Supporting Information

Norm-conserving pseudopotentials and basis sets to explore Lanthanide chemistry in complex environments

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Part A: SI for Section 2.1 "GTH pseudopotentials and MOLOPT basis sets"

GTH pseudopotentials contain local and non-local parts. The local part includes one or two coefficients. The non-local part has three scattering coefficients for *s* and *p* angular momentum, one scattering coefficient for *d* and *f* angular momentum. There is no standard way to optimize pseudopotential parameters. For this purpose, we minimized a user defined penalty function. In order to evaluate the penalty function for a given set of pseudopotential parameters, calculations on the radial Schrodinger equation of the pseudo-atom must be performed at each step of the fitting cycle. The penalty function consists of a weighted sum of square deviations to the reference data, which is derived from all-electron calculations of atom:

$$S = \sum_{n,\ l} w_{n,l,p}^2 (p_{n,l}^{AE} - p_{n,l}^{PS}) \qquad \text{(Eq. S1)}$$

Several important atomic properties were chosen for the penalty function:

(1) Atomic eigenvalue $\varepsilon_{n,\,l}$

(2) Charge density within a sphere of radius r_{loc}

(3) Nodeless of atomic orbitals.

The weights of each property were chosen carefully. In the present work, we gave the highest weight to the eigenvalue and the next highest weight to charge density. It is believed that the eigenvalues of valence orbitals are right if eigenvalues of lower core orbitals are correct. Therefore, we decreased the weight by one order of magnitude with increasing principle quantum number *n*.

After getting the pseudopotential parameters, we optimized the Gaussian exponents for the *s*, *p*, *d* and *f* orbitals. We tested the Gaussian exponents of *f* orbitals chosen from different existing basis set:

(1) *f*_crenbl_n: the Gaussian exponents of *f* orbitals are chosen from the Gaussian exponents of CRENBL ECP basis set, where *n* is the number of exponents of *f* orbitals.

(2) *f*_stg_n: the Gaussian exponents of *f* orbitals chosen from the Gaussian exponents of Stuttgart RSC ANO/ECP basis set. *n* is the number of exponents of *f* orbitals.

(3) *f*_spd_n: the Gaussian exponents of *f* orbitals are chosen from the Gaussian exponents of *s*, *p* and *d* states generated by ATOM code. *n* means the number of exponents of *f* orbitals.

The Gaussian exponents of *s*, *p* and *d* orbitals were optimized by the ATOM code in the CP2K package. The confinement parameters of GTH pseudopotentials are modulated to get different Gaussian exponents for the *s*, *p* and *d* orbitals.

The number of Gaussian exponents of *s*, *p* and *d* orbitals, the value of Gaussian exponents of *s*, *p* and *d* orbitals, and *f* orbitals are three key basis set factors. Tables S1 – S3 show the results of testing the three factors with Ce. Calculations are based on uncontracted basis sets. We tested the three key factors for all lanthanides. Here we take the testing of Ce as example:

For the testing of number of Gaussian exponents of *s*, *p* and *d* orbitals, the Gaussian exponents of *s*, *p*

and *d* orbitals are:

Gau_num = 6: 0.04091750, 0.12993444, 0.34483678, 0.77787686, 1.72233946, 3.96902350
Gau_num = 7: 0.04053284, 0.11467513, 0.30691395, 0.71562304, 1.51652533, 2.36809432, 4.15237224
Gau_num = 8: 0.04024110, 0.08851298, 0.19917888, 0.44370691, 0.93200530, 1.75066160, 2.51467732, 4.22060316

The Gaussian exponents of *f* orbitals which are grasped from CRENBL ECP basis set are: 6.9560000, 2.7930000, 1.0680000, 0.3499000.

For the testing of value of Gaussian exponents of *s*, *p* and *d* orbitals, we choose the following Gaussian exponents for *s*, *p* and *d* orbitals:

BS_1: 0.08775575, 0.16447973, 0.38040355, 0.83282172, 1.65665417, 2.58067085, 4.22245063
BS_2: 0.08104942, 0.14873630, 0.36148785, 0.80493102, 1.61437811, 2.51363675, 4.20565015
BS_3: 0.08019980, 0.12570707, 0.33070112, 0.75330363, 1.52928534, 2.37168605, 4.16775673
BS_4: 0.07073186, 0.13538834, 0.34409140, 0.77774428, 1.57616853, 2.45410483, 4.18880700
BS_5: 0.07055351, 0.11069368, 0.30792855, 0.71255837, 1.45299554, 2.25060217, 4.13501187
BS_6: 0.06073284, 0.12805253, 0.33396022, 0.76185311, 1.55595368, 2.42392346, 4.17935077
BS_7: 0.06091388, 0.10832212, 0.30262724, 0.70317879, 1.43381466, 2.22341926, 4.12774705
BS_8: 0.05648421, 0.09698038, 0.28029298, 0.65661696, 1.31944838, 2.08377973, 4.09031932
BS_9: 0.05097481, 0.12786786, 0.32763789, 0.75072837, 1.57226158, 2.47592859, 4.18237524
BS_10: 0.04053284, 0.11467513, 0.30691395, 0.71562304, 1.51652533, 2.36809432, 4.15237224
BS_11: 0.03787913, 0.10855637, 0.29403882, 0.69185398, 1.47593046, 2.28939079, 4.13019283
The *f* states are: 6.9560000, 2.7930000, 1.0680000, 0.3499000

For testing of value of Gaussian exponents for *f* states, we choose the following Gaussian exponents for *f* orbitals:

f_stg_4: 8.6013000, 3.8049000, 1.6176000, 0.6364000
f_stg_5: 19.4518000, 8.6013000, 3.8049000, 1.6176000, 0.6364000
f_stg_6: 43.9881000, 19.4518000, 8.6013000, 3.8049000, 1.6176000, 0.6364000
f_stg_7: 43.9881000, 19.4518000, 8.6013000, 3.8049000, 1.6176000, 0.6364000, 0.2164000
f_crenbl_4: 6.9560000, 2.7930000, 1.0680000, 0.3499000
f_crenbl_5: 18.2400000, 6.9560000, 2.7930000, 1.0680000, 0.3499000
f_spd_4: 0.65661696, 1.31944838, 2.08377973, 4.09031932
f_spd_5: 0.28029298, 0.65661696, 1.31944838, 2.08377973, 4.09031932
f_spd_6: 0.09698038, 0.28029298, 0.65661696, 1.31944838, 2.08377973, 4.09031932
f_spd_7: 0.05648421, 0.09698038, 0.28029298, 0.65661696, 1.31944838, 2.08377973, 4.09031932
The *s*, *p* and *d* states are: 0.05648421, 0.09698038, 0.28029298, 0.65661696, 1.31944838, 2.08377973, 4.09031932

Finally, the contract coefficients of *s*, *p* and *d* orbitals are optimized, and *f* orbitals are uncontracted. The training molecules of each lanthanide element consisted of lanthanide oxide, nitride, fluoride, hydride and chloride complexes. The lanthanide oxidation states in these complexes range from I to III, due to the fact that lanthanide chemistry is dominated by low oxidation states.

To double check our approach, we used the ATOM code in the CP2K package to verify that the same potentials can be generated.

Table S1. The number of Gaussian exponent of *s*, *p* and *d* orbitals tests for $CeCl_n$ (n = 1 - 3). The unit of energy is Hartree.

Gau_num	E(CeCl)	E($CeCl_2$)	E($CeCl_3$)	R1	R2
6	-53.66561	-68.77244	-83.87044	-0.13801	-0.12918
7	-53.66742	-68.77428	-83.87752	-0.13804	-0.13442
8	-53.66767	-68.77475	-83.87797	-0.13826	-0.13440
ADF_ref				-0.14564	-0.13469

Table S2. The *f* states tests for $CeCl_n$ (n = 1 - 3). The unit of energy is Hartree.

f state	E(CeCl)	E($CeCl_2$)	E($CeCl_3$)	R1	R2
*f*_ano_4	-53.71134	-68.81445	-83.90328	-0.13429	-0.12001
*f*_ano_5	-53.7097	-68.81458	-83.90334	-0.13606	-0.11994
*f*_ano_6	-53.70975	-68.81548	-83.90332	-0.13691	-0.11902
*f*_ano_7	-53.73602	-68.83488	-83.91793	-0.13004	-0.11423
*f*_crenbl_4	-53.66742	-68.77428	-83.87752	-0.13804	-0.13442
*f*_crenbl_5	-53.6875	-68.79152	-83.88961	-0.1352	-0.12927
*f*_spd_4	-53.70416	-68.81033	-83.90097	-0.13735	-0.12182
*f*_spd_5	-53.73329	-68.83375	-83.91185	-0.13164	-0.10928
*f*_spd_6	-53.73739	-68.83592	-83.91869	-0.12971	-0.11395
*f*_spd_7	-53.73754	-68.83606	-83.91276	-0.1297	-0.10788
ADF_ref				-0.14564	-0.13469

Table S3. The *s*, *p* and *d* states tests for $CeCl_n$ (n = 1 - 3). The unit of energy is Hartree.

spd state	E(CeCl)	E($CeCl_2$)	E($CeCl_3$)	R1	R2
BS_1	-53.64651	-68.76884	-83.87752	-0.15351	-0.13986
BS_2	-53.65155	-68.77003	-83.8774	-0.14966	-0.13855
BS_3	-53.65447	-68.77074	-83.87742	-0.14745	-0.13786
BS_4	-53.6574	-68.77155	-83.87749	-0.14533	-0.13712
BS_5	-53.65942	-68.77206	-83.87743	-0.14382	-0.13655
BS_6	-53.6618	-68.77274	-83.8775	-0.14212	-0.13594
BS_7	-53.66253	-68.77291	-83.87745	-0.14156	-0.13572
BS_8	-53.66412	-68.77335	-83.87743	-0.14041	-0.13526
BS_9	-53.66511	-68.77371	-83.87761	-0.13978	-0.13508
BS_10	-53.66721	-68.77422	-83.87744	-0.13819	-0.1344
BS_11	-53.66742	-68.77428	-83.87752	-0.13804	-0.13442
ADF_ref				-0.14564	-0.13469

Part B: SI for Section 2.2 "Computational methodology"

Table S4. The electronic configuration of LnCl, $LnCl_2$ and $LnCl_3$.

Complex	LnCl	$LnCl_2$	$LnCl_3$
La	$5d^2$	$5d^1$	$5s^25p^6$
Ce	$4f^15d^2$	$4f^16s^1$	$4f^1$
Pr	$4f^36s^1$	$4f^3$	$4f^2$
Nd	$4f^46s^1$	$4f^4$	$4f^3$
Pm	$4f^56s^1$	$4f^5$	$4f^4$
Sm	$4f^66s^1$	$4f^6$	$4f^5$
Eu	$4f^76s^1$	$4f^7$	$4f^6$
Gd	$4f^76s^2$	$4f^76s^1$	$4f^7$
Tb	$4f^96s^1$	$4f^9$	$4f^8$
Dy	$4f^{10}6s^1$	$4f^{10}$	$4f^9$
Ho	$4f^{11}6s^1$	$4f^{11}$	$4f^{10}$
Er	$4f^{12}6s^1$	$4f^{12}$	$4f^{11}$
Tm	$4f^{13}6s^1$	$4f^{13}$	$4f^{12}$
Yb	$4f^{14}6s^1$	$4f^{14}$	$4f^{13}$
Lu	$4f^{14}6s^2$	$4f^{14}6s^1$	$4f^{14}$

Discussion on ADF calculations

It is difficult to determine the occupation of *f* orbitals in lanthanide complexes using ADF. Here we calculate the energetics of the two redox reactions (R1 and R2, see main text) by three schemes:

(1) Scheme 1: Defining the occupation pattern. As the DFT wavefunction is a single-reference slater determinant, we must find an occupation pattern for *f* orbital electronic configuration. Here our standard is that M_z is largest.
(2) Scheme 2: Fully fragment occupation for *f* orbitals. As energy levels of *f* orbitals are very close, all *f* orbitals can occupy electrons. We divided electrons into every *f* orbitals in equal footing, *e. g.*, the occupation number is 1/7 if we have the electronic configuration f^1.
(3) Scheme 3: Dirac-Fermi smearing. Based on scheme 2, Dirac-Fermi smearing made electrons occupy different *f* orbitals fragmentally, but not with a fully fragmented occupation. Here, the smearing value is 0.01 Hartree.

The occupation of *f* orbitals in most solid state packages follows scheme 3. In order to check scheme 3's feasibility, we tested it for 3*d*-transition metal. As shown in Tables S5 and S6, the agreement between our new GTH pseudopotential results and ADF all-electron results are satisfactory. The error is relatively small.

Table S5. The energetics (kcal/mol) of two redox reactions by ADF through three schemes.

Element	Scheme_1		Scheme_2		Scheme_3	
	R1	R2	R1	R2	R1	R2
La	-91.0	-90.2	-94.2	-106.6	-94.1	-90.2
Ce	-90.5	-85.6	-89.9	-86.9	-91.4	-84.5
Pr	-79.8	-88.5	-78.7	-87.1	-86.8	-76.6
Nd	-79.4	-68.0	-80.1	-70.0	-79.7	-69.8
Pm	-82.4	-62.3	-79.6	-54.2	-79.8	-63.4
Sm	-72.3	-52.4	-76.9	-41.5	-77.4	-48.3
Eu	-75.0	-26.8	-75.0	-28.5	-75.0	-33.3
Gd	-78.7	-83.8	-78.7	-83.8	-93.5	-83.8
Tb	-83.1	-81.6	-83.1	-89.1	-70.1	-82.6
Dy	-73.1	-64.1	-74.9	-73.3	-74.2	-66.1
Ho	-75.1	-69.1	-74.9	-63.2	-73.0	-65.0
Er	-68.8	-52.2	-70.2	-58.1	-70.1	-66.0
Tm	-74.8	-42.5	-74.1	-44.2	-72.3	-52.1
Yb	-73.8	-35.0	-73.8	-34.7	-73.8	-39.5
Lu	-72.5	-85.2	-72.5	-85.2	-72.5	-85.2

Table S6. The reaction energies (kcal/mol) of MCl_2 + ½ Cl2 = MCl_3 (M = Sc-Cu) by CP2K and ADF at PBE level.

Element	CP2K	ADF	Error[a]
Sc	-88.1	-87.3	-0.8
Ti	-79.1	-80.0	0.9
V	-58.2	-59.5	1.3
Cr	-50.5	-50.7	0.2
Mn	-20.5	-20.5	0.0
Fe	-36.6	-36.1	-0.5
Co	-31.1	-30.0	-1.1
Ni	-12.5	-11.5	-0.9
Cu	-1.5	-2.9	1.4
Zn	15.8	17.8	-2.0
MAD			**0.9 kcal/mol**

[a] Error is calculated by E(CP2K) – E(ADF)

Table S7. The bond energies (kcal/mol) of M-Cl in MCl_3 (M = Sc-Cu) by CP2K and ADF at PBE level.

Element	CP2K	ADF	Error[a]
Sc	-122.1	-118.9	-3.2
Ti	-114.8	-109.9	-4.9
V	-94.3	-89.0	-5.3
Cr	-85.5	-81.3	-4.1
Mn	-55.3	-51.4	-4.0
Fe	-70.9	-67.4	-3.4
Co	-64.8	-61.9	-2.9
Ni	-46.3	-43.3	-3.1
Cu	-37.7	-32.3	-5.4
Zn	-17.0	-15.0	-2.0
MAD			**3.8 kcal/mol**

[a] Error is calculated by E(CP2K) – E(ADF)

Table S8. The cutoff (Ry) tests of density in CP2K for $CeCl_n$ (n = 1 - 3). The unit of energy is Hartree.

cutoff	E(CeCl)	E($CeCl_2$)	E($CeCl_3$)	R1	R2
100	-53.72723	-68.83262	-83.92163	-0.13767	-0.12129
200	-53.66146	-68.76988	-83.87110	-0.14070	-0.13350
300	-53.65818	-68.76592	-83.86964	-0.14002	-0.13600
400	-53.65741	-68.76613	-83.86966	-0.14100	-0.13581
500	-53.65726	-68.76531	-83.86926	-0.14033	-0.13623
600	-53.65696	-68.76587	-83.86962	-0.14119	-0.13603
700	-53.65702	-68.76588	-83.86957	-0.14114	-0.13597
800	-53.65690	-68.76572	-83.86954	-0.14110	-0.13610
900	-53.65687	-68.76578	-83.86945	-0.14119	-0.13595
1000	-53.65687	-68.76582	-83.86959	-0.14123	-0.13605

Table S9. The cutoff (Ry) tests of density in CP2K for $TbCl_n$ (n = 1 – 3). The unit of energy is Hartree.

cut-off	E(TbCl)	E($TbCl_2$)	E($TbCl_3$)	R1	R2
100	-429.21729	-445.46461	-459.97638	-1.27960	0.45595
200	-425.11815	-440.33827	-455.34466	-0.25240	-0.03867
300	-425.003	-440.08789	-455.17276	-0.11717	-0.11715
400	-424.95845	-440.04016	-455.13251	-0.11399	-0.12463
500	-424.9385	-440.01561	-455.11595	-0.10939	-0.13262
600	-424.92801	-440.00461	-455.10617	-0.10888	-0.13384
700	-424.92576	-440.00214	-455.10312	-0.10866	-0.13326
800	-424.92401	-440.00071	-455.10162	-0.10898	-0.13319
900	-424.92372	-440.00034	-455.10128	-0.10890	-0.13322
1000	-424.92383	-440.00077	-455.10133	-0.10922	-0.13284

Part C: SI for Section 3.1 "Pseudopotential transferability and reaction energies"

Table S10. The r_{loc} in medium-core pseudopotential tests for Tb. The unit energy is Hartree.

r_{loc}	E(TbCl)	E($TbCl_2$)	E($TbCl_3$)	E(R1)	E(R2)
0.48	-425.37674	-440.45817	-455.56388	-0.11261	-0.13689
0.49	-424.84606	-439.92731	-455.0342	-0.11243	-0.13807
0.5	-424.94475	-440.02427	-455.12763	-0.11070	-0.13454
0.51	-424.55303	-439.63273	-454.73568	-0.11088	-0.13413
ADF_ref				-0.11173	-0.13161

Table S11. The redox reaction energy (kcal/mol) by using CP2K optimized structure and Gaussian optimized structure in CP2K.

Element	CP2K optimized structure		Gaussian optimized structure	
	Reac_1	Reac_2	Reac_1	Reac_2
La	-89.9	-88.5	-89.7	-88.6
Ce	-88.4	-84.9	-88.5	-85.4
Pr	-86.2	-73.3	-85.7	-73.8
Nd	-80.5	-59.3	-77.6	-62.3
Pm	-79.7	-61.9	-80.6	-61.9
Sm	-78.8	-48.5	-78.9	-48.3
Eu	-75.9	-32.3	-75.9	-32.3
Gd	-90.0	-81.3	-89.3	-81.3
Tb	-74.2	-77.7	-68.4	-83.6
Dy	-76.9	-61.3	-76.4	-60.9
Ho	-71.8	-65.9	-71.7	-65.3
Er	-70.8	-66.0	-67.6	-68.6
Tm	-74.3	-50.4	-74.0	-50.1
Yb	-71.6	-41.5	-71.6	-41.0
Lu	-67.8	-87.5	-67.7	-87.2

Part D: SI for Section 3.2 "Molecular tests"

Table S12. Ln-Cl distance (Å) in $LnCl_n$ (Ln = La – Lu, n = 1 – 3) optimized by ADF and CP2K.

Element	Molecule	ADF/PBE	CP2K/PBE	Error[a]
La	LaCl	2.52	2.55	0.03
	$LaCl_2$	2.55	2.55	0.01
	$LaCl_3$	2.62	2.59	-0.03
Ce	CeCl	2.54	2.56	0.02
	$CeCl_2$	2.54	2.58	0.04
	$CeCl_3$	2.56	2.56	0.00
Pr	PrCl	2.55	2.60	0.05
	$PrCl_2$	2.54	2.59	0.05
	$PrCl_3$	2.54	2.54	0.00
Nd	NdCl	2.55	2.59	0.03
	$NdCl_2$	2.54	2.59	0.04
	$NdCl_3$	2.52	2.53	0.01
Pm	PmCl	2.56	2.58	0.03
	$PmCl_2$	2.56	2.58	0.02
	$PmCl_3$	2.51	2.52	0.01
Sm	SmCl	2.55	2.59	0.04
	$SmCl_2$	2.56	2.58	0.02
	$SmCl_3$	2.51	2.52	0.01
Eu	EuCl	2.54	2.58	0.04
	$EuCl_2$	2.55	2.58	0.03
	$EuCl_3$	2.53	2.56	0.03
Gd	GdCl	2.44	2.49	0.04
	$GdCl_2$	2.45	2.47	0.02
	$GdCl_3$	2.47	2.48	0.01
Tb	TbCl	2.45	2.47	0.02
	$TbCl_2$	2.47	2.51	0.05
	$TbCl_3$	2.46	2.49	0.03
Dy	DyCl	2.45	2.54	0.09
	$DyCl_2$	2.47	2.54	0.08
	$DyCl_3$	2.45	2.51	0.06
Ho	HoCl	2.46	2.51	0.05
	$HoCl_2$	2.47	2.51	0.04
	$HoCl_3$	2.44	2.48	0.04
Er	ErCl	2.46	2.50	0.05
	$ErCl_2$	2.45	2.52	0.07
	$ErCl_3$	2.43	2.48	0.05
	TmCl	2.46	2.56	0.10

Tm	$TmCl_2$	2.46	2.53	0.07
	$TmCl_3$	2.43	2.51	0.07
Yb	YbCl	2.45	2.53	0.07
	$YbCl_2$	2.45	2.50	0.05
	$YbCl_3$	2.44	2.50	0.06
Lu	LuCl	2.37	2.41	0.05
	$LuCl_2$	2.38	2.46	0.08
	$LuCl_3$	2.39	2.43	0.04
MAD				**0.04 Å**

[a] Error of bond length is calculated by $d_{CP2K} - d_{ADF}$.

Table S13. Cl-Ln-Cl angle (°) in $LnCl_n$ (Ln = La – Lu, n = 2, 3) optimized by ADF and CP2K.

Element	Molecule	ADF/PBE	CP2K/PBE	Error[a]
La	$LaCl_2$	115	113	-2
	$LaCl_3$	120	120	0
Ce	$CeCl_2$	115	117	2
	$CeCl_3$	120	120	0
Pr	$PrCl_2$	114	119	5
	$PrCl_3$	120	120	0
Nd	$NdCl_2$	115	120	5
	$NdCl_3$	120	121	1
Pm	$PmCl_2$	120	120	0
	$PmCl_3$	120	120	0
Sm	$SmCl_2$	121	122	1
	$SmCl_3$	120	120	0
Eu	$EuCl_2$	120	123	3
	$EuCl_3$	120	120	0
Gd	$GdCl_2$	119	120	1
	$GdCl_3$	118	116	-2
Tb	$TbCl_2$	119	126	7
	$TbCl_3$	120	120	0
Dy	$DyCl_2$	118	129	11
	$DyCl_3$	120	120	0
Ho	$HoCl_2$	121	123	2
	$HoCl_3$	120	120	0
Er	$ErCl_2$	119	132	13
	$ErCl_3$	120	120	0
Tm	$TmCl_2$	123	124	1
	$TmCl_3$	120	120	0
Yb	$YbCl_2$	123	125	2
	$YbCl_3$	120	120	0

Lu	$LuCl_2$	123	180	57
	$LuCl_3$	120	120	0
MAD				**4°**

[a] Error of angle is calculated by $a_{CP2K} - a_{ADF}$.

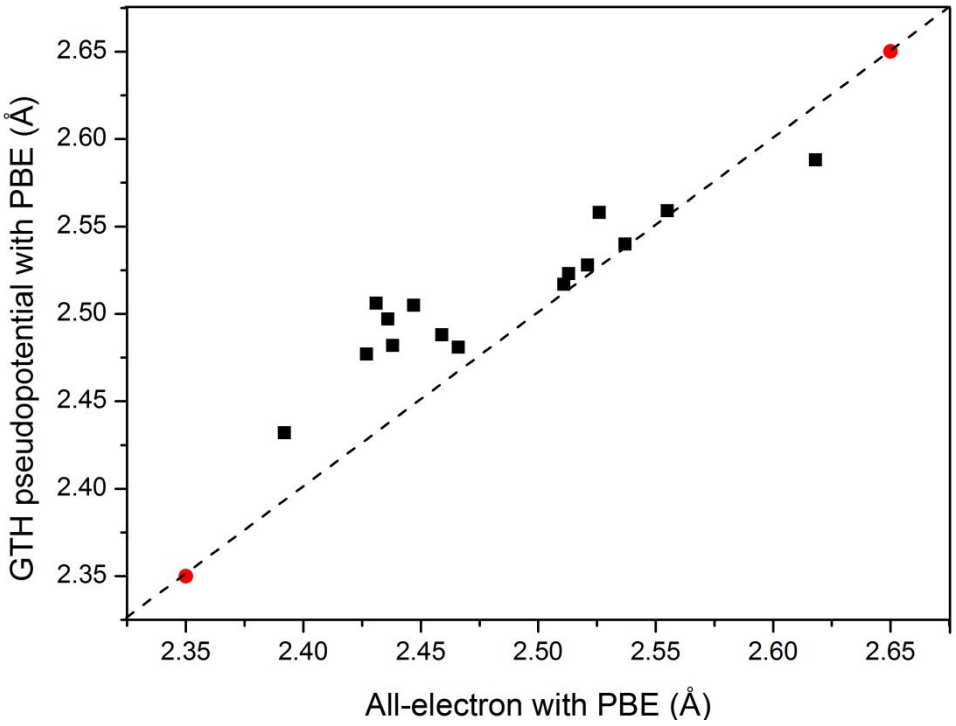

Figure S1. Comparison of the bond length for Ln-Cl (Ln = La - Lu) calculated with CP2K using the GTH pseudopotentials optimized for PBE and the bond length for Ln-Cl (Ln = La - Lu) obtained by ADF all-electron calculations with the TZ2P basis set.

Discussion on the calculation of enthalpies of formation with CP2K

Enthalpies of formation of $LnCl_n$ (n = 2, 3), LnF_n (n=1, 2, 3), and LnO were calculated using our LnPP1 GTH pseudopotentials and basis sets with uncontracted *f* states in the gas-phase under periodic conditions, based on varying the optimized structures and comparing with experiment. First, each structure was optimized in CP2K, yielded the "formed" structure (see Figure S1 below). Then, keeping the optimized angles, the optimized Ln-Cl, Ln-F, or Ln-O bond lengths were increased by a factor of *x*, to give the "non-formed" structure.

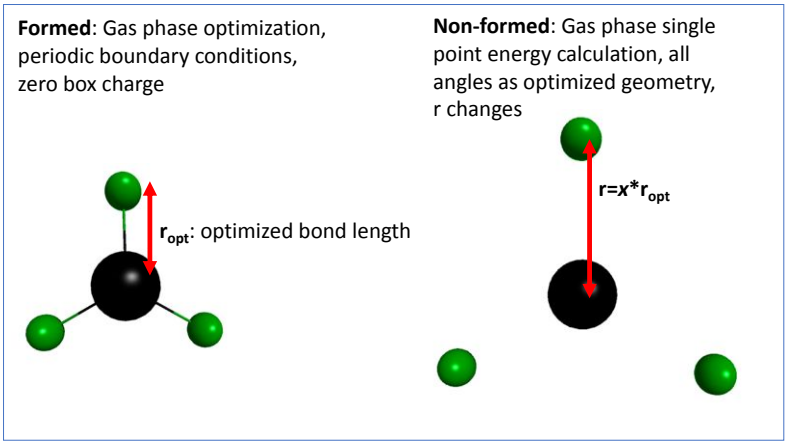

Figure S2. Structure of formed and non-formed structures, example with $CeCl_3$, with Ce^{3+} (black) and Cl^- (green).

The enthalpy of formation is simply calculated as:

$$H_f \cong E_{formed} - E_{non-formed} \qquad \text{(Eq. S2)}$$

There is one empirical parameter, $\boldsymbol{x}$, which is varied to match experiment. The fitting is performed in one early lanthanide, a middle lanthanide, and a late lanthanide, while retaining predictive values for the rest of the series. Tables S14 – S19 include all the calculated enthalpies of formation, with reported optimized $\boldsymbol{r_{opt}}$ values, as well as $\boldsymbol{x}$ values.

Although our procedure is not based on pure gas-phase reference molecules, it is a simple and fast procedure to calculate enthalpies of formation at acceptable with accuracy with respect to experiment. Calculations based on pure gas-phase reference molecules would include charged boxes (*e.g.*, Ln^{3+}, Cl^-). CP2K is not as well suited as quantum chemistry codes to perform calculations with charged boxes. Our approach avoids the use of charged boxes. Also, the Cl, F, and O MOLOPT basis sets are parametrized for short-range interactions.

Table S14. $LnCl_3$ enthalpies of formation (kcal/mol). Our calculations and the published[a] all-electron calculations both used the PBE functional.

	Ln-Cl average optimized bond length (Å)	$\boldsymbol{x}$	$\boldsymbol{H_f}$ with our pseudopotentials and basis sets	$\boldsymbol{H_f}$ with all-electron [a]	$\boldsymbol{H_f}$ from experiment[b]	Error between our calculations and experiment	Error between all-electron and experiment
La	2.59	1.50	-193.2	-175.4	-177.0	-16.2	1.6
Ce	2.55	1.50	-169.8	-178.2	-171.4	1.6	-6.8
Pr	2.53	1.50	-173.9	-155.4	-174.5	0.6	19.1
Nd	2.51	1.50	-158.9	-152.4	-172.4	13.5	19.7
Gd	2.48	1.45	-172.2	-152.7	-164.9	-7.3	-0.6
MAD						**7.8 kcal/mol**	**9.6 kcal/mol**

[a] S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

[b] Experimental data from various sources compiled in S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

Table S15. $LnCl_2$ enthalpies of formation (kcal/mol). Our calculations and the published[a] all-electron calculations both used the PBE functional.

	Ln-Cl average optimized bond length (Å)	x	H_f with our pseudopotentials and basis sets	H_f with all-electron[a]	H_f from experiment[b]	Error between our calculations and experiment	Error between all-electron and experiment
La	2.55	1.33	-75.2	-84.3	-85.0	9.8	0.7
Ce	2.52	1.33	-69.9	-93.1	-70.0	0.1	-23.1
Pr	2.51	1.33	-74.7	-83.8	-82.0	7.3	-1.8
Nd	2.54	1.33	-66.2	-91.4	-85.0	18.8	-6.4
Sm	2.53	1.33	-34.7	-104.4	-119.6	84.9	15.2
Eu	2.52	1.33	-66.7	-107.0	-109.1	42.4	2.1
Gd	2.44	1.33	-77.0	-81.1	-73.0	-4.0	-8.1
Tb	2.51	1.33	-73.8	-55.3	-73.0	-0.8	17.7
Ho	2.50	1.33	-78.0	-81.8	-85.0	7.0	3.2
Er	2.52	1.33	-72.7	-85.7	-84.0	11.3	-1.7
MAD						**18.6 kcal/mol**	**8.0 kcal/mol**

[a] S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

[b] Experimental data from various sources compiled in S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

Table S16. LnF_3 enthalpies of formation (kcal/mol). Our calculations and the published[a] all-electron calculations both used the PBE functional.

	Ln-F average optimized bond length (Å)	X	H_f with our pseudopotentials and basis sets	H_f with all-electron[a]	H_f from experiment[b]	Error between our calculations and experiment	Error between all-electron and experiment
La	2.12	2.00	-344.0	-309.0	-306.9	-37.1	-2.1
Ce	2.10	2.00	-313.5	-311.3	-315.1	1.6	3.8
Pr	2.09	2.00	-315.5	-287.5	-409.2	93.7	121.7
Nd	2.08	2.00	-300.9	-287.2	-315.3	14.4	28.1
Gd	2.03	1.75	-297.7	-295.9	-299.9	2.2	4.0
Tb	2.07	1.75	-293.6	-264.7	-298.7	5.1	34.0
Dy	2.07	2.30	-316.5	-255.6	-296.6	-19.9	41.0
Ho	2.05	2.30	-330.1	-251.5	-300.5	-29.6	49.0
Er	2.04	2.30	-292.5	-254.0	-301.2	8.7	47.2
Lu	2.02	2.30	-367.1	-285.2	-298.7	-68.4	13.5
MAD						**28.1 kcal/mol**	**34.4 kcal/mol**

[a] S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

[b] Experimental data from various sources compiled in S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

Table S17. LnF_2 enthalpies of formation (kcal/mol). Our calculations and the published[a] all-electron calculations both used the PBE functional.

	Ln-F average optimized bond length (Å)	***X***	H_f with our pseudopotentials and basis sets	H_f with all-electron[a]	H_f from experiment[b]	Error between our calculations and experiment	Error between all-electron and experiment
La	2.09	1.55	-149.0	-176.7	-147.0	-2.0	-29.7
Sm	2.05	1.80	-179.8	-185.0	-182.0	2.2	-3.0
Eu	2.04	1.80	-180.4	-184.0	-187.0	6.6	3.0
MAD						**3.6 kcal/mol**	**11.9 kcal/mol**

[a] S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

[b] Experimental data from various sources compiled in S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

Table S18. LnF enthalpies of formation (kcal/mol). Our calculations and the published[a] all-electron calculations both used the PBE functional.

	Ln-F average optimized bond length (Å)	***x***	H_f with our pseudopotentials and basis sets	H_f with all-electron[a]	H_f from experiment[b]	Error between our calculations and experiment	Error between all-electron and experiment
La	2.05	1.20	-21.5	-39.0	-21.0	-0.5	-18.0
Ho	2.05	1.85	-80.0	-40.1	-40.0	-40.0	-0.1
Yb	2.03	1.85	-75.8	-80.1	-80.0	4.2	-0.1
Lu	1.96	1.85	-104.3	-14.5	-14.0	-90.3	-0.5
MAD						**33.8 kcal/mol**	**4.7 kcal/mol**

[a] S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

[b] Experimental data from various sources compiled in S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

Table S19. LnO enthalpies of formation (kcal/mol). Our calculations and the published[a] all-electron calculations both used the PBE functional.

	Ln-O average optimized bond length (Å)	x	H_f with our pseudopotentials and basis sets	H_f with all-electron[a]	H_f from experiment[b]	Error between our calculations and experiment	Error between all-electron and experiment
La	1.87	1.18	-30.0	-39	-28.4	-1.6	-10.6
Ce	1.84	1.18	-29.3	-47.7	-31.5	2.2	-16.2
Pr	1.83	1.18	-30.4	-21.9	-34.7	4.3	12.8
Nd	1.82	1.18	-31.4	-17.2	-28.7	-2.7	11.5
Sm	1.81	1.10	-11.0	-19.3	-25.1	14.1	5.8
Eu	1.83	1.10	-11.1	-23.6	-13.5	2.4	-10.1
Gd	1.85	1.10	-11.5	-26.4	-16.3	4.8	-10.1
Tb	1.88	1.10	-23.8	46.5	-20.2	-3.6	66.7
Dy	1.90	1.10	-17.3	88.7	-17	-0.3	105.7
Ho	1.88	1.10	-15.3	27.6	-13.8	-1.5	41.4
Er	1.92	1.10	-22.9	17.4	-7.9	-15.0	25.3
Yb	1.92	1.10	-16.8	-4.9	-3.3	-13.5	-1.6
Lu	1.84	1.10	-11.6	-8.1	-3.8	-7.8	-4.3
MAD						**6.1 kcal/mol**	**23.0 kcal/mol**

[a] S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

[b] Experimental data from various sources compiled in S. Grimmel, G. Schoendorff, and Angela K. Wilson (2016), J. Chem. Theory Comput., 12, 1259−1266.

Part E: SI for Section 3.4 "DFT +U correction"

Table S20. The energies (Hartree) of Ce^{2+} and Ce^{3+} and the third ionization potential (IP) (kcal/mol) for Ce in different U (eV) value.

U	$E(Ce^{2+})$	$E(Ce^{3+})$	IP
0.05	-37.94316	-37.16058	491.1
0.1	-37.90733	-37.14571	477.9
0.15	-37.87412	-37.13098	466.3
0.2	-37.82359	-37.11635	443.8
Expt.			465.8

Table S21. The DFT and DFT+U results (kcal/mol) of fourth ionization potential for Ce and Tb by using the U value (eV) determined by the third ionization potential.

Element	U value	Exp.	DFT	DFT+U	Error(DFT)	Error(DFT+U)
Ce	4.08	848.0	937.2	909.1	89.2	61.1
Tb	2.45	908.3	1012.4	922.8	104.1	14.5

Part F: Our LnPP1 pseudopotentials and basis sets optimized for lanthanides (CP2K format)

The GTH pseudopotentials from La to Lu are:

```
La GTH-PBE-q11
4 6 1 0
0.536621886    2    20.187083390    -1.461153313
4
0.541927081    2     1.342431782     0.450502384
                                     -1.181781197
0.478778498    3     0.979802043     0.376291820     0.008876396
                                     -0.824267358    -0.021005386
                                                      0.029857280
0.626441601    1     0.328796868
0.300352697    1 -18.352319697

Ce GTH-PBE-q12
4 6 0 2
0.539004680    2    18.85114190 -0.77960224
4
0.498081460    2     1.19529290    0.62204575
                                   -1.62871685
0.47069322     2     1.18088300    0.55487102
                                   -1.53066452
0.65606409     1     0.07990999
0.30705426     1 -17.32458585

Pr GTH-PBE-q13
4 6 0 3
0.532486240    2     18.514048206    -0.582682182
4
0.524682347    2     1.598493376     0.440830770
                                     -1.719989118
0.461080992    2     0.890293235     0.917885701
                                     -1.989243896
0.571698543    1     0.017967543
0.301480737    1 -17.898484476
```

```
Nd GTH-PBE-q14
4 6 0 4
0.531351369    2    18.007481210        -0.707850932
4
0.494947486    2    1.730672873          0.811897086
                                         -2.169289264
0.483476215    2     0.754160538         0.658311314
                                         -1.583172072
0.380970008    1     0.043574337
0.294938171    1 -18.499650577

Pm GTH-PBE-q15
4 6 0 5
0.528636435 2      18.245839784    -0.486314424
4
0.484664155 2       1.282682334     0.959291011
                                    -2.495974035
0.472066166 2       0.193983187     0.667699003
                                    -1.597663074
0.473753437 1      -0.430483026
0.291785846 1    -19.326738513

Sm GTH-PBE-q16
4 6 0 6
0.525294147    2     17.251599705 -0.531859084
4
0.479541125    2       1.730666754   1.009862710
                                    -2.661213312
0.489714555    2    -0.091063341     0.465485746
                                    -1.109941887
0.470798767    1    -0.410255744
0.284444333    1 -19.973327048

Eu GTH-PBE-q17
4 6 0 7
0.522100054    2    17.359358474    -0.648460166
4
0.468993404    2     1.776114604     1.117935246
                                    -2.917719665
0.445886269    2     0.500415952     0.890257661
                                    -2.117280466
0.489972387    1    -0.425678559
0.278525768    1 -20.949306600
```

```
Gd GTH-PBE-q18
4 6 0 8
0.517500043    2    17.556643024    -0.667762780
4
0.462629406    2     1.481423675     1.139411985
                                     -2.993835804
0.457796465    2    -0.173373434     0.619844494
                                     -1.515130079
0.484114514    1    -0.566029370
0.273335071    1   -21.965466277

Tb GTH-PBE-q29
4 6 10 9
0.500000000    1    -7.272801776
4
0.259166302    2   229.865072125   -254.613943211
                                    295.196064370
0.270251357    2   398.718952272   -233.124922698
                                    168.561767849
0.245407837    1   -13.288686713
0.180455163    1   -33.908357345

Dy GTH-PBE-q30
4 6 10 10
0.500000000    1    -7.465003311
4
0.252286021    2   233.922888625   -255.562057179
                                    292.904849233
0.266723795    2   390.748247766   -235.364530025
                                    169.923036441
0.252493399    1   -13.041564434
0.180670438    1   -34.118812490

Ho GTH-PBE-q31
4 6 10 11
0.500000000    1    -7.070002029
4
0.242339356    2   237.963483100   -257.518622800
                                    293.321138600
0.261022150    2   434.742743633   -250.778942067
                                    168.922458067
0.259084819    1   -13.212357286
0.183863195    1   -33.334176813
```

```
Er GTH-PBE-q32
4 6 10 12
0.500000000    1     -8.440641881
4
0.240632456    2    229.192874000    -251.217461600
                                       288.569581400
0.262955148    2    452.933664300    -246.816963067
                                        158.044670000
0.251742832    1 -14.143079042
0.175943441    1 -37.369242337

Tm GTH-PBE-q33
4 6 10 13
0.500000000    1    -7.820984622
4
0.229824520    2    237.571048959    -252.425857659
                                       284.841129864
0.245648958    2    439.356463332    -234.946254129
                                       160.668667820
0.268822896    1 -13.225193664
0.180076023    1 -35.800235126

Yb GTH-PBE-q34
4 6 10 14
0.500000000    1    -7.857588330
4
0.217536443    2 261.367665552    -258.017370096
                                      284.373264272
0.278341463    2 452.078741970    -249.539240792
                                      147.629693880
0.269939733    1 -13.844248648
0.185203058    1 -33.857938124

Lu GTH-PBE-q35
4 6 11 14
0.490000000    1    -8.165545656
4
0.220945258    2 317.613963120    -245.012664370
                                      227.382408876
0.252847006    2 515.381490011    -256.956789586
                                      148.927676699
0.222838425    1 -20.524328427
```

The corresponding MOLOPT basis sets are:

```
La DZV-MOLOPT-GTH
  1
  2 0 3 7 3 2 2 1
        2.23164410000000       6.36920883417662E-01       1.03388584239346E-01      -
2.06422567585900E-01      -3.81942784914523E-01       5.20906412067701E-01
4.73211491187013E-01       1.15889907098170E-01       6.93296946031992E-01
        2.16099087000000      -6.75418466404104E-01      -8.42664751856035E-02
2.59875568312406E-01       4.37373559309912E-01      -5.93937877136375E-01      -
5.49741748678359E-01      -1.46277969697783E-01      -7.17876496123734E-01
        1.41317457000000      -9.66147249143188E-03      -6.64158779908996E-02      -
1.95672298632518E-02      -3.45694579474865E-02       4.93015199101241E-02
7.82129620957820E-02      -4.59601495753897E-02       5.97219634971428E-02
        0.50086179000000      -8.68264727109291E-02       1.43122704225202E-02      -
4.91563714156940E-01      -8.83582552959101E-03       1.94125027653746E-01
1.64040718482716E-01       5.59472052518504E-01       1.02116139497366E-02
        0.21269094000000       1.20385193656121E-01      -2.59245000182893E-01
9.30792964823556E-02      -4.37185110575576E-02       8.28677725547327E-02
3.63493703937926E-01       4.13305278955870E-01       1.77787925070400E-02
        0.08321277000000       2.55759093405274E-01       4.04459579776489E-01
7.53245521359801E-01      -6.07374755222125E-01      -4.25188404683415E-01
5.25530276796041E-01      -5.82889025583462E-01       2.19592403155021E-03
        0.03171828000000      -2.24953760541049E-01       8.64175057544492E-01      -
2.67944173556757E-01      -5.39198996607485E-01      -3.84872569218779E-01
1.80328935494740E-01      -3.73456961853954E-01       8.85102846811129E-04

Ce DZV-MOLOPT-SR-GTH
  5
  2 0 2 7 3 2 2
        4.18237524000000      -2.55709413226539E-02       7.36576300368870E-03      -
8.32438573926917E-03       2.04333917053169E-02       2.29293502559452E-02
4.66384465884811E-03      -8.97726446250939E-03
        2.47592859000000      -2.61623852719931E-02      -9.37132186661999E-03      -
9.37683158753043E-02      -8.00136921826957E-02      -8.65367299268815E-02      -
1.86416546876456E-02       3.42415058191077E-02
        1.57226158000000       4.66464565930329E-01       3.87198316643733E-02
4.44948388737540E-01      -2.48812113210110E-01      -2.75619313069053E-02      -
8.97742761765502E-02      -8.58222366695025E-02
        0.75072837000000      -4.20528848168514E-01      -3.47952702110125E-01      -
1.49275170519501E-01       6.39627584982445E-01       4.43127743588676E-02
3.29281054212053E-01       1.74049133412445E-01
        0.32763789000000      -5.33826251578161E-01      -1.26830422964948E-01      -
2.34686468638884E-01       6.24649527433705E-01       3.92774887661322E-01
8.23168217192468E-01       2.23887188040902E-01
```

0.12786786000000 2.96715777665402E-01 4.92379406450713E-01 -
7.68362738102677E-01 3.88855309298648E-02 3.67189162504830E-01
4.52912034209198E-01 -5.82760423497633E-01
0.05097481000000 -4.80850531409368E-01 7.86615649161499E-01
3.54142664048518E-01 -3.61178977485272E-01 8.36757051839597E-01 -
2.02504175300237E-02 -7.55879082055740E-01
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2 3 3 1 1
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2 3 3 1 1
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Pr DZV-MOLOPT-SR-GTH
5
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4.39591796000000 -8.31398562719043E-03 2.48341744528545E-02 -
1.05972247582625E-02 8.58418269268012E-03 5.07646221667311E-02
1.06767902584073E-02 -2.78422614696601E-02
2.56776794000000 -4.00895549063873E-02 -1.64983644699875E-02 -
8.21043904821067E-02 -7.00378391774263E-02 -1.65003425642652E-01 -
3.74162025200424E-02 7.94712225353780E-02
1.57060627000000 4.19900087764128E-01 -1.19913081586626E-01
5.08085579909234E-01 -2.50298914949444E-01 3.92597332654944E-02 -
6.33497448055813E-02 -1.31375996370367E-01
0.78494829000000 -4.11629409226868E-01 -2.13669589456472E-01 -
3.19766212239041E-01 6.31410227527661E-01 3.59110194577983E-02
3.00047271985428E-01 2.07285766973044E-01
0.34094115000000 -5.37128529423651E-01 -7.73284440975443E-02 -
2.03032949162405E-01 6.21327598603499E-01 4.23614239083528E-01
8.12607734494970E-01 1.66127500478465E-01
0.13034558000000 4.62826527141755E-01 5.12750571651612E-01 -
6.64754606346762E-01 1.43895101247268E-01 1.60039642730230E-01
4.93094570446624E-01 -3.54360969191497E-01
0.06078830000000 -3.87109028102813E-01 8.18647903655388E-01
3.86812765798592E-01 -3.56284725591908E-01 8.73100328649776E-01
3.11591524425493E-02 -8.82899795419343E-01
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2 3 3 1 1

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Nd DZV-MOLOPT-SR-GTH
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5.53525329419063E-03 5.56890457881671E-03
2.94144388000000 1.01926178672860E-03 6.82301673560527E-02 -
1.39775590815099E-01 -4.50888096035157E-02 -4.28783480100715E-03 -
1.56539638362687E-02 -1.82796944740753E-02
1.71549407000000 3.54281925073881E-01 -1.33505211062628E-01
5.56087176114545E-01 -2.79326785460719E-01 -1.12804459149377E-01 -
7.60155188722356E-02 2.61790755541406E-03
0.82158105000000 -4.19471586499703E-01 -1.01795034100547E-01 -
2.77163650823389E-01 5.87707373866578E-01 6.44180068537356E-02
2.79449538875024E-01 3.75493084850524E-02
0.35689793000000 -5.34441025251133E-01 -2.26726231314072E-01 -
2.66709969039673E-01 7.22249475705701E-01 9.89946373806572E-02
8.06719400515469E-01 3.72451471686853E-01
0.13970930000000 4.80328814451032E-01 4.87706533380857E-01 -
6.24869521097830E-01 8.95239761610916E-02 4.68417975184734E-01
5.14754612271509E-01 -5.91500265323160E-01
0.04267857000000 -4.26430515513051E-01 8.23290773053057E-01
3.64313149311161E-01 -2.11079014770283E-01 8.68156443786987E-01
9.24167070790305E-03 -7.13880503603356E-01
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Pm DZV-MOLOPT-SR-GTH
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2.34560596362279E-03 -3.70164731710461E-02

3.01992424000000 1.65468640015913E-01 6.02490312219872E-02 -
3.15683735249028E-01 -4.93109777090491E-02 -4.68500836801754E-02 -
7.74261033322132E-03 9.13208096639175E-02
1.70203691000000 3.24279923539893E-01 -9.26069834332587E-02
5.90566444993444E-01 -2.29964246796829E-01 -1.03377395525530E-01 -
6.97801846876570E-02 -1.28203327972035E-01
0.80884291000000 -5.21039727425882E-01 -8.81250330722742E-02 -
2.45708085096556E-01 6.32870724812486E-01 7.45952924969446E-02
3.40680528075547E-01 1.86335061049843E-01
0.34454172000000 -5.84429805020692E-01 -2.60319099057729E-01 -
1.05500973055877E-01 7.04779397263568E-01 1.19311339943585E-01
8.18304397046495E-01 2.81203925014031E-01
0.13230951000000 3.42175004945482E-01 3.95063277048839E-01 -
6.13637771395787E-01 3.54229131136986E-02 5.35757309214463E-01
4.57576773823515E-01 -6.62078753574052E-01
0.04044278000000 -3.60852058566285E-01 8.69560250501888E-01
3.07848772048831E-01 -2.14625493980741E-01 8.23812310640968E-01 -
1.86106546082244E-03 -6.49391652840782E-01
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2 3 3 1 1
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Sm DZV-MOLOPT-SR-GTH
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5.14254447000000 -1.75691630797067E-02 6.88448709667556E-03
2.64406530493253E-03 1.27601492233223E-02 1.22259234557231E-02
2.48227490115885E-03 -3.89035740349234E-03
3.24872886000000 1.61140068846953E-02 9.95758193327239E-03 -
8.13721665631279E-02 -3.79293152582252E-02 -2.70323214814880E-03 -
9.04265838870334E-03 9.99838622349882E-03
1.73741769000000 4.34358694121963E-01 -9.32786458763034E-02
4.22598620911098E-01 -2.14150370856299E-01 -1.19678269071057E-01 -
5.94645929148260E-02 -3.57105257243345E-02
0.80945305000000 -6.75730471699104E-01 -9.96399131690378E-02 -
4.46786201674431E-02 6.44953302809119E-01 1.28181264720146E-01
3.71291486166409E-01 1.34362126196976E-01

0.34005735000000 -4.07389323001301E-01 -1.02589168771824E-01 -
3.51262287446826E-01 6.99237232026099E-01 9.46907752651636E-02
8.49626757416796E-01 2.90715740298455E-01
0.12681129000000 3.65140632089842E-01 3.91848727038729E-01 -
7.61919176440182E-01 -2.66419900330171E-02 6.58296122067300E-01
3.69659537814207E-01 -8.05646702579844E-01
0.03914979000000 -2.34219503992175E-01 9.03965967221909E-01
3.29969362095629E-01 -2.16635406176179E-01 7.25789065341539E-01
2.20506571353798E-03 -4.96965012604975E-01
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2 3 3 1 1
4.27900000000000 1.00000000000000
2 3 3 1 1
1.88830000000000 1.00000000000000
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Eu DZV-MOLOPT-SR-GTH
5
2 0 2 7 3 2 2
5.34652456000000 -2.30514587773043E-02 1.97109917409011E-03
7.65388089517225E-03 1.79127928238801E-02 1.20910455595489E-02
3.15865034443402E-03 -1.20089921571522E-04
3.31221973000000 5.65857145370741E-02 1.48139364756093E-02 -
1.10077430163909E-01 -7.56109875194690E-02 -7.26551238711004E-03 -
1.08781591014321E-02 -4.39078342094372E-04
1.80858834000000 3.14760419467496E-01 -2.42194637852433E-02
5.39638657898657E-01 -1.93297982388296E-01 -1.27914269745379E-01 -
5.35186229333161E-02 -2.57654687557505E-02
0.85719456000000 -5.80792172328951E-01 -1.53483337910867E-01 -
2.32486179370011E-01 6.23754797394118E-01 1.68111784667483E-01
3.26834702065335E-01 1.29445726707913E-01
0.35821506000000 -4.09137777535115E-01 -1.76814321555905E-01 -
3.20829100639841E-01 7.09575935204607E-01 9.74337317661253E-02
8.08824650762631E-01 4.16961656850869E-01
0.13357636000000 5.13372537647854E-01 3.36415594349455E-01 -
7.12766226532001E-01 1.66530558728500E-03 5.64155610279647E-01
4.81286580073895E-01 -7.17118799104300E-01
0.04023003000000 -3.59065362628116E-01 9.11698518252094E-01
1.77757326929121E-01 -2.53035910668474E-01 7.92093745526737E-01
6.59729889214731E-02 -5.42644765876224E-01
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4.31490000000000 1.00000000000000
2 3 3 1 1
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Gd DZV-MOLOPT-SR-GTH
5
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5.59677090000000 -1.35878826330605E-02 6.72095162666337E-04
1.28533946067190E-02 8.85750972497280E-03 1.75484692016863E-02
3.12812909049960E-03 -2.12036726871308E-03
3.50230035000000 5.91186819277078E-03 2.02410127252069E-02 -
1.15129279080133E-01 -3.77008387521725E-02 -1.02465087973366E-02 -
1.11420421226726E-02 5.47631500541752E-03
1.89721105000000 4.70003822920645E-01 -1.10331786006664E-01
3.96861575545720E-01 -2.03550364664786E-01 -1.42685632578040E-01 -
5.12878754602592E-02 -2.79764089883626E-02
0.91341748000000 -6.55628449474706E-01 -5.14502449345743E-02
1.67966445478583E-02 6.37811214261053E-01 1.40238291412014E-01
3.42640447025973E-01 1.09841840429153E-01
0.38939153000000 -4.94687687723429E-01 -8.43818737817532E-02 -
2.22733172437647E-01 6.84442075224601E-01 1.79556488628826E-01
8.39165265433382E-01 3.37788624190813E-01
0.14481943000000 3.01933944230836E-01 2.81384869857912E-01 -
8.37666389976923E-01 -2.97743830042233E-02 5.45815691374363E-01
4.18792162468975E-01 -7.48983095760161E-01
0.05080032000000 -1.14665369120110E-01 9.47877562439806E-01
2.78394962116352E-01 -2.84458831934409E-01 7.93350988279841E-01
1.57457785779562E-02 -5.58606097604262E-01
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Tb DZV-MOLOPT-SR-GTH
6
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12.62165193000000 2.63150313397855E-01 2.75920747568875E-01 -
2.29534159641915E-01 3.23339286916022E-03 4.42554560140806E-02
3.66201960419660E-03 2.45332089696323E-02 5.58475121303479E-04
6.72437222000000 1.12100175379467E-01 3.30092032872405E-01 -
5.42442558532860E-01 2.10462052221002E-01 -1.16624848593508E-01 -
3.08486266065285E-01 -3.44853285345094E-01 -6.06346654165208E-01
3.41699150000000 3.00217068620189E-01 -5.87552929899234E-02
5.17949773393714E-01 -4.85442534528900E-01 2.88189417112265E-01 -
2.97287705027119E-01 3.55693485753689E-01 -6.17295238061172E-01
1.58752341000000 -2.07461573961026E-01 -2.35011210559256E-01
3.27973544983787E-01 7.92373070753704E-02 -5.47909128510165E-01
9.32072675152942E-02 6.26385825767554E-01 -2.31015730704829E-01
0.65525611000000 -5.96201511927088E-01 -5.15564402503859E-01 -
3.70688898005795E-01 7.56159063449380E-01 9.54644339323703E-02
3.22225314789337E-01 -5.57532338519373E-01 -3.35364771268595E-01
0.24700792000000 7.57032345089346E-02 7.15323211326409E-02 -
2.72623762736979E-01 3.69657224072585E-01 4.84757097453906E-01
8.06993522602640E-01 1.85663574943541E-01 -2.61362721260212E-01
0.05016424000000 -6.51020466450908E-01 6.96635493824822E-01
2.55924893608170E-01 7.30703225488018E-02 -5.97561414739346E-01
2.29537665688777E-01 -1.27608789503469E-01 -1.30926477266939E-01
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Dy DZV-MOLOPT-SR-GTH
6
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12.56351020000000 1.01946219877616E-02 5.30482693003410E-03 -
3.76876660244758E-02 8.48062946355694E-03 8.85392342837320E-03
1.49696948253197E-03 2.13887030494234E-03 2.68641670216845E-03
6.70570037000000 2.43612524808085E-01 -3.04390742530620E-02 -
2.96034516351133E-01 1.78436344905181E-01 7.93368397957354E-02
9.49883082377906E-02 1.20455701847814E-01 -4.90817398295040E-01
3.49925238000000 -4.53996389328329E-01 5.09401495444984E-02
5.23194485404674E-01 -4.13540069787687E-01 -1.88544862045712E-01
8.42174340487461E-02 2.45503456954647E-01 -7.40788288552856E-01

1.61426607000000 1.09913768476936E-01 -3.11635564438197E-02
1.04967358280927E-01 5.41768248520402E-02 1.62197229205481E-02
7.98472407187404E-02 5.32084121930716E-02 -3.39305549873108E-01
0.64232855000000 -3.38884634569258E-01 -7.32588443193683E-02 -
7.85010704959878E-01 7.44567651957788E-01 2.32011470829891E-01
6.54211023933368E-01 -6.59968502658633E-02 6.21098379814215E-02
0.22899342000000 7.40793097792432E-01 -2.98301199419964E-01
6.77717437482374E-02 3.52006590293371E-01 1.52285365706923E-01
6.50127816446322E-01 -4.52268365211918E-01 -1.84175346371598E-02
0.07047220000000 2.42359320289416E-01 9.49278037056382E-01 -
7.31968096556035E-02 -3.40365104142343E-01 9.38506725746041E-01 -
3.56158083972736E-01 -8.44677754060626E-01 -3.01659878850734E-01
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10.98000000000000 1.00000000000000
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1.59200000000000 1.00000000000000
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0.50630000000000 1.00000000000000

Ho DZV-MOLOPT-SR-GTH
6
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12.30521213000000 -1.09498800559440E-02 -6.73056881188356E-03 -
5.73216470190983E-02 1.20748030014911E-02 3.89315123139363E-03
2.89861562096413E-03 7.00110635542719E-03 7.70333920625418E-03
6.89265331000000 9.04473460907019E-02 -1.63214032728612E-02 -
3.50974648148776E-01 1.35767925014780E-01 1.28924983132233E-01
1.44050961927728E-02 -4.62773533071611E-02 -4.25684611516166E-01
3.74514009000000 -1.76834465914595E-01 2.56788971158107E-02
6.63538933842279E-01 -3.21056401626826E-01 -2.87964852316636E-01 -
4.91771078981686E-03 2.56508278332187E-02 -8.19487178211919E-01
1.73557208000000 2.29124216333128E-01 8.10209457811409E-02
6.57130385447420E-02 -3.41922315822506E-03 6.05679671746153E-02 -
4.16969980457300E-02 -8.64145527850708E-02 -3.81161909915498E-01
0.70494403000000 -6.56905573137469E-01 -3.09163255173879E-01 -
5.03533091094195E-01 7.10509992228838E-01 3.44697363965317E-01
6.42333275737345E-01 3.81350731707496E-02 -3.01507808256395E-02
0.26191801000000 6.56261520761308E-01 -2.01810548823772E-02 -
3.66680107547675E-01 2.25659292495078E-01 4.02921559040170E-01
6.11281004260719E-01 -6.26033177283207E-01 2.41189582625750E-02

0.04869946000000 -2.13838554090060E-01 9.46823805414948E-01 -
2.02309243195857E-01 -5.67966372429461E-01 7.84607482641121E-01 -
4.60179056907483E-01 -7.72212489094851E-01 1.99709671065952E-02
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2 3 3 1 1
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2 3 3 1 1
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2 3 3 1 1
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Er DZV-MOLOPT-SR-GTH
7
2 0 2 7 3 2 3
13.29786362000000 -1.88258997114713E-02 -1.12923622850546E-02 -
3.59985774371780E-02 2.91511147970692E-03 -2.85616974662119E-03
4.46317596728246E-03 5.55068354904846E-03 6.09241047822173E-03
7.20153202000000 2.69801972656753E-02 -3.14201631140619E-02 -
3.91538351118871E-01 1.55734983485313E-01 1.16291132649830E-01 -
6.65793505476507E-02 -5.72197683089214E-02 -4.33672919878100E-01
3.89149849000000 -7.45325639894567E-02 3.91965351360620E-02
6.79857949307220E-01 -3.36553888837010E-01 -2.43485178180600E-01 -
1.44060017335282E-01 1.10111833987007E-02 -7.99137864521800E-01
1.82211428000000 2.41658115702460E-01 1.20026388872551E-01
3.05374279128731E-02 -3.15752575901760E-02 5.16366934175329E-02 -
1.14364636559818E-01 -8.40688648901724E-02 -3.66104555330849E-01
0.74199544000000 -6.87232115788379E-01 -4.34879554084935E-01 -
3.48116489314522E-01 7.53344784053229E-01 2.40969421567430E-01
6.15851897059552E-01 -1.19652008766815E-01 -1.36134886731089E-01
0.27368575000000 5.48780601953728E-01 6.00335122120518E-02 -
4.75675470753964E-01 2.84899449743213E-01 3.87598221257440E-01
4.36960349456210E-01 -7.60521407772502E-01 -3.30104134632282E-02
0.04998959000000 -4.01890254204036E-01 8.88942239046961E-01 -
1.86574951899199E-01 -4.61277075017281E-01 8.46296751387000E-01 -
6.25705046601108E-01 -6.29918131935292E-01 1.40051470432146E-01
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10.21230000000000 1.00000000000000
2 3 3 1 1
4.65890000000000 1.00000000000000

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 2 3 3 1 1
        0.29960000000000        1.00000000000000

Tm DZV-MOLOPT-SR-GTH
 6
 2 0 2 7 3 2 3
        13.07159832000000        4.82640863819318E-01        5.30962714586083E-01
4.81035344739481E-01        1.54527161195963E-02        2.73918158063254E-02
1.94326800952731E-02        1.26343666206368E-01        2.20430204249432E-02
         7.69124664000000       -1.40423485625341E-01        7.94492570837716E-02
2.69608993815981E-01        1.60976620751110E-01       -1.29694839216536E-01       -
1.45524359039653E-01       -4.30024180852938E-01       -2.43106400772169E-01
         4.14640499000000        3.46080701857410E-01       -7.55963373952662E-02       -
4.55438266261998E-01       -3.88879276828200E-01        2.57769097415642E-01       -
4.56195408905127E-01       -8.46920750804919E-02       -7.68665681565586E-01
         1.90595415000000       -2.04332883476302E-01       -1.67129705586130E-01       -
3.84395783723523E-01       -3.48504723505320E-02       -3.08933145344357E-01       -
1.29944762570536E-01        7.36416808121053E-01       -3.11042111935698E-01
         0.77641959000000       -4.25703349727655E-01       -4.05224581951437E-01
5.22087096462240E-01        8.01263862408963E-01        3.72438344330715E-01
2.34823968919336E-01       -4.71414933279035E-01       -2.32789244687416E-01
         0.28494144000000       -1.84170524385739E-01       -5.25601562277349E-02
1.42354998125132E-01        4.19977595100555E-01       -5.31270827649495E-01
7.85476150749333E-01        1.53381690023162E-01       -4.13625079522189E-01
         0.05144299000000       -6.08829317433754E-01        7.14947364993345E-01       -
2.18842347047294E-01        5.47826266039235E-02        6.32127620495588E-01
2.85184255648461E-01       -6.22578258038006E-02       -1.65959991829858E-01
 2 3 3 1 1
        35.69000000000000        1.00000000000000
 2 3 3 1 1
        13.54000000000000        1.00000000000000
 2 3 3 1 1
         5.29900000000000        1.00000000000000
 2 3 3 1 1
         1.97000000000000        1.00000000000000
 2 3 3 1 1
         0.61620000000000        1.00000000000000
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Yb    DZV-MOLOPT-SR-GTH
 6
 2 0 2 7 3 2 3
        12.17373040000000        2.07362348970018E-04        1.47446619839764E-01       -
1.09022057680819E-01       -2.26197536087966E-02       -2.50123759552461E-03       -
8.13330415547623E-01        2.03729589600744E-01       -4.64591114630638E-01
        6.86323383000000       -1.82670457585053E-01        1.24214794565837E-02       -
2.48388016900023E-03        1.00691019419771E-01        1.31104615382825E-01
1.33501650894867E-01        5.72399632057193E-02       -3.34757394645174E-01
        3.87428635000000        4.42159476178132E-01       -3.35744116700326E-01
2.45970619601946E-01       -2.03661757828175E-01       -3.20891816087186E-01
4.69782601167514E-01        1.83487196328873E-01       -6.94605645824650E-01
        1.84256942000000       -2.76505025141618E-01       -1.68171598954976E-01
2.02033253141520E-01        2.38589211197316E-01        3.34519917406246E-02
2.18894837608890E-01        8.61239507450086E-02       -2.83153705299204E-01
        0.75839072000000       -5.19918580936243E-01        2.05718231618124E-01       -
5.75119844827363E-01       -1.69853029653692E-01        8.45138662995554E-01       -
2.23656386738234E-01       -2.84209442147208E-01       -2.51781588606154E-01
        0.27877809000000        6.12342350285100E-01        5.88020817282881E-01       -
3.33304631992987E-01        3.92808504188513E-01        3.89175896956264E-01
2.60451585081994E-02       -8.55689597000181E-01       -1.89187697633446E-01
        0.05199452000000       -2.22245235873348E-01        6.70082778598187E-01
6.67033902117279E-01       -8.41307416708132E-01        1.14006542755405E-01       -
3.68750925614999E-02       -3.18046005170323E-01       -1.01260088897879E-01
 2 3 3 1 1
        40.55000000000000        1.00000000000000
 2 3 3 1 1
        14.98000000000000        1.00000000000000
 2 3 3 1 1
         5.86000000000000        1.00000000000000
 2 3 3 1 1
         2.16900000000000        1.00000000000000
 2 3 3 1 1
         0.67220000000000        1.00000000000000

Lu DZV-MOLOPT-SR-GTH
 7
 2 0 2 7 3 2 3
        12.64717647000000       -7.46473450415586E-02       -1.25017022698197E-02       -
8.45216706360905E-02        1.43102338976610E-02        1.26295142205977E-02
9.65694842306751E-03        7.80936697122167E-03       -5.45048154100064E-02
        7.31232980000000       -4.70383438631707E-02       -5.44258989060867E-03       -
1.36821300357875E-01        7.64829732394098E-02        6.51444846689639E-02
6.26840514242059E-02        1.98598556769331E-02       -7.27964859722694E-01
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3.50738970000000 3.67222823169333E-01 7.63693693465873E-02
6.20098730302488E-01 -3.38372487389196E-01 -2.81597037450698E-01 -
2.92463179712271E-02 5.51097975893657E-02 -6.52038066927356E-01
1.57826189000000 -4.72308055533134E-02 3.91918956568624E-02 -
2.72035695698600E-01 1.91838648929557E-01 1.87980237143454E-01
1.10822507645460E-01 -4.78959753056949E-02 -1.94475956844904E-01
0.64515220000000 -6.20042694984947E-01 -5.75136636811166E-01 -
7.95333334332790E-02 7.73251787600272E-01 2.82041547226272E-01
7.82074534398664E-01 2.24413920041415E-01 4.65196306045735E-02
0.23243020000000 2.69750411342863E-01 3.22639300949557E-01 -
6.89246132356973E-01 8.86174748654325E-02 3.99227488185820E-01
5.82338001287281E-01 -5.40148023467217E-01 7.64656887296803E-03
0.05975385000000 -6.30803922951773E-01 7.46704446803679E-01
1.84998329203890E-01 -4.86699271302765E-01 8.01269747803634E-01 -
1.79115472894852E-01 -8.07522130294446E-01 -4.35809998258455E-02
1 3 3 1 1
28.36240000000000 1.00000000000000
1 3 3 1 1
13.24780000000000 1.00000000000000
1 3 3 1 1
6.14400000000000 1.00000000000000
1 3 3 1 1
2.76230000000000 1.00000000000000
1 3 3 1 1
1.15740000000000 1.00000000000000
1 3 3 1 1
0.42440000000000 1.00000000000000