# A Supporting Information

## A.1 Unveiling the interplay: comparing solvent extraction, structure, and stability constants in a phenanthroline diamide system with trivalent lanthanides

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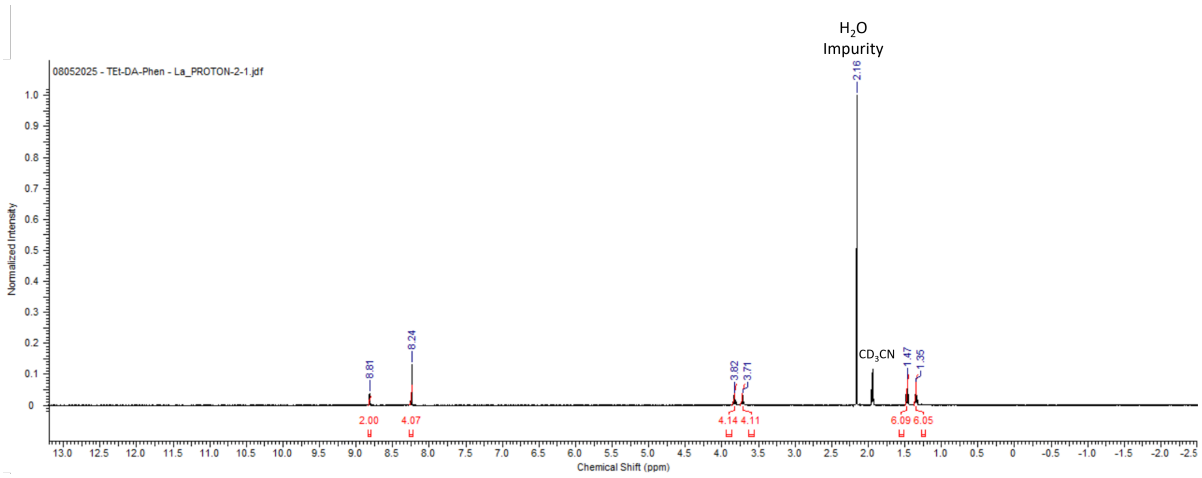


Fig. 1 $^{1}$H NMR of La(TEtDAPhen)$(NO_3)_3$ complex in $CD_3CN$.

The $^{1}$H NMR peaks observed in Figure 1 show two phenanthroline resonances at 8.81 and 8.24 ppm as a result of peak averaging, two resonances at 3.82 and 3.71 ppm which represent the $CH_2$ resonance on the ethyl arms, and two resonances at 1.47 and 1.35 ppm which represent the $CH_3$ resonance on the ethyl arms. The $^{1}$H NMR spectrum show in Figure 2 for the Ce-TEtDAPhen complex shows

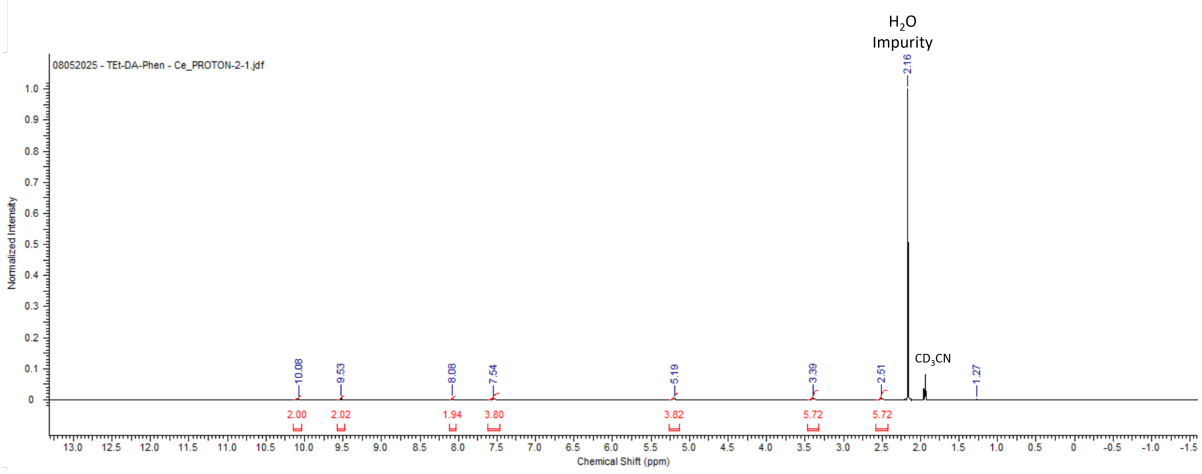


Fig. 2 $^{1}$H NMR of Ce(TEtDAPhen)$(NO_3)_3$ complex in $CD_3CN$.

three phenanthroline resonances at 10.08, 9.53, and 8.08 ppm, two $CH_2$ resonances at 7.54 and 5.19 ppm, and two $CH_3$ resonances at 3.39 and 2.51 ppm.

The $^{1}$H NMR spectrum show in Figure 3 for the Sm-TEtDAPhen complex shows three phenanthroline resonances at 8.94, 8.60, and 8.27 ppm, two $CH_2$ resonances at 4.39 and 4.09 ppm, and one $CH_3$ resonance at 1.68 ppm.

The $^{1}$H NMR spectrum show in Figure 4 for the Yb-TEtDAPhen complex shows paramagnetic features.

Table 1 Crystal data for La(TEtDAPhen)$(NO_3)_3$. The ethyl arms on the ligand had a 50/50 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450883 |
| Empirical formula | $La_{0.5}C_{24}H_{29}N_{6.50}O_{6.50}$ |
| Formula weight | 581.99 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_1$ |
| Space group (number) | $P2_1$, 4 |
| $a$ [{dist_unit}] | 12.5844(8) |
| $b$ [{dist_unit}] | 20.8564(15) |
| $c$ [{dist_unit}] | 20.0610(14) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 98.248(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 5210.9(6) |
| $Z$ | 8 |
| $\rho_{calc}$ [gcm$^3$] | 1.484 |
| $\mu$ [mm$^1$] | 0.477 |
| $F(000)$ | 2392 |
| Crystal size [mm$^3$] | 0.511 x 0.158 x 0.15 |
| Crystal color | Clear yellow |
| Crystal shape | Prism |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 38.000° |
| Reflections collected | 131723 |
| Data / Restraints / Parameters | 26970/ 1 / 1392 |
| Absorption correction $T_{min}/T_{max}$ (method) | 0.6666 / 0.7448 multi-scan |
| Goodness-of-fit on $F^2$ | 1.028 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0349 w$R_2$ = 0.0821 |
| Final $R$ indexes [all data] | $R_1$ = 0.0373 w$R_2$ = 0.0844 |
| Largest peak/hole [eÅ$^3$] | 1.105 / -0.620 |

Table 2 Crystal data for Ce(TEtDAPhen)$(NO_3)_3$. The ethyl arms on the ligand had a 65/35 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450881 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Ce$ |
| Formula weight | 704.62 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.7201(7) |
| $b$ [{dist_unit}] | 17.5441(12) |
| $c$ [{dist_unit}] | 15.8709(11) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.587(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2688.6(3) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.741 |
| $\mu$ [mm$^1$] | 0.936 |
| $F(000)$ | 1412 |
| Crystal size [mm$^3$] | 0.904 x 0.776 x 0.634 |
| Crystal color | Clear yellow |
| Crystal shape | block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2Θ range [°] | 47.262° |
| Reflections collected | 120568 |
| Data / Restraints / Parameters | 8170 / 12 / 398 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.5791/ 0.7449 |
| Goodness-of-fit on $F^2$ | 1.092 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0230 w$R_2$ = 0.0550 |
| Final $R$ indexes [all data] | $R_1$ = 0.0244 w$R_2$ = 0.0559 |
| Largest peak/hole [eÅ$^3$] | 1.422 / -0.926 |

Table 3 Crystal data for Sm(TEtDAPhen)$(NO_3)_3$. The ethyl arms on the ligand had a 60/40 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450523 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Sm$ |
| Formula weight | 714.85 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.5877(7) |
| $b$ [{dist_unit}] | 17.4923(13) |
| $c$ [{dist_unit}] | 15.9192(12) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.539(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2652.5(3) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.790 |
| $\mu$ [mm$^1$] | 1.219 |
| $F(000)$ | 1428 |
| Crystal size [mm$^3$] | 0.267 x 0.174 x 0.138 |
| Crystal color | Clear yellow |
| Crystal shape | Block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 43.998° |
| Reflections collected | 48428 |
| Data / Restraints / Parameters | 6595 / 10 / 388 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.6835 / 0.7426 |
| Goodness-of-fit on $F^2$ | 1.103 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0274 w$R_2$ = 0.0686 |
| Final $R$ indexes [all data] | $R_1$ = 0.0298 w$R_2$ = 0.0709 |
| Largest peak/hole [eÅ$^3$] | 1.530 / -0.761 |

Table 4 Crystal data for Eu(TEtDAPhen)$(NO_3)_3$. The ethyl arm on the ligand had a 60/40 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450882 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Eu$ |
| Formula weight | 716.46 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.5497(7) |
| $b$ [{dist_unit}] | 17.4884(12) |
| $c$ [{dist_unit}] | 15.9305(11) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.558(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2643.1(3) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.800 |
| $\mu$ [mm$^1$] | 1.300 |
| $F(000)$ | 1432 |
| Crystal size [mm$^3$] | 0.551 x 0.309 x 0.277 |
| Crystal color | Clear yellow |
| Crystal shape | Block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 46.608° |
| Reflections collected | 117078 |
| Data / Restraints / Parameters | 119117/ 10 / 388 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.6123 / 0.7449 |
| Goodness-of-fit on $F^2$ | 1.129 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0227 w$R_2$ = 0.0532 |
| Final $R$ indexes [all data] | $R_1$ = 0.0245 w$R_2$ = 0.0546 |
| Largest peak/hole [eÅ$^3$] | 1.413/ -0.985 |

Table 5 Crystal data for Gd(TEtDAPhen)$(NO_3)_3$. The ethyl arm on the ligand had a 60/40 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450524 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Gd$ |
| Formula weight | 721.75 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.5653(10) |
| $b$ [{dist_unit}] | 17.4897(19) |
| $c$ [{dist_unit}] | 15.9187(18) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.412(3)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2646.4(5) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.811 |
| $\mu$ [mm$^1$] | 1.377 |
| $F(000)$ | 1436 |
| Crystal size [mm$^3$] | 0.507 x 0.35 x 0.24 |
| Crystal color | Clear yellow |
| Crystal shape | Block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 43.939° |
| Reflections collected | 56149 |
| Data / Restraints / Parameters | 57154/ 12 / 388 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.6280/ 0.7447 |
| Goodness-of-fit on $F^2$ | 1.134 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0241 w$R_2$ = 0.0576 |
| Final $R$ indexes [all data] | $R_1$ = 0.0253 w$R_2$ = 0.0584 |
| Largest peak/hole [eÅ$^3$] | 1.197/ -0.859 |

Table 6 Crystal data for Dy(TEtDAPhen)$(NO_3)_3$. The ethyl arm on the ligand had a 70/30 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450520 |
| Empirical formula | $C_{21.987}H_{5.934}N_7O_{11}Dy$ |
| Formula weight | 726.78 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.5190(6) |
| $b$ [{dist_unit}] | 17.4566(11) |
| $c$ [{dist_unit}] | 15.9292(11) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.408(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2630.4(3) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.835 |
| $\mu$ [mm$^1$] | 1.558 |
| $F(000)$ | 1443 |
| Crystal size [mm$^3$] | 0.484 x 0.234 x 0.21 |
| Crystal color | Clear yellow |
| Crystal shape | Block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2Θ range [°] | 46.702° |
| Reflections collected | 72328 |
| Data / Restraints / Parameters | 73301/ 1 / 392 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.6168 / 0.7449 |
| Goodness-of-fit on $F^2$ | 1.117 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0225 w$R_2$ = 0.0531 |
| Final $R$ indexes [all data] | $R_1$ = 0.0250 w$R_2$ = 0.0551 |
| Largest peak/hole [eÅ$^3$] | 1.780/ -0.851 |

Table 7 Crystal data for Ho(TEtDAPhen)$(NO_3)_3$. The ethyl arm on the ligand had a 70/30 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450519 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Ho$ |
| Formula weight | 729.43 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.584(4) |
| $b$ [{dist_unit}] | 17.569(7) |
| $c$ [{dist_unit}] | 16.203(7) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.835(14)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2709.0(19) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.788 |
| $\mu$ [mm$^1$] | 1.601 |
| $F(000)$ | 1448 |
| Crystal size [mm$^3$] | 0.235 x 0.207 x 0.157 |
| Crystal color | Clear yellow |
| Crystal shape | prism |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 43.188° |
| Reflections collected | 87354 |
| Data / Restraints / Parameters | 89187/ 1 / 385 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.6175 / 0.7171 |
| Goodness-of-fit on $F^2$ | 1.082 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0302 w$R_2$ = 0.0730 |
| Final $R$ indexes [all data] | $R_1$ = 0.0344 w$R_2$ = 0.0775 |
| Largest peak/hole [eÅ$^3$] | 1.000/ -0.548 |

Table 8 Crystal data for Er(TEtDAPhen)$(NO_3)_3$. The ethyl arm on the ligand had a 70/30 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450521 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Er$ |
| Formula weight | 731.76 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.5494(10) |
| $b$ [{dist_unit}] | 17.5693(18) |
| $c$ [{dist_unit}] | 16.1606(15) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.922(3)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2691.6(5) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.806 |
| $\mu$ [mm$^1$] | 1.705 |
| $F(000)$ | 1452 |
| Crystal size [mm$^3$] | 0.153 x 0.147 x 0.13 |
| Crystal color | Clear yellow |
| Crystal shape | Prism |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 43.134° |
| Reflections collected | 49237 |
| Data / Restraints / Parameters | 50139/ 10 / 388 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.6279 / 0.7445 |
| Goodness-of-fit on $F^2$ | 1.036 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0293 w$R_2$ = 0.0722 |
| Final $R$ indexes [all data] | $R_1$ = 0.0350 w$R_2$ = 0.0772 |
| Largest peak/hole [eÅ$^3$] | 0.872/ -0.663 |

Table 9 Crystal data for Tm(TEtDAPhen)$(NO_3)_3$. The ethyl arm on the ligand had a 75/25 disorder changing the orientation of the arm. The SADI command was applied to constrain the bond lengths for the disorder. Then a SIMU command was used to ensure similar thermal parameters when modeling the disorder of the atoms on the ethyl arm.

| | |
|---|---|
| CCDC number | 2450522 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Tm$ |
| Formula weight | 733.43 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | 14 |
| $a$ [{dist_unit}] | 9.5417(4) |
| $b$ [{dist_unit}] | 17.5481(7) |
| $c$ [{dist_unit}] | 16.1542(7) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 96.875(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2685.39(19) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.814 |
| $\mu$ [mm$^1$] | 1.806 |
| $F(000)$ | 1456 |
| Crystal size [mm$^3$] | 0.204 x 0.19 x 0.155 |
| Crystal color | Clear yellow |
| Crystal shape | Block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 44.1° |
| Reflections collected | 62772 |
| Data / Restraints / Parameters | 63908 / 10 / 388 |
| Absorption correction $T_{min}/T_{max}$ (method) | Multi-scan 0.5994/ 0.6651 |
| Goodness-of-fit on $F^2$ | 1.058 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0369 w$R_2$ = 0.0936 |
| Final $R$ indexes [all data] | $R_1$ = 0.0428 w$R_2$ = 0.1009 |
| Largest peak/hole [eÅ$^3$] | 1.193/ -0.695 |

Table 10 Crystal data for Yb(TEtDAPhen)$(NO_3)_3$. Due to the smaller cation of the yterbium, one of the nitrates was monodentate. This nitrate moiety showed two different orientations depending on the sterics of the ethyl arms. Both nitrates were modeled individually. The ISOR command was applied to each nitrate to control the thermal parameters during refinements.

| | |
|---|---|
| CCDC number | 2450518 |
| Empirical formula | $C_{22}H_{26}N_7O_{11}Yb$ |
| Formula weight | 737.54 |
| Temperature [K] | 100 |
| Crystal system | Monoclinic, $P2_{1/c}$ |
| Space group (number) | $P2_{1/c}$, 14 |
| $a$ [{dist_unit}] | 9.8307(6) |
| $b$ [{dist_unit}] | 17.5745(9) |
| $c$ [{dist_unit}] | 15.1393(9) |
| $\alpha$ [°] | 90° |
| $\beta$ [°] | 99.408(2)° |
| $\gamma$ [°] | 90° |
| Volume [{vol_unit}$^3$] | 2580.4(3) |
| $Z$ | 4 |
| $\rho_{calc}$ [gcm$^3$] | 1.898 |
| $\mu$ [mm$^1$] | 1.984 |
| $F(000)$ | 1460 |
| Crystal size [mm$^3$] | 0.157 x 0.153 x 0.096 |
| Crystal color | Clear yellow |
| Crystal shape | Block |
| Radiation | Ag $K\alpha$ ($\lambda$=0.56086) |
| 2$\Theta$ range [°] | 40.702° |
| Reflections collected | 95263 |
| Data / Restraints / Parameters | 7949 / 91 / 431 |
| Absorption correction $T_{min}/T_{max}$ (method) | 0.6592 / 0.7449 multi-scan |
| Goodness-of-fit on $F^2$ | 1.026 |
| Final $R$ indexes [$I \geq 2\sigma(I)$] | $R_1$ = 0.0296 w$R_2$ = 0.0688 |
| Final $R$ indexes [all data] | $R_1$ = 0.0335 w$R_2$ = 0.0714 |
| Largest peak/hole [eÅ$^3$] | 2.106 / -1.591 |

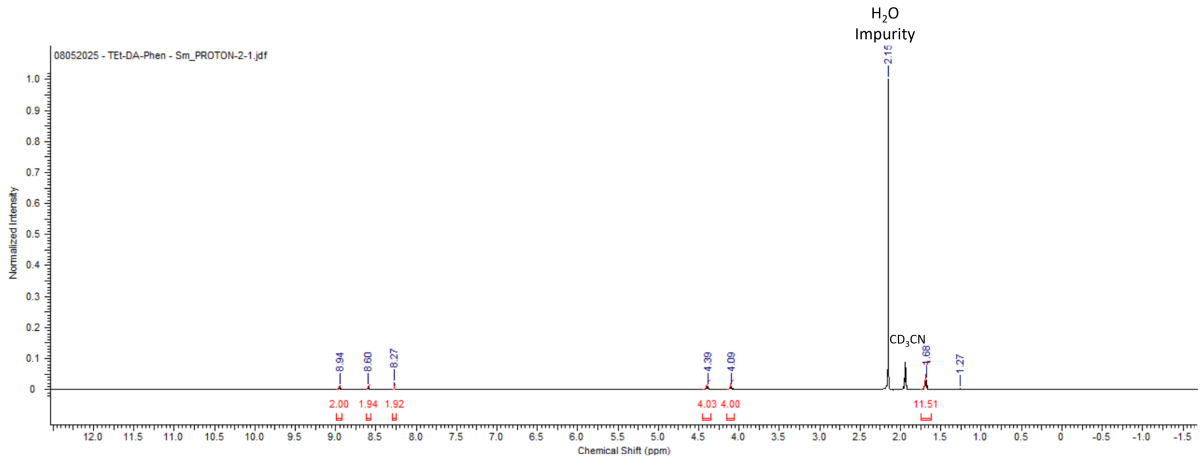


Fig. 3 $^{1}$H NMR of Sm(TEtDAPhen)$(NO_3)_3$ complex in $CD_3CN$.

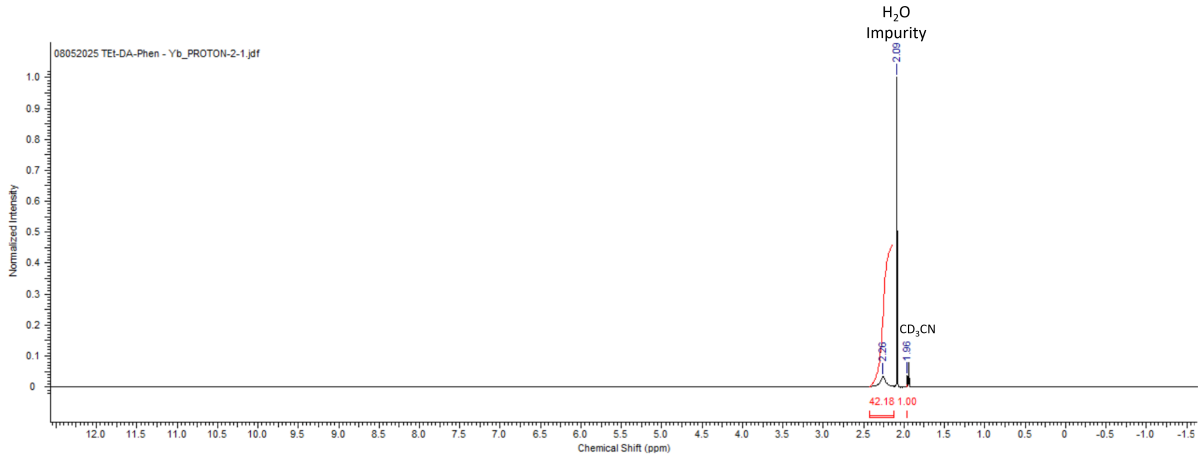


Fig. 4 $^{1}$H NMR of Yb(TEtDAPhen)$(NO_3)_3$ complex in $CD_3CN$.

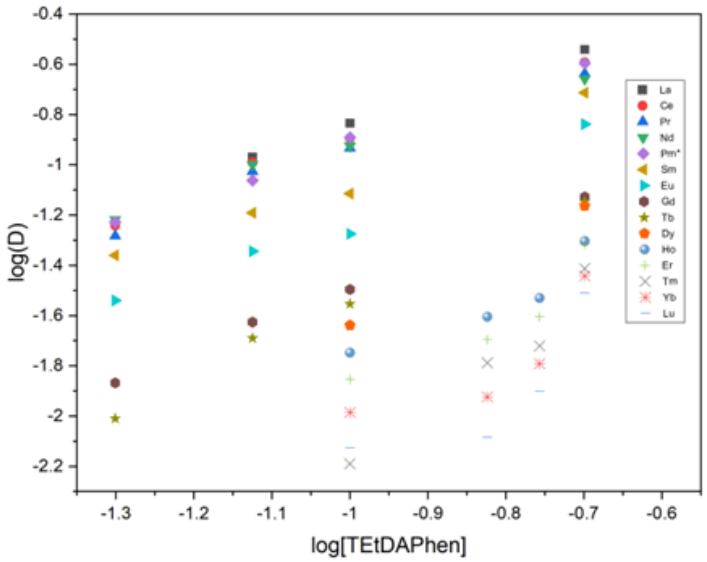


Fig. 5 The plotted logD values of each lanthanide with respect to the log[TEtDAPhen] for slope analysis.

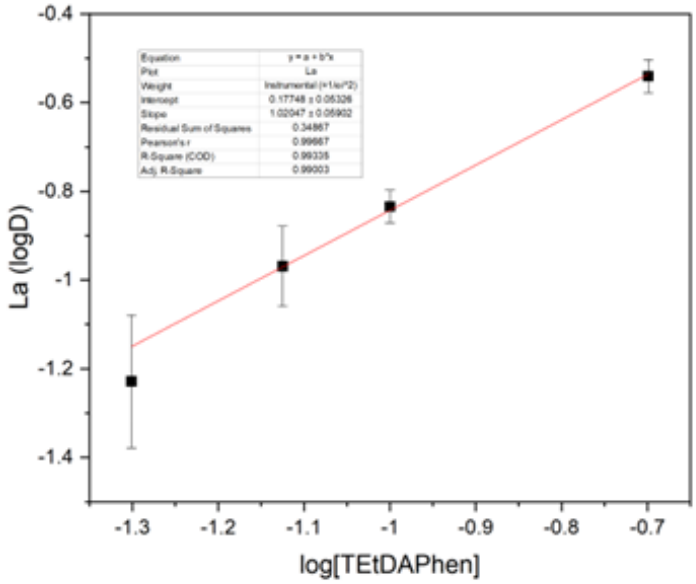


Fig. 6 La(III) slope analysis plot with a weighted linear fit. The slope value is 1.02 ± 0.06.

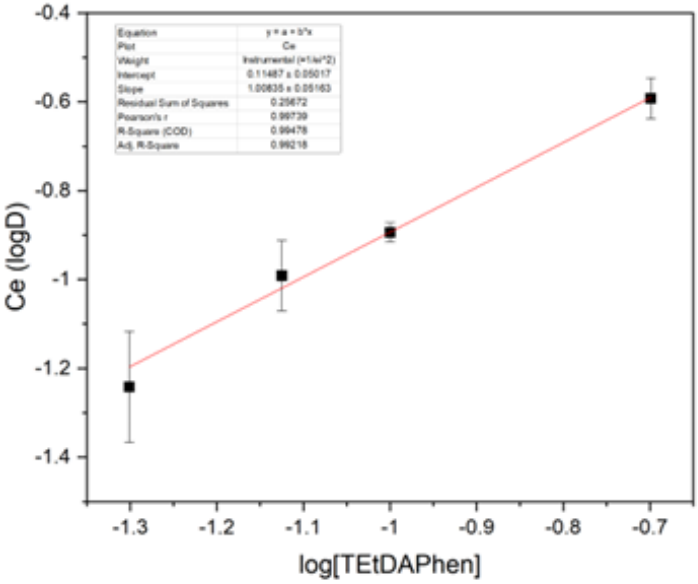


Fig. 7 Ce(III) slope analysis plot with a weighted linear fit. The slope value is 1.01 ± 0.05.

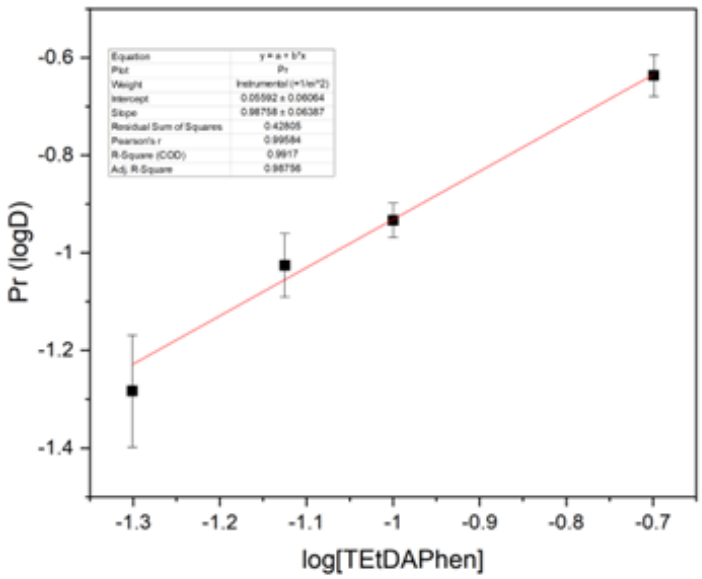


Fig. 8 Pr(III) slope analysis plot with a weighted linear fit. The slope value is 0.99 ± 0.06.

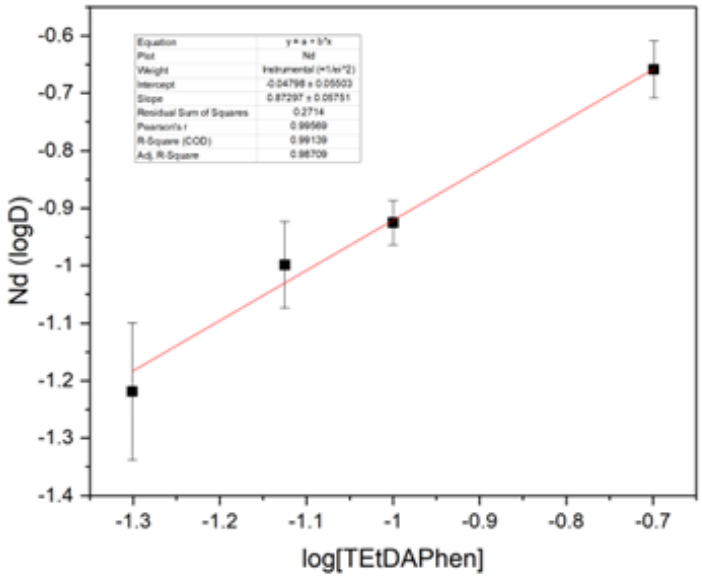


Fig. 9 Nd(III) slope analysis plot with a weighted linear fit. The slope value is 0.87 ± 0.06.

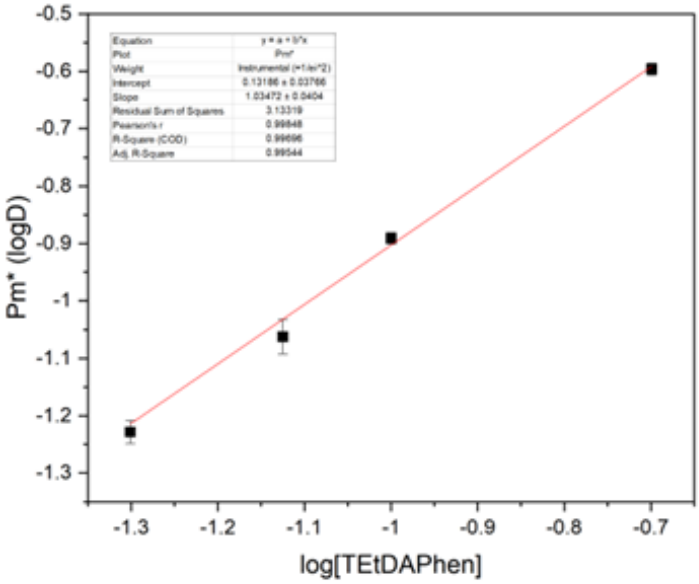


Fig. 10 Pm(III) slope analysis plot with a weighted linear fit. The slope value is 1.03 ± 0.04.

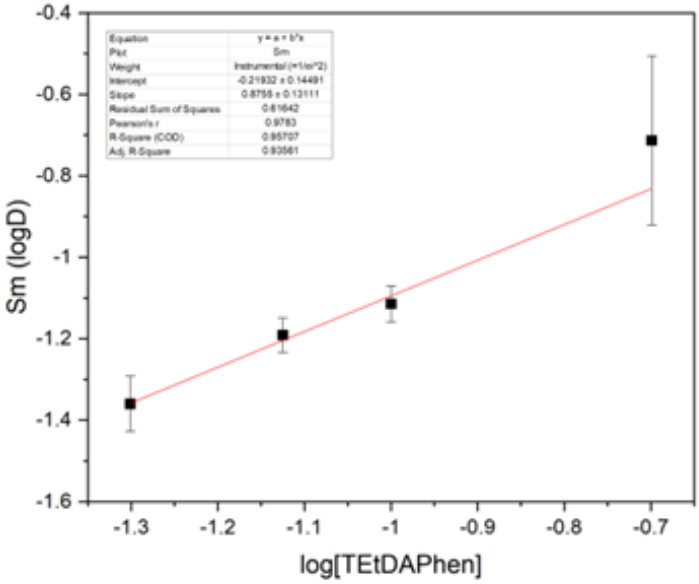


Fig. 11 Sm(III) slope analysis plot with a weighted linear fit. The slope value is 0.9 ± 0.1.

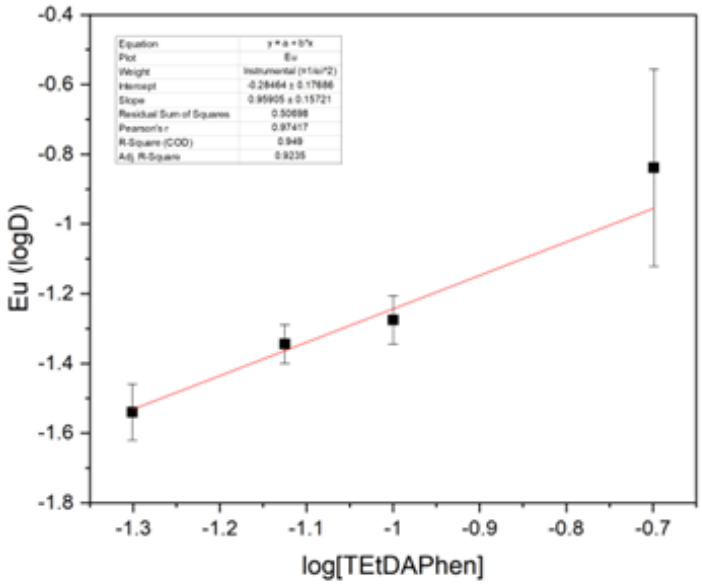


Fig. 12 Eu(III) slope analysis plot with a weighted linear fit. The slope value is 1.0 ± 0.2.

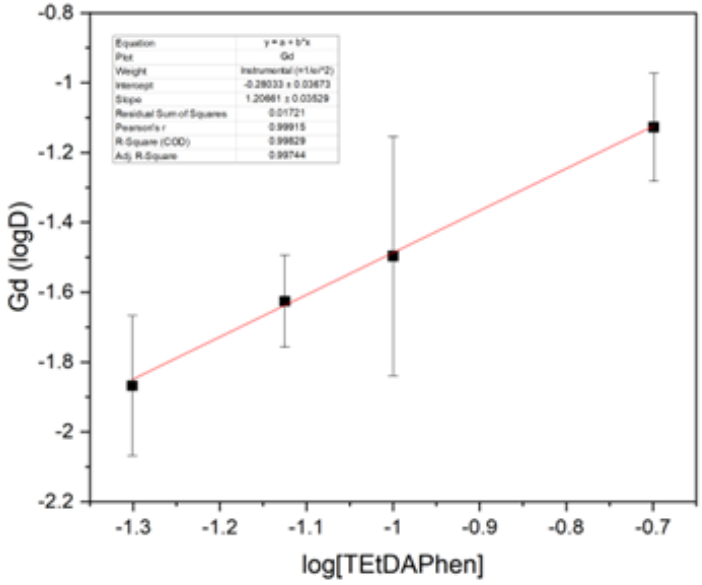


Fig. 13 Gd(III) slope analysis plot with a weighted linear fit. The slope value is 1.21 ± 0.04.

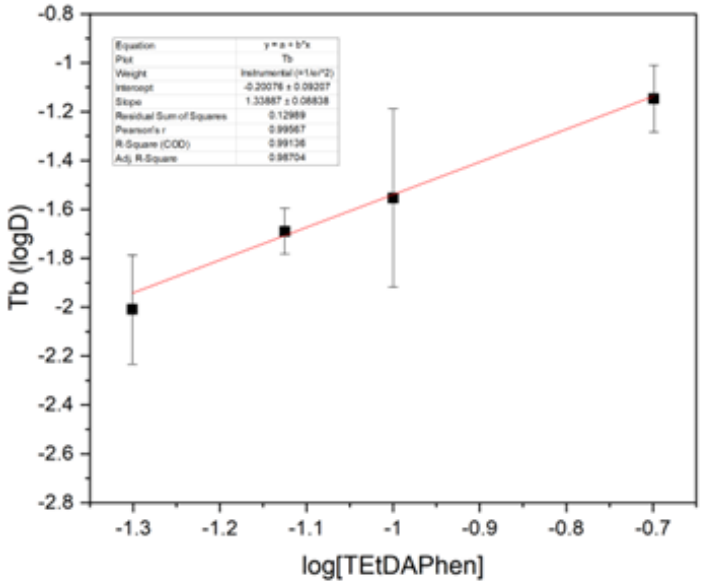


Fig. 14 Tb(III) slope analysis plot with a weighted linear fit. The slope value is 1.33 ± 0.09.

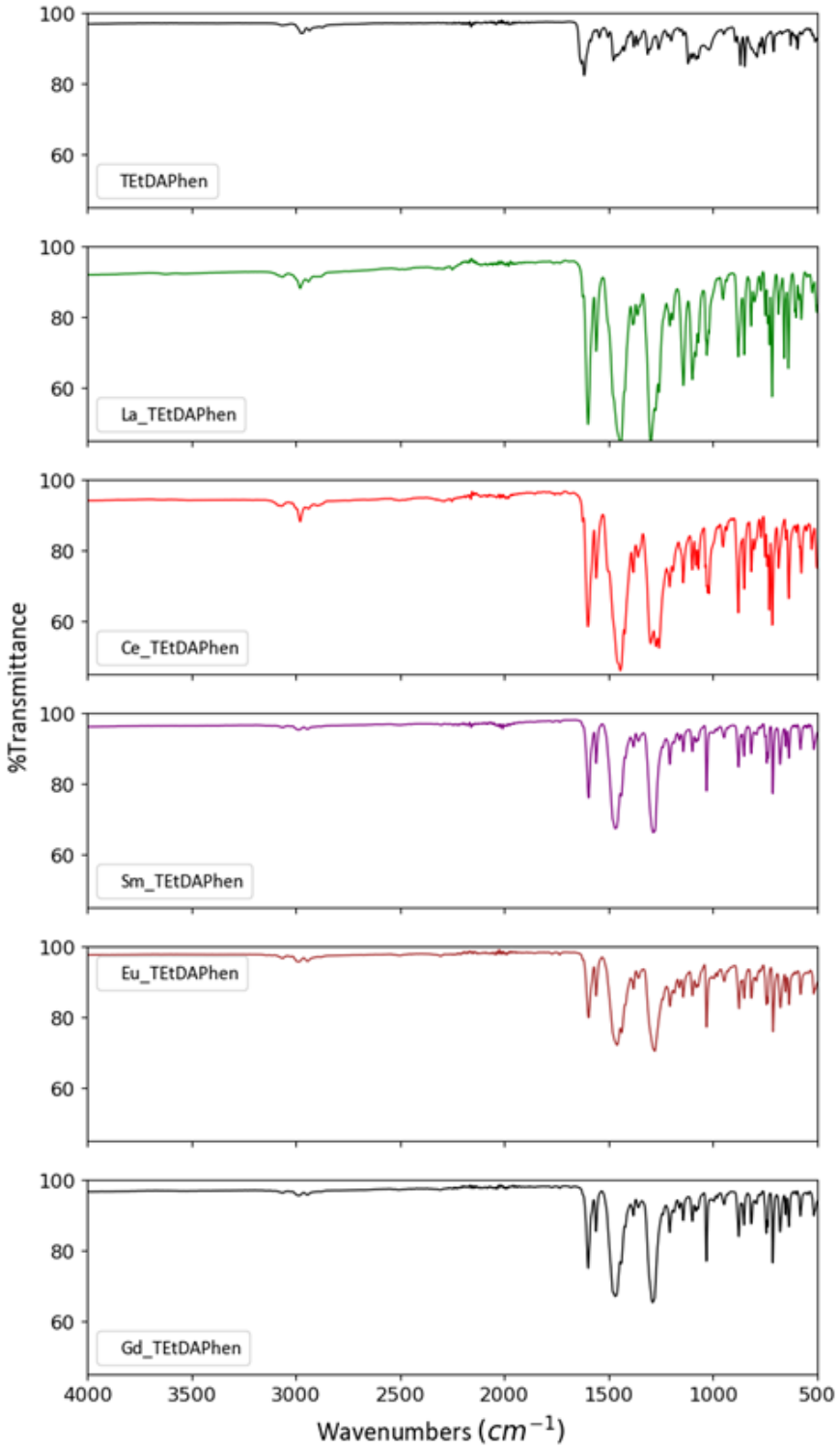


Fig. 15 FTIR spectra of the solid Ln(TEtDAPhen)$(NO_3)_3$ complexes where the Ln = La, Ce, Sm, Eu, Gd.

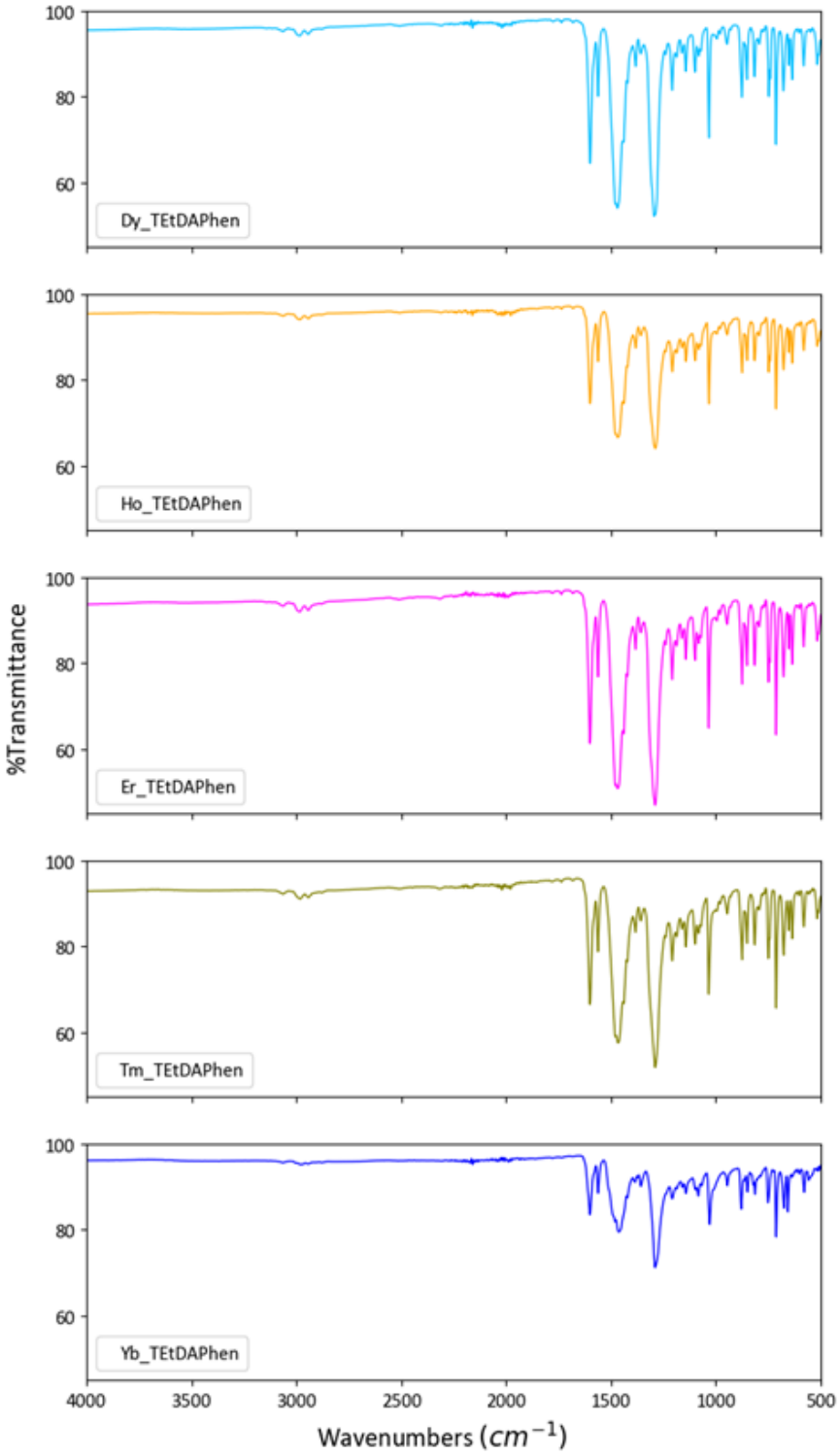


Fig. 16 FTIR spectra of the solid Ln(TEtDAPhen)$(NO_3)_3$ complexes where the Ln = Dy, Ho, Er, Tm, Yb.