
| 303 | 549 | 347 | 331 | 1991 | 1788 | 1500 | 596 | 369 |
| 313 | 611 | 387 | 360 | 2116 | 1867 | 1761 | 654 | 407 |
| 323 | 646 | 428 | 395 | 2181 | 2225 | 1923 | 702 | 456 |
| 333 | 683 | 449 | 413 | 1985 | 2488 | 2196 | 743 | 477 |
| 343 | 679 | 462 | 420 | 1661 | 1962 | 1726 | 758 | 502 |
| 353 | 682 | 464 | 430 | 1558 | 1720 | 1498 | 775 | 513 |

## 5 Walden Data

Table 12: Molar conductivities of the liquid electrolytes.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $\Lambda_m$ (S cm$^2$ mol$^{-1}$) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $\Lambda_m$ (S cm$^2$ mol$^{-1}$) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ $\Lambda_m$ (S cm$^2$ mol$^{-1}$) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 45.12 | 37.81 | 30.93 | 58.04 | 45.42 | 36.01 | 40.53 | 28.03 |
| 303 | 52.11 | 43.66 | 36.17 | 66.69 | 52.69 | 42.04 | 47.14 | 32.84 |
| 313 | 59.13 | 49.60 | 41.49 | 74.91 | 59.57 | 47.90 | 53.40 | 37.88 |
| 323 | 65.95 | 55.40 | 45.68 | 82.90 | 66.04 | 53.76 | 59.57 | 42.95 |
| 333 | 72.40 | 61.74 | 51.15 | 90.32 | 72.29 | 59.20 | 65.68 | 47.61 |
| 343 | 79.97 | 67.55 | 55.66 | 97.57 | 78.25 | 65.32 | 71.42 | 52.00 |
| 353 | 86.06 | 73.18 | 60.33 | 104.22 | 83.78 | 69.94 | 76.98 | 56.72 |

Table 13: Inverse viscosities of the liquid electrolytes.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $\eta^{-1}$ (P$^{-1}$) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $\eta^{-1}$ (P$^{-1}$) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ $\eta^{-1}$ (P$^{-1}$) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 76.63 | 67.89 | 61.65 | 72.05 | 60.20 | 52.08 | 63.68 | 45.41 |
| 303 | 90.33 | 80.52 | 73.42 | 85.18 | 71.68 | 62.62 | 75.75 | 54.92 |
| 313 | 104.82 | 93.81 | 86.13 | 98.04 | 83.61 | 73.58 | 88.32 | 65.04 |
| 323 | 119.90 | 107.64 | 99.30 | 112.74 | 95.79 | 85.18 | 101.50 | 75.49 |
| 333 | 135.87 | 122.70 | 112.99 | 127.06 | 108.34 | 97.47 | 114.84 | 86.03 |
| 343 | 152.21 | 135.69 | 127.39 | 141.84 | 122.10 | 110.25 | 128.87 | 96.19 |
| 353 | 168.92 | 151.52 | 142.25 | 158.23 | 136.05 | 123.15 | 143.12 | 106.87 |

Table 14: Walden product of the liquid electrolytes.

| Samples / T (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ $\Lambda_m.\eta$ (S cm$^2$ mol$^{-1}$ Pa s) | | | $Co^{3+}(bpy)_3(NTf_2)_3$ $\Lambda_m.\eta$ (S cm$^2$ mol$^{-1}$ Pa s) | | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ $\Lambda_m.\eta$ (S cm$^2$ mol$^{-1}$ Pa s) | |
|---|---|---|---|---|---|---|---|---|
| | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** | ***0.15 M*** | ***0.05 M*** | ***0.10 M*** |
| 293 | 0.05888 | 0.0557 | 0.05017 | 0.08056 | 0.07545 | 0.06913 | 0.06365 | 0.06171 |
| 303 | 0.05769 | 0.05423 | 0.04926 | 0.0783 | 0.0735 | 0.06714 | 0.06223 | 0.05979 |
| 313 | 0.05641 | 0.05288 | 0.04817 | 0.07641 | 0.07125 | 0.06509 | 0.06047 | 0.05824 |
| 323 | 0.055 | 0.05146 | 0.046 | 0.07353 | 0.06895 | 0.06312 | 0.05869 | 0.0569 |
| 333 | 0.05328 | 0.05032 | 0.04527 | 0.07108 | 0.06672 | 0.06074 | 0.05719 | 0.05534 |
| 343 | 0.05254 | 0.04978 | 0.04369 | 0.06879 | 0.06409 | 0.05924 | 0.05542 | 0.05405 |
| 353 | 0.05095 | 0.0483 | 0.04241 | 0.06587 | 0.06158 | 0.05679 | 0.05379 | 0.05308 |

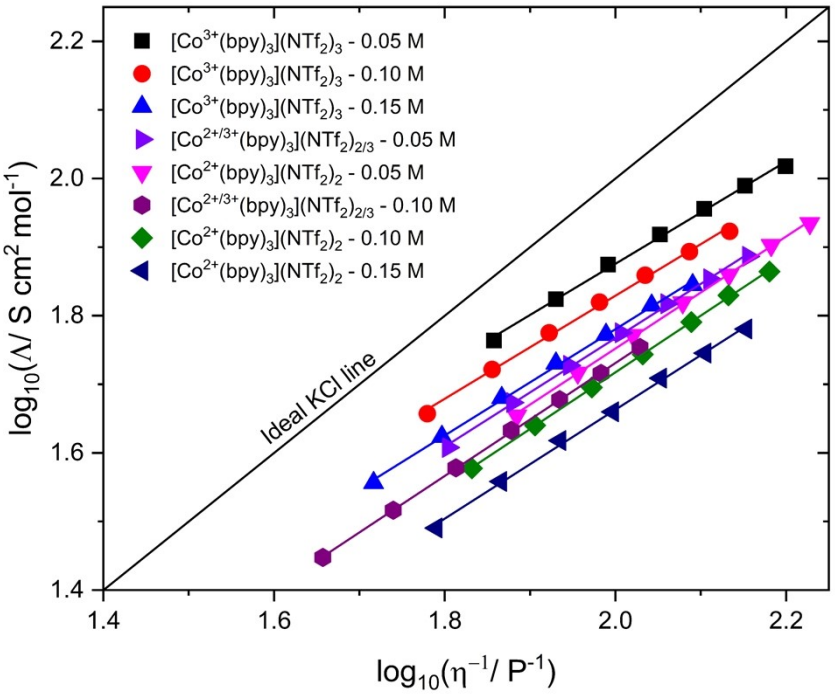


*Figure 1: Walden plots for liquid electrolytes.*

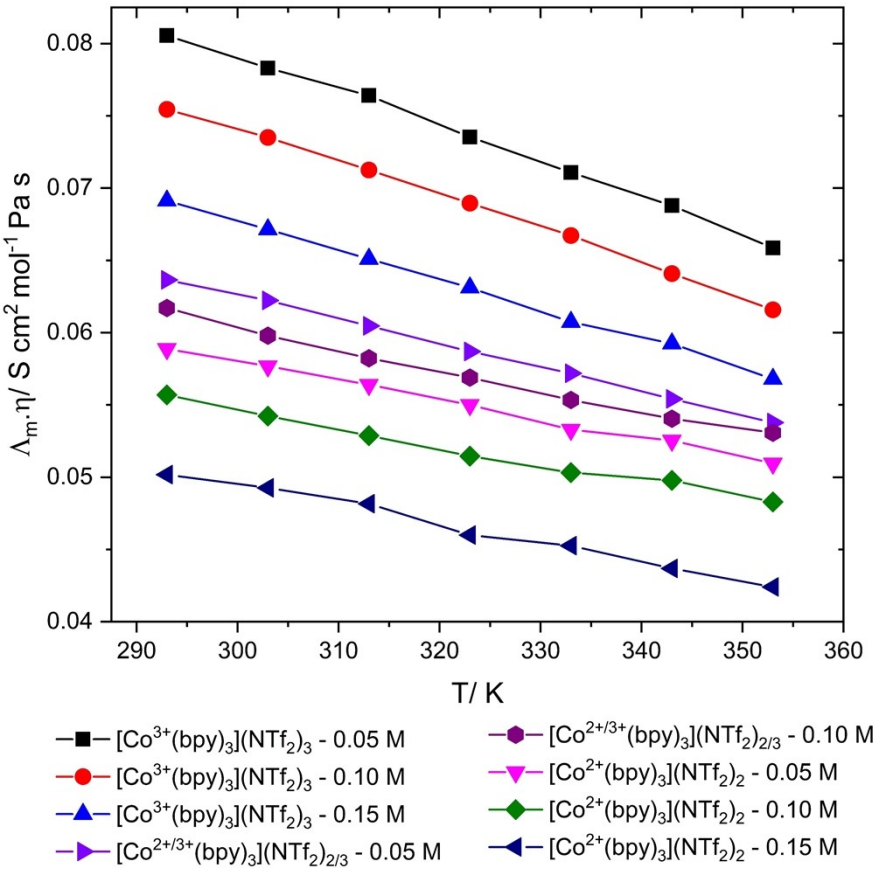


*Figure 2: The temperature dependence of the Walden products of the liquid electrolytes.*

## 6 Nernst–Einstein equation

The experimental molar conductivity $\Lambda_{exp}$and the molar conductivity calculated from the Nernst–Einstein equation ($\Lambda_{NE}$) are compared:

$$\Lambda_{NE} = \frac{F^2}{RT}(z_i^2 D_i)$$

$$\Lambda_{NE} = \frac{F^2}{RT}(z_{cation}^{2} D^{+} + z_{anion}^{2} D^{-})$$

$D^+$ = diffusion coefficients of $Co^{2+}(bpy)_3$ or $Co^{3+}(bpy)_3$
$D^-$ = diffusion coefficient of $NTf_2^-$

Table 15: $\Lambda_{exp}$ and $\Lambda_{NE}$ values of $Co^{2+}(bpy)_3(NTf_2)_2$ samples.

| Temperature (K) | $Co^{2+}(bpy)_3(NTf_2)_2$ -0.05 M | | $Co^{2+}(bpy)_3(NTf_2)_2$ -0.10 M | | $Co^{2+}(bpy)_3(NTf_2)_2$ -0.15 M | |
|---|---|---|---|---|---|---|
| | $\Lambda_{exp}$ | $\Lambda_{NE}$ | $\Lambda_{exp}$ | $\Lambda_{NE}$ | $\Lambda_{exp}$ | $\Lambda_{NE}$ |
| 293 | 45.12 | 65.77 | 37.81 | 58.94 | 30.93 | 54.27 |
| 303 | 52.11 | 77.26 | 43.66 | 69.58 | 36.17 | 65.58 |
| 313 | 59.13 | 94.32 | 49.60 | 83.31 | 41.49 | 78.65 |
| 323 | 65.95 | 111.80 | 55.40 | 99.50 | 45.68 | 93.97 |
| 333 | 72.40 | 128.93 | 61.74 | 118.90 | 51.15 | 110.74 |
| 343 | 80.00 | 150.93 | 67.55 | 135.31 | 55.66 | 127.99 |
| 353 | 86.06 | 172.52 | 73.18 | 156.98 | 60.33 | 148.72 |

Table 16: $\Lambda_{exp}$ and $\Lambda_{NE}$ values of $Co^{3+}(bpy)_3(NTf_2)_3$ samples.

| Temperature (K) | $Co^{3+}(bpy)_3(NTf_2)_3$ -0.05 M | | $Co^{3+}(bpy)_3(NTf_2)_3$ -0.10 M | | $Co^{3+}(bpy)_3(NTf_2)_3$ -0.15 M | |
|---|---|---|---|---|---|---|
| | $\Lambda_{exp}$ | $\Lambda_{NE}$ | $\Lambda_{exp}$ | $\Lambda_{NE}$ | $\Lambda_{exp}$ | $\Lambda_{NE}$ |
| 293 | 58.04 | 102.29 | 45.42 | 89.24 | 36.01 | 75.30 |
| 303 | 66.69 | 125.41 | 52.69 | 108.19 | 42.04 | 88.41 |
| 313 | 74.91 | 150.92 | 59.57 | 131.15 | 47.90 | 108.02 |
| 323 | 82.90 | 180.62 | 66.04 | 156.09 | 53.76 | 128.73 |
| 333 | 90.32 | 212.59 | 72.29 | 185.34 | 59.20 | 152.78 |
| 343 | 97.57 | 241.78 | 78.25 | 209.65 | 65.32 | 174.38 |
| 353 | 104.22 | 283.77 | 83.78 | 239.45 | 69.94 | 205.28 |

Table 17: $\Lambda_{exp}$ and $\Lambda_{NE}$ values of $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ samples.

| Temperature (K) | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ - 0.05 M | | $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ - 0.10 M | |
|---|---|---|---|---|
| | $\Lambda_{exp}$ | $\Lambda_{NE}$ | $\Lambda_{exp}$ | $\Lambda_{NE}$ |
| 293 | 40.53 | 131.66 | 28.03 | 114.54 |
| 303 | 47.14 | 160.71 | 32.84 | 134.64 |
| 313 | 53.40 | 194.92 | 37.88 | 159.23 |
| 323 | 59.57 | 227.82 | 42.95 | 189.67 |
| 333 | 65.68 | 268.92 | 47.61 | 226.72 |
| 343 | 71.42 | 309.35 | 52.00 | 259.60 |
| 353 | 76.98 | 353.37 | 56.72 | 299.51 |

## 7 Stokes-Einstein radii at 293 K and 353 K by changing the arbitrary constant.

In the Stokes-Einstein equation, the constant may also be varied depending on systems studied. The constant was reported to 3~4 for organic electrolyte solutions. Various arbitrary constants were used to understand which one was better for ion radius calculations.

$$R_H = \frac{k_B T}{c\pi\eta D}$$

Table 18: Stokes-Einstein radii at 293 K and 353 K with constant, 6.

| **Sample** | **$R_H$ ($NTf_2^-$) ($\times 10^{-10}$ m )** | | **$R_H$ ($[Co(bpy)_3]^{2+}$) ($\times 10^{-10}$ m )** | | **$R_H$ ($[Co(bpy)_3]^{3+}$) ($\times 10^{-10}$ m )** | |
|---|---|---|---|---|---|---|
| | ***293 K*** | ***353 K*** | ***293 K*** | ***353 K*** | ***293 K*** | ***353 K*** |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.05 M | 4.1 | 3.8 | 5.0 | 4.1 | - | - |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.10 M | 3.8 | 3.6 | 5.0 | 4.1 | - | - |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.15 M | 3.9 | 3.7 | 4.9 | 4.0 | - | - |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.05 M | 4.1 | 4.0 | - | - | 6.0 | 4.7 |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.10 M | 4.3 | 3.7 | - | - | 5.7 | 4.8 |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.15 M | 4.2 | 3.8 | - | - | 5.9 | 5.1 |
| $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ 0.05 M | 4.2 | 3.9 | 5.3 | 4.3 | 5.9 | 5.0 |
| $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ 0.10 M | 3.6 | 3.2 | 4.2 | 3.9 | 4.9 | 4.3 |

Table 19: Stokes-Einstein radii at 293 K and 353 K with constant, 5.

| **Sample** | **$R_H$ ($NTf_2^-$) ($\times 10^{-10}$ m )** | | **$R_H$ ($[Co(bpy)_3]^{2+}$) ($\times 10^{-10}$ m )** | | **$R_H$ ($[Co(bpy)_3]^{3+}$) ($\times 10^{-10}$ m )** | |
|---|---|---|---|---|---|---|
| | ***293 K*** | ***353 K*** | ***293 K*** | ***353 K*** | ***293 K*** | ***353 K*** |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.05 M | 4.9 | 4.6 | 6.0 | 4.9 | - | - |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.10 M | 4.5 | 4.3 | 6.1 | 4.9 | - | - |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.15 M | 4.7 | 4.7 | 5.9 | 4.8 | - | - |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.05 M | 5.0 | 4.8 | - | - | 7.2 | 5.6 |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.10 M | 5.1 | 4.5 | - | - | 6.9 | 5.8 |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.15 M | 5.0 | 4.6 | - | - | 7.1 | 6.1 |
| $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ 0.05 M | 5.0 | 4.6 | 6.4 | 5.1 | 7.1 | 6.0 |
| $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ 0.10 M | 4.3 | 3.9 | 5.0 | 4.6 | 5.9 | 5.2 |

Table 20: Stokes-Einstein radii at 293 K and 353 K with constant, 4.

| **Sample** | **$R_H$ ($NTf_2^-$) ($\times 10^{-10}$ m )** | | **$R_H$ ($[Co(bpy)_3]^{2+}$) ($\times 10^{-10}$ m )** | | **$R_H$ ($[Co(bpy)_3]^{3+}$) ($\times 10^{-10}$ m )** | |
|---|---|---|---|---|---|---|
| | ***293 K*** | ***353 K*** | ***293 K*** | ***353 K*** | ***293 K*** | ***353 K*** |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.05 M | 6.1 | 5.7 | 7.5 | 6.1 | - | - |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.10 M | 5.7 | 5.4 | 7.6 | 6.1 | - | - |
| $Co^{2+}(bpy)_3(NTf_2)_2$ 0.15 M | 5.9 | 5.6 | 7.3 | 6.0 | - | - |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.05 M | 6.2 | 6.0 | - | - | 9.1 | 7.0 |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.10 M | 6.4 | 5.6 | - | - | 8.6 | 7.2 |
| $Co^{3+}(bpy)_3(NTf_2)_3$ 0.15 M | 6.3 | 5.7 | - | - | 8.9 | 7.6 |
| $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ 0.05 M | 6.3 | 5.8 | 7.9 | 6.4 | 8.8 | 7.5 |
| $Co^{2+/3+}(bpy)_3(NTf_2)_{2/3}$ 0.10 M | 5.3 | 4.9 | 6.3 | 5.8 | 7.4 | 6.5 |