Electronic Supplementary Information
for
In situ Raman investigation of Dy complexation in Cl-bearing aqueous solutions at 20-300 °C

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Supplementary Materials:

- Details of calculations used to simulate Dy speciation in experimental fluids
- Supplementary figures S1 and S2
- Supplementary tables S1 through S10
- Supplementary tables in xlx format (Supplementary_materials_tables)
- Zipped folder containing reference raw spectra for solutions and $DyCl_3$ solids measured in this study (Supp_raw files).
    - Supp_raw_MilliQ
    - Supp_raw_3mNaCl
    - Supp_raw_1.7mNaCl_pH2
    - Supp_raw_1.7mNaCl_pH4
    - Supp_raw_1mNaCl_pH2
    - Supp_raw_1mNaCl_pH5
    - Supp_raw_0.14mDyCl3_pH2
    - Supp_raw_0.3mDyCl3_pH2
    - Supp_raw_0.6mDyCl3_pH2
    - Supp_raw_1.8mDyCl3_pH2
    - Supp_raw_Capillary
    - Supp_rawDyCl3_solids
- Reference spectra for Dy_2O_3 and $Dy(OH)_3$ solids area available in the supplementary materials provided by Hurtig et al.[1] (doi:10.1039/D4DT01086H).

Speciation calculations

Thermodynamic data for the minerals and aqueous species in the Dy-Cl-O-H component system used to calculate speciation models presented in Figures 8 and 10 were taken from the MINES thermodynamic database (Gysi et al., 2023) with data sources used summarized in Table S6. The TSolMod library [2] is used in GEM-Selektor for all of the activity and equation of state (EoS) calculations. The revised Helgeson-Kirkham-Flowers EoS [3–6] was used to calculate all thermodynamic properties of aqueous species and the IAPS-84 EoS [7] was used to calculate the properties of H_2O. The activity coefficients (γ_i) of charged aqueous species were calculated using the extended Debye-Hückel equation (Robinson and Stokes, 1968)

$$\log(\gamma_i) = \frac{-A \cdot z_i^2 \cdot \sqrt{I}}{1 + a \cdot B \cdot \sqrt{I}} + \Gamma_\gamma + b_\gamma \cdot I \qquad \text{(S1)}$$

with the ionic strength (I) calculated as

$$I = \frac{1}{2} \sum_i m_i \cdot z_i^2 \qquad \text{(S2)}$$

where A and B are the Debye-Hückel parameters from Helgeson et al. [3]; $\mathring{a}_i$ is the ion size parameter taken from Kielland [8] for charged species; Γ_γ is a mole fraction to molality conversion factor; $b\gamma$ is the extended term parameter; m_i and z_i are the molal concentration and the charge of the ith aqueous species respectively. The γ_i was set to unity for all neutral aqueous species and the γ_i of water was calculated from the osmotic coefficient [3].

Peak fitting of the water stretching band and low frequency region

An example of the 3-peak de-convolution used to fit the water stretching band (Fig. 4 B, D, E; ESI Table S4) is shown in Figure S1 panels A and B. Sub-peak S-1 represents the most fully hydrogen bonded network water, S-2 represents intermediately networked water, and S-3 represents the least strongly hydrogen bonded water. Comparison of the relative contributions of each sub-peak at ambient temperature vs. 300 °C illustrates the changing structure of water with increasing temperature, i.e. with increasing temperature the hydrogen bond network of water decreases.

The peaks used to fit the Dy-O ($\nu_{1,Dy\text{-}O}$) and Dy-Cl ($\nu_{Dy\text{-}Cl}$) vibrational modes measured in the low frequency region (Fig. 6) of the $DyCl_3$-bearing solutions and reported in Table 4 and ESI Table S5 are shown in Figure S1 panels C and D. At ambient temperature only the $\nu_{1,Dy\text{-}O}$ could be fitted due to the overlap of the water translational region with the region for $\nu_{Dy\text{-}Cl}$, whereas at 300 °C both $\nu_{1,Dy\text{-}O}$ and $\nu_{Dy\text{-}Cl}$ could be fitted in the 1.8 mol/kg solution. Each vibrational mode was fitted using a single Gaussian peak.

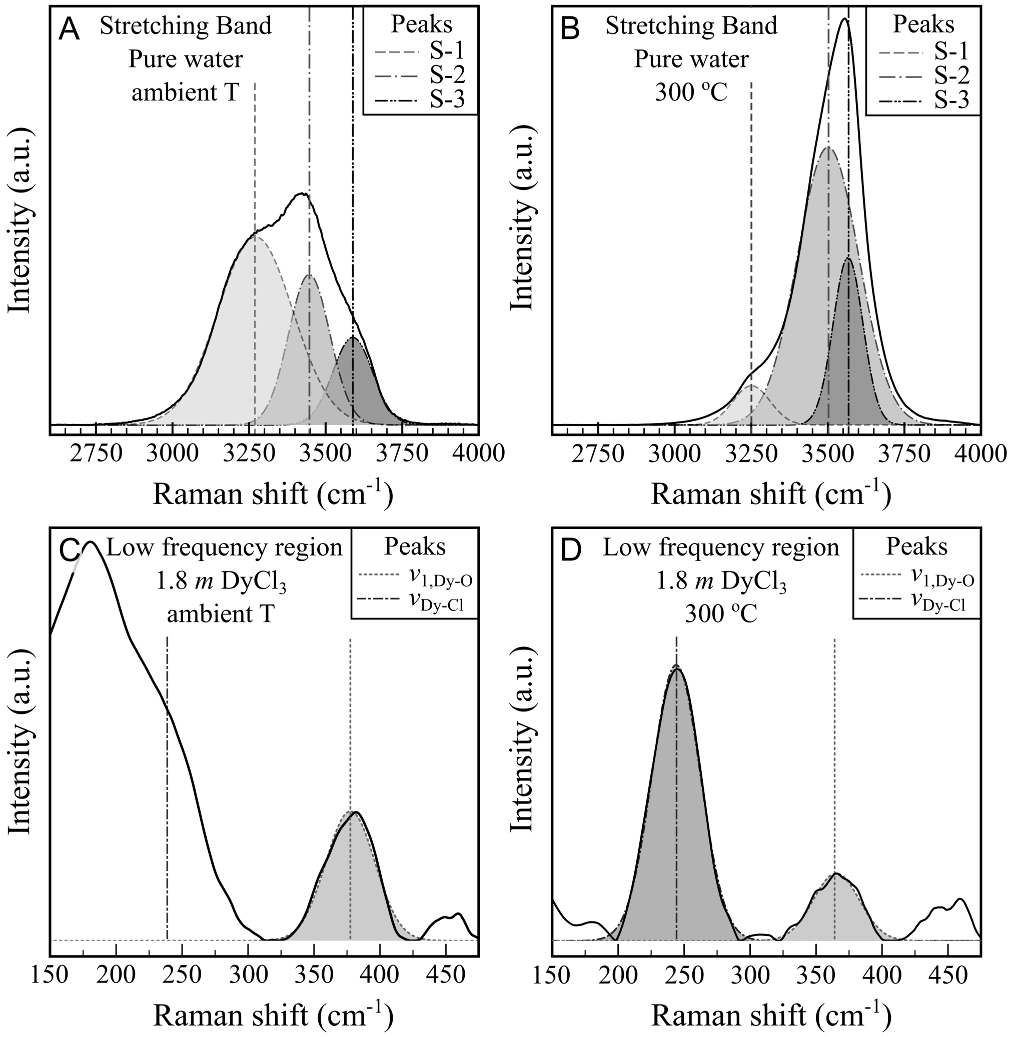

Figure S1: Representative examples peak fittings and de-convolution used in this study. Raman spectra of pure water (MilliQ) illustrating the fitting of the water stretching band with three sub-peaks (S-1 to S-3) at (A) ambient temperature and (B) 300 °C. Similar fits were used for other spectra as described in text and reported in Table S4. Raman spectra of 1.8 mol/kg $DyCl_3$ experimental solution illustrating the fitting of the low frequency region with peak(s) for (C) the Dy-O stretching mode of the Dy^{3+} aqua ion ($\nu_{1,Dy\text{-}O}$) at ambient temperature and (D) $\nu_{1,Dy\text{-}O}$ of the Dy^{3+} aqua ion and the Dy-Cl vibrational mode of Dy chloride complexes ($\nu_{Dy\text{-}Cl}$) at 300 °C. Similar fits were used for other spectra as described in text and reported in Table S5.

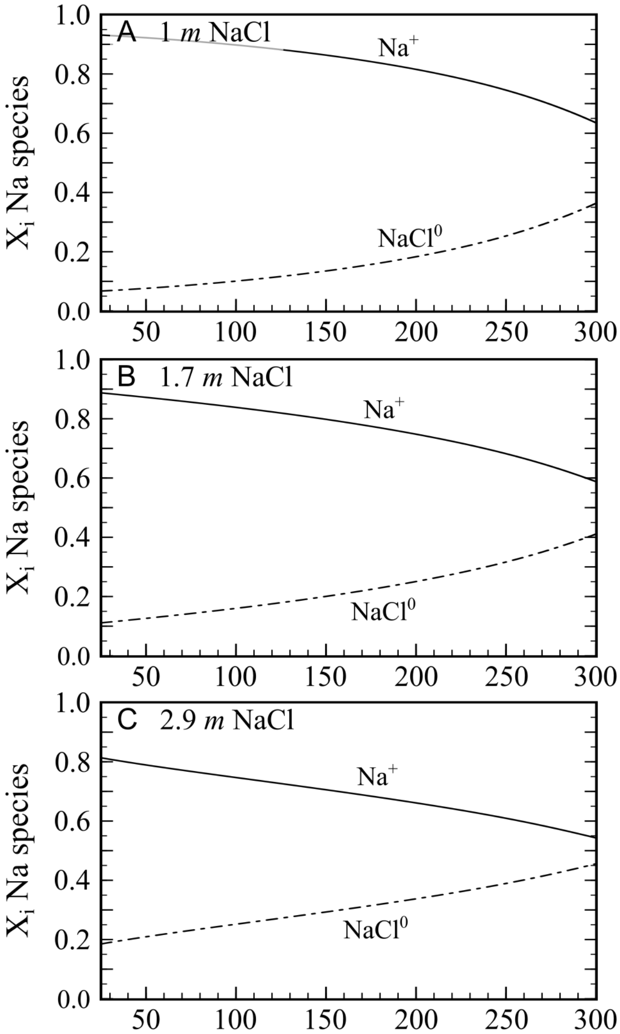

Figure S2: Simulated speciation in mole fraction (X_i) Na species as a function of temperature. A) The 1 mol/kg NaCl, pH 2 background solution, B) the 1.7 mol/kg NaCl, pH 2 background solution, and C) the 2.9 mol/kg NaCl, pH 7 background solution. Models of the 1 mol/kg NaCl, pH 5 and 1.7 mol/kg NaCl, pH 4 background solutions predict speciation very similar to the lower pH models and are therefore not included. Thermodynamic equilibrium calculations were conducted using the GEMS code package and the MINES thermodynamic database. The thermodynamic properties for the Na species are from Shock and Helgeson [4] for Na^+ and Miron et al. [9] for $NaCl^0$ and $NaOH^0$.

Table S1: Crystal lattice parameters of Dy hydroxide [$Dy(OH)_3(s)$] with a hexagonal crystal structure in the $P6_3/m$ space group derived from Rietveld refinement of powder XRD data.

Data source	a Å	c Å	α °	γ °
This study	6.287	3.575	90.1	119.9
Beall et al. [10]	6.286	3.577	90.0	120.0

Table S2: Peak centers for vibrational modes in reference Dy solids measured using the 532 nm and the 266 nm excitation lasers.

Dy_2O_3 cm^{-1} 532 nm	Dy_2O_3 cm^{-1} 266 nm	Vibrational modes (a-c)	$Dy(OH)_3$ cm^{-1} 532 nm	$Dy(OH)_3$ cm^{-1} 266 nm	Vibrational modes (c-d)	$DyCl_3$ anhydr. cm^{-1} 266 nm	Vibrational modes (f,g)	$DyCl_3 \cdot xH_2O$ cm^{-1} 532 nm	$DyCl_3 \cdot xH_2O$ cm^{-1} 266 nm	Vibrational modes (g)
95		F_g	133					62		lattice vibrations
119		F_g+A_g/A_g	262					74		lattice vibrations
135		F_g	314	311	A_g			85		
145		E_g	395	392	A_g/ E_{2g}			94		δ_{ODyCl} and δ_{ClDyCl}
176		F_g+E_g/F_g	406	405	A_g			106		δ_{ODyCl} and δ_{ClDyCl}
264	264	E_g/E_g+F_g	502	499	E_{1g}			111		δ_{ODyCl} and δ_{ClDyCl}
327	326		697	692				135		δ_{ODyCl} and δ_{ClDyCl}
	349		736	731				153		
372	370	F_g+A_g /F_g	1077	1079	δ_{OH}			159		ν_{Dy-Cl}
424	424	F_g/A_g	3597	3598	ν_{OH}			168		ν_{Dy-Cl}
463	461	F_g	3611	3612	ν_{OH}			193		δ_{ODyO}
	522							209	211	δ_{ODyO}
567	561					242	ν_{Dy-Cl}	240	237	ν_{Dy-Cl}
584	583	F_g				255	ν_{Dy-Cl}		269	ν_{Dy-Cl}
592	596					272	ν_{Dy-Cl}	315	314	ν_{Dy-H2O}
						340	ν_{Dy-H2O}	359	348	ν_{Dy-H2O}
						383			485	
						425		528		ω_{H2O}
						527	ω_{H2O}	564	569	ω_{H2O}
								600	604	ω_{H2O}
						647	ω_{H2O}	618	635	ω_{H2O}
								670	672	ω_{H2O}
						1598	δ_{HOH}	1621	1622	δ_{HOH}
						1638	δ_{HOH}	1637	1638	δ_{HOH}
								1648	1649	δ_{HOH}
								3224	3231	ν_{H2O}
								3285	3280	ν_{H2O}
						3383	ν_{H2O}	3344	3343	ν_{H2O}
						3471	ν_{H2O}	3394	3397	ν_{H2O}
						3534	ν_{H2O}	3442	3444	ν_{H2O}

References: (a) Schaack and Koningstein[11]; (b) Abrashev et al.[12]; (c) Hurtig et al.[1]; (d) Arunachalam et al.[5]; (e) Sanivarapu et al.[6]; (f) Paptheodorou[13]; (g) Oczko and Macalik[14]. The notation for the vibration types for coordinated OH^- or H_2O molecules: ω – wagging; ν – stretching modes; δ – bending mode.

Table S3: Peak center positions, peak areas, and full width half maximum (FWHM) for Raman modes in the low frequency region for background H_2O-NaCl solutions.

	$\nu_{H2O,L-1}$ cm^{-1}	$A_{H2O,L-1}$	FWHM cm^{-1}
100 °C			
MilliQ	367.3	1.197	39.94
1 *m* NaCl-pH2	372.4	1.237	40.68
1.7 *m* NaCl -pH2	367.0	1.236	36.49
1.7 *m* NaCl-pH4	369.5	1.224	35.16
1 *m* NaCl l-pH5	372.9	1.162	34.52
3 *m* NaCl-pH7	374.3	1.252	44.81
average		*1.218*	
3σ		*0.099*	
150 °C			
MilliQ	370.5	1.077	31.33
1 *m* NaCl-pH2	369.7	1.318	33.00
1.7 *m* NaCl -pH2	367.4	1.610	32.46
1.7 *m* NaCl-pH4	370.2	1.738	34.41
1 *m* NaCl l-pH5	369.8	1.185	28.34
3 *m* NaCl-pH7	370.6	1.944	37.57
average		*1.478*	
3σ		*1.017*	
200 °C			
MilliQ	370.8	1.263	30.00
1 *m* NaCl-pH2	371.5	1.261	31.00
1.7 *m* NaCl -pH2	369.9	1.880	32.58
1.7 *m* NaCl-pH4	367.9	1.942	31.96
1 *m* NaCl l-pH5	370.1	1.347	34.04
3 *m* NaCl-pH7	370.7	2.263	38.92
average		*1.659*	
3σ		*1.277*	
250 °C			
MilliQ	370.2	2.360	38.68H_2O-NaCl
1 *m* NaCl-pH2	372.7	1.552	32.86
1.7 *m* NaCl -pH2	369.3	2.031	33.82
1.7 *m* NaCl-pH4	367.3	2.110	37.35
1 *m* NaCl l-pH5	374.6	1.654	35.17
3 *m* NaCl-pH7	368.8	1.645	39.00
average		*1.892*	
3σ		*0.9669*	

Table S3: continued.

	$\nu_{H2O,L-1}$ cm^{-1}	$A_{H2O,L-1}$	FWHM cm^{-1}
300 °C			
MilliQ	367.6	1.264	31.67
1 *m* NaCl-pH2	367.1	1.575	35.29
1.7 *m* NaCl-pH2	376.2	1.920	41.70
1.7 *m* NaCl-pH4	368.1	2.442	34.95
1 *m* NaCl-pH5	365.3	1.692	38.62
3 *m* NaCl-pH7	370.0	1.962	44.72
average		*1.809*	
3σ		*1.201*	

Symbols: $\nu_{H2O,L-1}$ = peak center position of the librational mode in background solutions used to correct $\nu_{1,Dy-O}$; $\tilde{\nu}_{H2O,T-1}$ = peak center position for peak T-1 of the water translational region; $A_{H2O,L-1}$ = peak area of the librational mode in background solutions used to correct $\nu_{1,Dy-O}$; $A_{H2O,T-1}$ = peak area of peak T-1 of the water translational region used to correct ν_{Dy-Cl}.

Table S4: Peak center positions, peak areas, and FWHM for Raman modes in the water stretching band at 3000-3800 cm^{-1} for background H_2O-NaCl and $DyCl_3$-bearing solutions.

	Sub-peak S-1				*Sub-peak S-2*				*Sub-peak S-3*				*Total*
	$\nu_{H2O,S-1}$	$A_{H2O,S-1}$	FWHM	$A_{H2O,S-1}/A_{H2O,S}$	$\nu_{H2O,S-2}$	$A_{H2O,S-2}$	FWHM	$A_{H2O,S-2}/A_{H2O,S}$	$\nu_{H2O,S-3}$	$A_{H2O,S-3}$	FWHM	$A_{H2O,S-3}/A_{H2O,S}$	$A_{H2O,S}$
Solution ID	cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		
						Background solutions							
Ambient T													
MilliQ	3271	58667	288.7	0.5931	3446	25806	158.8	0.2609	3588	14437	152.6	0.1460	98909
1 *m* NaCl-pH2	3271	51059	275.3	0.5144	3446	33814	166.9	0.3406	3588	14396	148.9	0.1450	99268
1.7 *m* NaCl -pH2	3271	47254	265.8	0.4762	3446	37604	168.5	0.3789	3588	14377	148.7	0.1449	99235
1.7 *m* NaCl-pH4	3271	47220	266.3	0.4760	3446	37551	169.1	0.3785	3588	14430	149.8	0.1455	99201
1 *m* NaCl l-pH5	3271	51088	274.6	0.5147	3446	33925	167.8	0.3418	3588	14254	150.5	0.1436	99266
3 *m* NaCl-pH7	3271	40324	252.0	0.4067	3446	44811	173.4	0.4520	3588	14006	148.4	0.1413	99140
100 °C													
MilliQ	3288	42680	270.0	0.4315	3452	34246	166.3	0.4315	3584	21990	160.0	0.3462	98916
1 *m* NaCl-pH2	3288	36956	259.6	0.3741	3452	40094	169.1	0.4058	3584	21744	156.1	0.2201	98795
1.7 *m* NaCl -pH2	3288	34788	255.8	0.3526	3452	42447	168.1	0.4302	3584	21423	153.3	0.2171	98658
1.7 *m* NaCl-pH4	3288	34846	254.9	0.3522	3452	42677	169.7	0.4314	3584	21405	155.4	0.2164	98928
1 *m* NaCl l-pH5	3288	37047	260.6	0.3752	3452	39984	169.3	0.4049	3584	21720	156.3	0.2199	98751
3 *m* NaCl-pH7	3288	30995	246.0	0.3133	3452	47264	170.0	0.4777	3584	20675	153.5	0.2090	98933
150 °C													
MilliQ	3293	32370	257.6	0.3286	3453	37651	167.7	0.3822	3578	28496	164.1	0.2893	98516
1 *m* NaCl-pH2	3293	28627	251.6	0.2913	3453	41527	167.3	0.4225	3578	28135	160.0	0.2862	98288
1.7 *m* NaCl -pH2	3293	27470	246.4	0.2776	3453	43800	167.3	0.4427	3578	27678	159.1	0.2797	98948
1.7 *m* NaCl-pH4	3293	27741	250.1	0.2807	3453	43109	166.0	0.4362	3578	27967	160.8	0.2830	98818
1 *m* NaCl l-pH5	3293	28662	251.5	0.2903	3453	41818	167.9	0.4235	3578	28260	160.0	0.2862	98740
3 *m* NaCl-pH7	3293	25488	239.6	0.2575	3453	47119	165.9	0.4760	3578	26387	156.0	0.2666	98993
200 °C													
MilliQ	3291	22646	247.0	0.2294	3466	46019	185.4	0.4662	3579	30038	167.2	0.3043	98702
1 *m* NaCl-pH2	3291	23216	254.5	0.2351	3466	49495	191.5	0.5011	3579	26054	169.1	0.2638	98764
1.7 *m* NaCl -pH2	3291	21592	243.4	0.2195	3466	49947	182.8	0.5077	3579	26847	164.2	0.2729	98386
1.7 *m* NaCl-pH4	3291	21252	240.6	0.2151	3466	50617	184.3	0.5122	3579	26947	163.1	0.2727	98815
1 *m* NaCl l-pH5	3291	23224	254.5	0.2352	3466	49481	191.4	0.5011	3579	26045	169.0	0.2637	98751
3 *m* NaCl-pH7	3291	20151	240.9	0.2039	3466	54730	188.0	0.5538	3579	23949	163.9	0.2423	98830

Table S4: continued

	Sub-peak S-1				Sub-peak S-2				Sub-peak S-3				Total
	$\nu_{H2O,S-1}$	$A_{H2O,S-1}$	FWHM	$A_{H2O,S-1}/A_{H2O,S}$	$\nu_{H2O,S-2}$	$A_{H2O,S-2}$	FWHM	$A_{H2O,S-2}/A_{H2O,S}$	$\nu_{H2O,S-3}$	$A_{H2O,S-3}$	FWHM	$A_{H2O,S-3}/A_{H2O,S}$	$A_{H2O,S}$
Solution ID	cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		
Background solutions													
250 °C													
MilliQ	3247	8811	166.5	0.0896	3498	78652	240.6	0.8001	3571	10840	112.1	0.11027	98303
1 *m* NaCl-pH2	3247	10285	184.5	0.1038	3498	83620	240.3	0.8437	3571	5201	85.58	0.05248	99107
1.7 *m* NaCl -pH2	3247	10248	184.2	0.1038	3498	83857	237.1	0.8492	3571	4645	85.02	0.04703	98749
1.7 *m* NaCl-pH4	3247	10269	185.9	0.1038	3498	83977	237.8	0.8491	3571	4656	84.32	0.04708	98901
1 *m* NaCl l-pH5	3247	10257	184.6	0.1037	3498	83474	240.8	0.8437	3571	5205	85.92	0.05261	98936
3 *m* NaCl-pH7	3247	11040	195.1	0.1118	3498	83212	230.1	0.8424	3571	4528	85.00	0.04584	98781
300 °C													
MilliQ	3251	6097	143.2	0.0624	3501	71050	235.5	0.7276	3566	20498	114.15	0.20992	97645
1 *m* NaCl-pH2	3251	6701	149.4	0.0681	3501	76600	237.2	0.7783	3566	15116	104.93	0.15359	98417
1.7 *m* NaCl -pH2	3251	7294	158.3	0.0745	3501	76932	233.5	0.7855	3566	13709	105.11	0.13998	97935
1.7 *m* NaCl-pH4	3251	7238	156.8	0.0737	3501	77645	234.6	0.7911	3566	13266	102.62	0.13516	98149
1 *m* NaCl l-pH5	3251	6858	152.5	0.0697	3501	76191	236.2	0.7743	3566	15351	105.74	0.15600	98399
3 *m* NaCl-pH7	3251	8141	161.8	0.0832	3501	79981	228.9	0.8171	3566	9762	93.30	0.09973	97884
Water-$DyCl_3$-HCl													
Ambient T													
0.14 *m* $DyCl_3$-pH2	3271	56760	286.7	0.5713	3446	28591	160.6	0.2878	3588	13996	151.9	0.1409	99347
0.27 *m* $DyCl_3$-pH2	3271	55116	286.1	0.5555	3446	30482	163.6	0.3072	3588	13617	150.9	0.1372	99215
0.59 *m* $DyCl_3$-pH2	3271	51938	285.5	0.5242	3446	34641	169.9	0.3496	3588	12506	149.3	0.1262	99085
1.8 *m* $DyCl_3$-pH2	3271	44029	290.1	0.4483	3446	46800	185.7	0.4765	3588	7390	139.2	0.07524	98219
100 °C													
0.14 *m* $DyCl_3$-pH2	3288	40913	271.6	0.4140	3452	36335	168.1	0.3677	3584	21578	158.9	0.2183	98827
0.27 *m* $DyCl_3$-pH2	3288	40802	274.5	0.4132	3452	37350	170.8	0.3782	3584	20602	156.5	0.2086	98754
0.59 *m* $DyCl_3$-pH2	3288	40786	284.2	0.4131	3452	38967	171.8	0.3947	3584	18972	156.7	0.1922	98725
1.8 *m* $DyCl_3$-pH2	3288	42760	312.7	0.4339	3452	43929	180.4	0.4457	3584	11865	151.3	0.1204	98553
150 °C													
0.14 *m* $DyCl_3$-pH2	3293	33357	266.8	0.3385	3453	38115	168.3	0.3867	3578	27084	163.2	0.2748	98556
0.27 *m* $DyCl_3$-pH2	3293	31907	267.2	0.3235	3453	39442	169.5	0.3999	3578	27285	161.0	0.2766	98633
0.59 *m* $DyCl_3$-pH2	3293	33352	282.1	0.3380	3453	40439	171.0	0.4098	3578	24881	161.1	0.2522	98672
1.8 *m* $DyCl_3$-pH2	3293	40315	322.6	0.4079	3453	42504	176.2	0.4300	3578	16018	154.6	0.1621	98836

Table S4: continued

	Sub-peak S-1				*Sub-peak S-2*				*Sub-peak S-3*				*Total*
	$\nu_{H2O,S-1}$	$A_{H2O,S-1}$	FWHM	$A_{H2O,S-1}/A_{H2O,S}$	$\nu_{H2O,S-2}$	$A_{H2O,S-2}$	FWHM	$A_{H2O,S-2}/A_{H2O,S}$	$\nu_{H2O,S-3}$	$A_{H2O,S-3}$	FWHM	$A_{H2O,S-3}/A_{H2O,S}$	$A_{H2O,S}$
Solution ID	cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		cm^{-1}		
Water-$DyCl_3$-HCl													
200 °C													
0.14 *m* $DyCl_3$-pH2	3291	24825	262.3	0.2518	3466	45610	184.8	0.4626	3579	28166	166.2	0.2857	98601
0.27 *m* $DyCl_3$-pH2	3291	23953	263.1	0.2431	3466	46749	186.9	0.4744	3579	27836	163.0	0.2825	98538
0.59 *m* $DyCl_3$-pH2	3291	27424	276.7	0.2781	3466	47553	188.3	0.4822	3579	23633	169.3	0.2397	98611
1.8 *m* $DyCl_3$-pH2	3291	35638	327.3	0.3616	3466	47890	192.1	0.4859	3579	15039	162.7	0.15257	98567
250 °C													
0.14 *m* $DyCl_3$-pH2	3247	10261	183.7	0.1045	3498	78961	238.6	0.8041	3571	8981	106.31	0.09145	98202
0.27 *m* $DyCl_3$-pH2	3247	10966	193.0	0.1116	3498	79249	240.5	0.8066	3571	8035	98.10	0.08178	98250
0.59 *m* $DyCl_3$-pH2	3247	14257	228.1	0.1448	3498	78945	236.9	0.8019	3571	5243	85.40	0.05326	98445
1.8 *m* $DyCl_3$-pH2	3247	25113	293.3	0.2524	3498	74026	227.8	0.7440	3571	360	38.0	0.00362	99499
300 °C													
0.14 *m* $DyCl_3$-pH2	3251	6233	149.8	0.0642	3501	68645	237.8	0.7073	3566	22169	115.95	0.2284	97047
0.27 *m* $DyCl_3$-pH2	3251	7913	168.0	0.0811	3501	73109	234.7	0.7492	3566	16564	105.24	0.1697	97586
0.59 *m* $DyCl_3$-pH2	3251	9520	193.7	0.0976	3501	71749	233.7	0.7353	3566	16304	105.35	0.1671	97574
1.8 *m* $DyCl_3$-pH2	3251	20103	273.6	0.2053	3501	72271	220.7	0.7379	3566	5566	85.06	0.0568	97940

Symbols: $\nu_{H2O,S-1}$ to $\nu_{H2O,S-3}$ = peak center positions of sub-peaks S-1 to S-3 of the water stretching band; $A_{H2O,S-1}$ to $A_{H2O,S-3}$ = peak areas of sub-peaks S-1 to S-3 of the water stretching band; $A_{H2O,S}$ = total peak area of the water stretching band; $A_{H2O,S-1}/A_{H2O,S}$ to $A_{H2O,S-3}/A_{H2O,S}$ = ratios of peak areas of sub-peaks S-1 to S-3 to the total peak area of the water stretching band.

Table S5: Peak center positions, peak areas, and FWHM for Raman modes in the region of 200-400 cm^{-1} in $DyCl_3$-bearing solutions.

	Dy-O				Dy-Cl		
	$\nu_{1,Dy-O}$ cm^{-1}	$A_{Dy-O,raw}$	FWHM cm^{-1}	$A_{Dy-O,corr.}$	ν_{Dy-Cl} cm^{-1}	A_{Dy-Cl}	FWHM cm^{-1}
Ambient T							
0.14 *m* $DyCl_3$-pH2	384.3	1.533	38.20	-	-	-	-
0.27 *m* $DyCl_3$-pH2	381.4	2.711	46.34	-	-	-	-
0.59 *m* $DyCl_3$-pH2	378.6	5.399	44.89	-	-	-	-
1.8 *m* $DyCl_3$-pH2	378.0	12.80	43.97	-	-	-	-
100 °C							
0.14 *m* $DyCl_3$-pH2	374.0	1.786	32.00	0.5677	-	-	-
0.27 *m* $DyCl_3$-pH2	372.8	3.188	39.73	1.970	236.5	0.5215	32.70
0.59 *m* $DyCl_3$-pH2	373.3	5.640	46.55	4.422	239.0	0.7784	27.64
1.8 *m* $DyCl_3$-pH2	372.7	11.88	44.27	10.66	240.8	2.683	31.02
150 °C							
0.14 *m* $DyCl_3$-pH2	368.7	1.665	29.74	0.1865	-	-	-
0.27 *m* $DyCl_3$-pH2	370.7	3.212	41.37	1.734	239.5	0.4825	26.23
0.59 *m* $DyCl_3$-pH2	370.6	5.114	45.67	3.635	238.4	1.000	32.39
1.8 *m* $DyCl_3$-pH2	370.3	10.95	42.99	9.473	240.4	3.838	32.85
200 °C							
0.14 *m* $DyCl_3$-pH2	*367.8*	*2.214*	*43.15*	*0.5546*	240.7	0.6008	28.64
0.27 *m* $DyCl_3$-pH2	370.3	2.958	40.15	1.299	238.5	0.9373	29.44
0.59 *m* $DyCl_3$-pH2	367.6	4.901	43.25	3.242	238.1	1.780	32.43
1.8 *m* $DyCl_3$-pH2	367.1	9.666	41.94	8.006	237.0	6.612	40.26
250 °C							
0.14 *m* $DyCl_3$-pH2	*369.6*	*2.329*	*29.36*	*0.4363*	237.7	0.9141	28.52
0.27 *m* $DyCl_3$-pH2	367.6	2.811	51.35	*0.9192*	238.5	1.559	33.14
0.59 *m* $DyCl_3$-pH2	367.6	4.665	44.80	2.773	239.0	3.283	42.07
1.8 *m* $DyCl_3$-pH2	366.1	8.056	41.74	6.164	239.3	12.05	39.18
300 °C							
0.14 *m* $DyCl_3$-pH2	*370.0*	*2.697*	*36.05*	*0.8882*	241.0	1.589	37.00
0.27 *m* $DyCl_3$-pH2	369.3	2.911	37.36	*1.101*	242.1	3.047	29.21
0.59 *m* $DyCl_3$-pH2	365.5	4.516	47.81	2.707	243.7	7.601	40.16
1.8 *m* $DyCl_3$-pH2	365.3	6.151	41.25	4.342	244.5	26.16	42.29

Symbols: $\nu_{1,Dy-O}$ = peak center position of the main stretching vibration of the Dy-O bond; ν_{Dy-Cl} = peak center position of the main stretching vibration of the Dy-Cl bond; $A_{\nu1,Dy-O,\ raw}$ = uncorrected peak area of the Dy-O mode; $A_{\nu1,Dy-O,\ corr.}$ = peak area of the Dy-O mode corrected using the average peak areas of $\nu_{H2O,L-1}$ reported in Table S3; A_{Dy-Cl} = peak area of the Dy-Cl mode. Italic notation: high uncertainty, excluded from evaluation of speciation.

Table S6: Thermodynamic data for solid and aqueous species included in calculations of aqueous Dy speciation.

Solids	$\Delta_f G°$ ($J \cdot mol^{-1}$)	$\Delta_f H°$ ($J \cdot mol^{-1}$)	$S°$ ($J \cdot K^{-1} \cdot mol^{-1}$)	$V°$ ($J \cdot bar^{-1}$)	$C_P°$ ($J \cdot K^{-1} \cdot mol^{-1}$)	a_0 ($J \cdot K^{-1} \cdot mol^{-1}$)	$a_1 \times 10^4$ ($J \cdot K^{-2} \cdot mol^{-1}$)	$a_2 \times 10^{-3}$ ($J \cdot K \cdot mol^{-1}$)	$a_3 \times 10^{-2}$ ($J \cdot mol^{-1}$)	Refs
$Dy(OH)_3(s)$	-1,294,800	-1,428,400	130.3	3.5493	114.05	174.60	122.88	1,137.2	-229.6	(a)

Aqueous species	$\Delta_f G°$ ($J \cdot mol^{-1}$)	$\Delta_f H°$ ($J \cdot mol^{-1}$)	$S°$ ($J \cdot K^{-1} \cdot mol^{-1}$)	$V°$ ($J \cdot bar^{-1}$)	$C_p°$ ($J \cdot K^{-1} \cdot mol^{-1}$)	a_1 ($cal \cdot bar^{-1} \cdot mol^{-1}$)	a_2 ($cal \cdot mol^{-1}$)	a_3 ($cal \cdot K \cdot bar^{-1} \cdot mol^{-1}$)	a_4 ($cal \cdot K \cdot mol^{-1}$)	c_1 ($cal \cdot K^{-1} \cdot mol^{-1}$)	c_2 ($cal \cdot K \cdot mol^{-1}$)	w_0 ($cal \cdot mol^{-1}$)	
Dy^{3+}	-664,001	-696,711	-231.0	-4.1428	-131.61	-0.30003	-1,510.74	11.6879	-21,545	9.5076	-99,419	237,920	(b,c)
$DyCl^{2+}$	799,270	-866,777	-170.8	-1.990	146.0	-0.03106	-853.96	9.1065	-24260	42.1744	40696	170640	(d)
$DyCl_2^+$	-929,057	-1,020,000	-72.51	0.413	67.24	0.26363	-134.44	6.2785	-27234	22.9371	2368	80650	(d)
$DyOH^{2+}$	-856,465	-903,493	-45.6	0.2827	-103.24	0.26154	-139.41	6.2950	-27,213	2.8769	-80,863	122,060	(e)
DyO^+	-809,186	-836,884	19.3	0.5453	-276.36	0.26935	-120.56	6.2255	-27,291	-28.1511	-164,991	47,990	(e)
DyO_2H^0	-996,629	-1,050,602	163.6	2.151	-473.22	0.46969	368.55	4.3051	-29,313	-60.3933	-260,730	-3,000	(e)
HCl^0	-127,240	-179,450	1.76	1.638	149.2	1.61573	-1143.11	-46.1866	-23036.4	46.4716	-52811	0.0	(f)
Cl^-	-131,290	-167,080	56.74	1.734	-122.5	0.40320	480.10	5.5630	-28470	-4.40	-57140	14560	(c)
OH^-	-157,297	-230,024	-10.7	-0.4708	-136.34	0.12527	7.38	1.8423	-27,821	4.15000	-103,460	172,460	(c)
H^+	0	0	0	0	0	0	0	0	0	0	0	-	(c)
H_2O^0	-237183	-285881	69.9	1.8068	75.36	-	-	-	-	-	-	-	(g)

(a) Diakonov et al.[15], C_p estimated from the data for $Tb(OH)_3$; (b) Shock et al.[16]; (c) Shock and Helgeson[4]; (d) Migdisov et al.[17]; (e) Haas et al.[18]; (f) Tagirov et al. [19]; (g) IAPS-84 equation-of-state[7].

Table S7: Aqueous speciation calculations using GEM-Selektor and the composition of the 0.14 mol/kg $DyCl_3$ experimental solution (Table 1). Listed are the calculated pH values at temperature T, the concentrations of aqueous Dy species and the combined concentration of Dy chloride complexes (Dy-Cl) used to plot Figure 8A, and the ratio of Dy chloride species to the Dy^{3+} aqua ion (Dy-Cl/Dy^{3+}) used in Figure 10.

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
25	2.141	$1.270x10^{-1}$	$1.448x10^{-2}$	$4.969x10^{-4}$	$2.045x10^{-8}$	$3.237x10^{-15}$	$5.850x10^{-22}$	$1.498x10^{-2}$	0.1180
30	2.139	$1.274x10^{-1}$	$1.409x10^{-2}$	$5.261x10^{-4}$	$3.388x10^{-8}$	$8.209x10^{-15}$	$2.377x10^{-21}$	$1.462x10^{-2}$	0.1147
35	2.138	$1.276x10^{-1}$	$1.384x10^{-2}$	$5.599x10^{-4}$	$5.506x10^{-8}$	$2.005x10^{-14}$	$9.087x10^{-21}$	$1.440x10^{-2}$	0.1129
40	2.138	$1.277x10^{-1}$	$1.371x10^{-2}$	$5.994x10^{-4}$	$8.803x10^{-8}$	$4.725x10^{-14}$	$3.280x10^{-20}$	$1.431x10^{-2}$	0.1121
45	2.138	$1.277x10^{-1}$	$1.369x10^{-2}$	$6.444x10^{-4}$	$1.382x10^{-7}$	$1.077x10^{-13}$	$1.122x10^{-19}$	$1.434x10^{-2}$	0.1123
50	2.138	$1.275x10^{-1}$	$1.376x10^{-2}$	$6.956x10^{-4}$	$2.133x10^{-7}$	$2.376x10^{-13}$	$3.643x10^{-19}$	$1.446x10^{-2}$	0.1134
55	2.139	$1.273x10^{-1}$	$1.392x10^{-2}$	$7.535x10^{-4}$	$3.244x10^{-7}$	$5.086x10^{-13}$	$1.127x10^{-18}$	$1.467x10^{-2}$	0.1152
60	2.140	$1.270x10^{-1}$	$1.416x10^{-2}$	$8.189x10^{-4}$	$4.856x10^{-7}$	$1.058x10^{-12}$	$3.329x10^{-18}$	$1.498x10^{-2}$	0.1179
65	2.142	$1.266x10^{-1}$	$1.447x10^{-2}$	$8.925x10^{-4}$	$7.167x10^{-7}$	$2.139x10^{-12}$	$9.413x10^{-18}$	$1.537x10^{-2}$	0.1214
70	2.143	$1.262x10^{-1}$	$1.487x10^{-2}$	$9.748x10^{-4}$	$1.043x10^{-6}$	$4.215x10^{-12}$	$2.553x10^{-17}$	$1.584x10^{-2}$	0.1256
75	2.145	$1.256x10^{-1}$	$1.534x10^{-2}$	$1.067x10^{-3}$	$1.498x10^{-6}$	$8.094x10^{-12}$	$6.657x10^{-17}$	$1.640x10^{-2}$	0.1306
80	2.148	$1.250x10^{-1}$	$1.588x10^{-2}$	$1.171x10^{-3}$	$2.124x10^{-6}$	$1.519x10^{-11}$	$1.671x10^{-16}$	$1.705x10^{-2}$	0.1364
85	2.150	$1.242x10^{-1}$	$1.649x10^{-2}$	$1.286x10^{-3}$	$2.975x10^{-6}$	$2.786x10^{-11}$	$4.049x10^{-16}$	$1.778x10^{-2}$	0.1431
90	2.153	$1.234x10^{-1}$	$1.719x10^{-2}$	$1.415x10^{-3}$	$4.120x10^{-6}$	$4.998x10^{-11}$	$9.478x10^{-16}$	$1.861x10^{-2}$	0.1508
95	2.156	$1.225x10^{-1}$	$1.797x10^{-2}$	$1.559x10^{-3}$	$5.640x10^{-6}$	$8.786x10^{-11}$	$2.147x10^{-15}$	$1.952x10^{-2}$	0.1594
100	2.159	$1.214x10^{-1}$	$1.882x10^{-2}$	$1.719x10^{-3}$	$7.636x10^{-6}$	$1.512x10^{-10}$	$4.716x10^{-15}$	$2.054x10^{-2}$	0.1691
105	2.162	$1.203x10^{-1}$	$1.977x10^{-2}$	$1.898x10^{-3}$	$1.023x10^{-5}$	$2.555x10^{-10}$	$1.005x10^{-14}$	$2.166x10^{-2}$	0.1800
110	2.166	$1.191x10^{-1}$	$2.079x10^{-2}$	$2.096x10^{-3}$	$1.357x10^{-5}$	$4.237x10^{-10}$	$2.083x10^{-14}$	$2.289x10^{-2}$	0.1922
115	2.170	$1.177x10^{-1}$	$2.192x10^{-2}$	$2.317x10^{-3}$	$1.783x10^{-5}$	$6.903x10^{-10}$	$4.199x10^{-14}$	$2.423x10^{-2}$	0.2058
120	2.173	$1.163x10^{-1}$	$2.313x10^{-2}$	$2.563x10^{-3}$	$2.318x10^{-5}$	$1.106x10^{-9}$	$8.246x10^{-14}$	$2.570x10^{-2}$	0.2210
125	2.177	$1.147x10^{-1}$	$2.444x10^{-2}$	$2.834x10^{-3}$	$2.988x10^{-5}$	$1.742x10^{-9}$	$1.579x10^{-13}$	$2.728x10^{-2}$	0.2378
130	2.181	$1.130x10^{-1}$	$2.586x10^{-2}$	$3.135x10^{-3}$	$3.816x10^{-5}$	$2.702x10^{-9}$	$2.951x10^{-13}$	$2.899x10^{-2}$	0.2567
135	2.186	$1.111x10^{-1}$	$2.738x10^{-2}$	$3.470x10^{-3}$	$4.827x10^{-5}$	$4.127x10^{-9}$	$5.390x10^{-13}$	$3.085x10^{-2}$	0.2777
140	2.190	$1.091x10^{-1}$	$2.900x10^{-2}$	$3.839x10^{-3}$	$6.064x10^{-5}$	$6.211x10^{-9}$	$9.624x10^{-13}$	$3.284x10^{-2}$	0.3010
145	2.194	$1.069x10^{-1}$	$3.073x10^{-2}$	$4.246x10^{-3}$	$7.548x10^{-5}$	$9.210x10^{-9}$	$1.681x10^{-12}$	$3.498x10^{-2}$	0.3270
150	2.199	$1.046x10^{-1}$	$3.257x10^{-2}$	$4.695x10^{-3}$	$9.316x10^{-5}$	$1.347x10^{-8}$	$2.877x10^{-12}$	$3.726x10^{-2}$	0.3561
155	2.204	$1.022x10^{-1}$	$3.452x10^{-2}$	$5.190x10^{-3}$	$1.140x10^{-4}$	$1.943x10^{-8}$	$4.824x10^{-12}$	$3.971x10^{-2}$	0.3886
160	2.208	$9.956x10^{-2}$	$3.657x10^{-2}$	$5.733x10^{-3}$	$1.385x10^{-4}$	$2.768x10^{-8}$	$7.927x10^{-12}$	$4.231x10^{-2}$	0.4250
165	2.213	$9.678x10^{-2}$	$3.873x10^{-2}$	$6.329x10^{-3}$	$1.668x10^{-4}$	$3.890x10^{-8}$	$1.279x10^{-11}$	$4.506x10^{-2}$	0.4656
170	2.218	$9.383x10^{-2}$	$4.099x10^{-2}$	$6.980x10^{-3}$	$1.994x10^{-4}$	$5.396x10^{-8}$	$2.023x10^{-11}$	$4.797x10^{-2}$	0.5112
175	2.222	$9.074x10^{-2}$	$4.333x10^{-2}$	$7.690x10^{-3}$	$2.364x10^{-4}$	$7.396x10^{-8}$	$3.148x10^{-11}$	$5.102x10^{-2}$	0.5623
180	2.227	$8.750x10^{-2}$	$4.576x10^{-2}$	$8.463x10^{-3}$	$2.780x10^{-4}$	$1.001x10^{-7}$	$4.806x10^{-11}$	$5.422x10^{-2}$	0.6196
185	2.232	$8.412x10^{-2}$	$4.826x10^{-2}$	$9.301x10^{-3}$	$3.244x10^{-4}$	$1.339x10^{-7}$	$7.216x10^{-11}$	$5.756x10^{-2}$	0.6843
190	2.237	$8.059x10^{-2}$	$5.082x10^{-2}$	$1.021x10^{-2}$	$3.756x10^{-4}$	$1.770x10^{-7}$	$1.066x10^{-10}$	$6.103x10^{-2}$	0.7573
195	2.242	$7.694x10^{-2}$	$5.343x10^{-2}$	$1.120x10^{-2}$	$4.316x10^{-4}$	$2.313x10^{-7}$	$1.549x10^{-10}$	$6.462x10^{-2}$	0.8399

Table S7: continued

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
200	2.247	$7.319x10^{-2}$	$5.606x10^{-2}$	$1.226x10^{-2}$	$4.919x10^{-4}$	$2.989x10^{-7}$	$2.217x10^{-10}$	$6.831x10^{-2}$	0.9333
205	2.253	$6.936x10^{-2}$	$5.869x10^{-2}$	$1.340x10^{-2}$	$5.563x10^{-4}$	$3.819x10^{-7}$	$3.125x10^{-10}$	$7.208x10^{-2}$	1.039
210	2.258	$6.545x10^{-2}$	$6.131x10^{-2}$	$1.462x10^{-2}$	$6.244x10^{-4}$	$4.829x10^{-7}$	$4.343x10^{-10}$	$7.593x10^{-2}$	1.160
215	2.264	$6.149x10^{-2}$	$6.388x10^{-2}$	$1.593x10^{-2}$	$6.954x10^{-4}$	$6.042x10^{-7}$	$5.954x10^{-10}$	$7.981x10^{-2}$	1.298
220	2.270	$5.752x10^{-2}$	$6.639x10^{-2}$	$1.732x10^{-2}$	$7.685x10^{-4}$	$7.484x10^{-7}$	$8.057x10^{-10}$	$8.371x10^{-2}$	1.455
225	2.276	$5.354x10^{-2}$	$6.880x10^{-2}$	$1.881x10^{-2}$	$8.429x10^{-4}$	$9.182x10^{-7}$	$1.077x10^{-9}$	$8.762x10^{-2}$	1.636
230	2.284	$4.960x10^{-2}$	$7.109x10^{-2}$	$2.039x10^{-2}$	$9.177x10^{-4}$	$1.116x10^{-6}$	$1.423x10^{-9}$	$9.148x10^{-2}$	1.844
235	2.291	$4.571x10^{-2}$	$7.322x10^{-2}$	$2.207x10^{-2}$	$9.916x10^{-4}$	$1.344x10^{-6}$	$1.861x10^{-9}$	$9.529x10^{-2}$	2.085
240	2.300	$4.192x10^{-2}$	$7.517x10^{-2}$	$2.384x10^{-2}$	$1.064x10^{-3}$	$1.606x10^{-6}$	$2.409x10^{-9}$	$9.901x10^{-2}$	2.362
245	2.309	$3.824x10^{-2}$	$7.692x10^{-2}$	$2.571x10^{-2}$	$1.133x10^{-3}$	$1.902x10^{-6}$	$3.090x10^{-9}$	$1.026x10^{-1}$	2.684
250	2.319	$3.470x10^{-2}$	$7.842x10^{-2}$	$2.768x10^{-2}$	$1.199x10^{-3}$	$2.237x10^{-6}$	$3.932x10^{-9}$	$1.061x10^{-1}$	3.058
255	2.331	$3.133x10^{-2}$	$7.968x10^{-2}$	$2.973x10^{-2}$	$1.259x10^{-3}$	$2.612x10^{-6}$	$4.969x10^{-9}$	$1.094x10^{-1}$	3.492
260	2.344	$2.814x10^{-2}$	$8.066x10^{-2}$	$3.188x10^{-2}$	$1.314x10^{-3}$	$3.029x10^{-6}$	$6.239x10^{-9}$	$1.125x10^{-1}$	3.999
265	2.358	$2.516x10^{-2}$	$8.135x10^{-2}$	$3.412x10^{-2}$	$1.364x10^{-3}$	$3.492x10^{-6}$	$7.795x10^{-9}$	$1.155x10^{-1}$	4.590
270	2.374	$2.239x10^{-2}$	$8.176x10^{-2}$	$3.644x10^{-2}$	$1.406x10^{-3}$	$4.004x10^{-6}$	$9.697x10^{-9}$	$1.182x10^{-1}$	5.279
275	2.393	$1.984x10^{-2}$	$8.188x10^{-2}$	$3.884x10^{-2}$	$1.442x10^{-3}$	$4.569x10^{-6}$	$1.203x10^{-8}$	$1.207x10^{-1}$	6.085
280	2.413	$1.751x10^{-2}$	$8.171x10^{-2}$	$4.130x10^{-2}$	$1.470x10^{-3}$	$5.190x10^{-6}$	$1.489x10^{-8}$	$1.230x10^{-1}$	7.023
285	2.436	$1.542x10^{-2}$	$8.128x10^{-2}$	$4.381x10^{-2}$	$1.492x10^{-3}$	$5.873x10^{-6}$	$1.841x10^{-8}$	$1.251x10^{-1}$	8.113
290	2.461	$1.354x10^{-2}$	$8.059x10^{-2}$	$4.636x10^{-2}$	$1.508x10^{-3}$	$6.624x10^{-6}$	$2.276x10^{-8}$	$1.269x10^{-1}$	9.377
295	2.489	$1.187x10^{-2}$	$7.969x10^{-2}$	$4.892x10^{-2}$	$1.518x10^{-3}$	$7.450x10^{-6}$	$2.816x10^{-8}$	$1.286x10^{-1}$	10.83
300	2.521	$1.041x10^{-2}$	$7.859x10^{-2}$	$5.147x10^{-2}$	$1.522x10^{-3}$	$8.359x10^{-6}$	$3.492x10^{-8}$	$1.301x10^{-1}$	12.50

Table S8: Aqueous speciation calculations using GEM-Selektor and the composition of the 0.27 mol/kg $DyCl_3$ experimental solution (Table 1). Listed are the calculated pH values at temperature T, the concentrations of aqueous Dy species and the combined concentration of Dy chloride complexes (Dy-Cl) used to plot Figure 8B, and the ratio of Dy chloride species to the Dy^{3+} aqua ion (Dy-Cl/Dy^{3+}) used in Figure 10.

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^{+}$	$DyOH^{2+}$	$Dy(OH)_2^{+}$	$Dy(OH)_3^{0}$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
25	2.172	2.337×10^{-1}	3.540×10^{-2}	1.869×10^{-3}	2.775×10^{-8}	3.783×10^{-15}	7.506×10^{-22}	3.727×10^{-2}	0.1594
30	2.167	2.345×10^{-1}	3.452×10^{-2}	1.988×10^{-3}	4.564×10^{-8}	9.450×10^{-15}	2.999×10^{-21}	3.651×10^{-2}	0.1557
35	2.164	2.349×10^{-1}	3.396×10^{-2}	2.123×10^{-3}	7.372×10^{-8}	2.278×10^{-14}	1.130×10^{-20}	3.608×10^{-2}	0.1536
40	2.162	2.351×10^{-1}	3.365×10^{-2}	2.275×10^{-3}	1.170×10^{-7}	5.308×10^{-14}	4.025×10^{-2}	3.592×10^{-2}	0.1528
45	2.161	2.350×10^{-1}	3.356×10^{-2}	2.445×10^{-3}	1.826×10^{-7}	1.196×10^{-13}	1.360×10^{-19}	3.601×10^{-2}	0.1532
50	2.160	2.347×10^{-1}	3.368×10^{-2}	2.634×10^{-3}	2.806×10^{-7}	2.617×10^{-13}	4.374×10^{-19}	3.632×10^{-2}	0.1547
55	2.160	2.342×10^{-1}	3.399×10^{-2}	2.845×10^{-3}	4.244×10^{-7}	5.552×10^{-13}	1.341×10^{-18}	3.683×10^{-2}	0.1573
60	2.161	2.335×10^{-1}	3.446×10^{-2}	3.077×10^{-3}	6.325×10^{-7}	1.147×10^{-12}	3.931×10^{-18}	3.754×10^{-2}	0.1608
65	2.162	2.326×10^{-1}	3.510×10^{-2}	3.336×10^{-3}	9.299×10^{-7}	2.306×10^{-12}	1.104×10^{-17}	3.844×10^{-2}	0.1653
70	2.163	2.315×10^{-1}	3.590×10^{-2}	3.619×10^{-3}	1.348×10^{-6}	4.518×10^{-12}	2.977×10^{-17}	3.952×10^{-2}	0.1707
75	2.166	2.302×10^{-1}	3.685×10^{-2}	3.932×10^{-3}	1.929×10^{-6}	8.638×10^{-12}	7.721×10^{-17}	4.078×10^{-2}	0.1771
80	2.168	2.288×10^{-1}	3.794×10^{-2}	4.276×10^{-3}	2.727×10^{-6}	1.614×10^{-11}	1.930×10^{-16}	4.222×10^{-2}	0.1845
85	2.171	2.272×10^{-1}	3.918×10^{-2}	4.653×10^{-3}	3.808×10^{-6}	2.948×10^{-11}	4.658×10^{-16}	4.384×10^{-2}	0.1930
90	2.175	2.254×10^{-1}	4.057×10^{-2}	5.067×10^{-3}	5.257×10^{-6}	5.271×10^{-11}	1.087×10^{-15}	4.564×10^{-2}	0.2025
95	2.178	2.234×10^{-1}	4.211×10^{-2}	5.519×10^{-3}	7.177×10^{-6}	9.236×10^{-11}	2.457×10^{-15}	4.763×10^{-2}	0.2132
100	2.182	2.212×10^{-1}	4.380×10^{-2}	6.015×10^{-3}	9.695×10^{-6}	1.587×10^{-10}	5.384×10^{-15}	4.981×10^{-2}	0.2252
105	2.187	2.188×10^{-1}	4.563×10^{-2}	6.556×10^{-3}	1.296×10^{-5}	2.676×10^{-10}	1.146×10^{-14}	5.219×10^{-2}	0.2385
110	2.192	2.162×10^{-1}	4.762×10^{-2}	7.147×10^{-3}	1.715×10^{-5}	4.432×10^{-10}	2.372×10^{-14}	5.477×10^{-2}	0.2533
115	2.197	2.134×10^{-1}	4.976×10^{-2}	7.789×10^{-3}	2.246×10^{-5}	7.214×10^{-10}	4.778×10^{-14}	5.755×10^{-2}	0.2696
120	2.202	2.104×10^{-1}	5.206×10^{-2}	8.488×10^{-3}	2.919×10^{-5}	1.155×10^{-9}	9.380×10^{-14}	6.054×10^{-2}	0.2877
125	2.207	2.072×10^{-1}	5.451×10^{-2}	9.248×10^{-3}	3.756×10^{-5}	1.818×10^{-9}	1.797×10^{-13}	6.376×10^{-2}	0.3077
130	2.213	2.038×10^{-1}	5.712×10^{-2}	1.007×10^{-2}	4.789×10^{-5}	2.820×10^{-9}	3.358×10^{-13}	6.719×10^{-2}	0.3298
135	2.219	2.001×10^{-1}	5.989×10^{-2}	1.096×10^{-2}	6.055×10^{-5}	4.307×10^{-9}	6.141×10^{-13}	7.085×10^{-2}	0.3541
140	2.225	1.962×10^{-1}	6.280×10^{-2}	1.193×10^{-2}	7.587×10^{-5}	6.481×10^{-9}	1.097×10^{-12}	7.473×10^{-2}	0.3809
145	2.232	1.921×10^{-1}	6.589×10^{-2}	1.297×10^{-2}	9.433×10^{-5}	9.621×10^{-9}	1.919×10^{-12}	7.885×10^{-2}	0.4106
150	2.238	1.877×10^{-1}	6.911×10^{-2}	1.409×10^{-2}	1.163×10^{-4}	1.408×10^{-8}	3.287×10^{-12}	8.320×10^{-2}	0.4433
155	2.245	1.831×10^{-1}	7.248×10^{-2}	1.529×10^{-2}	1.422×10^{-4}	2.034×10^{-8}	5.519×10^{-12}	8.777×10^{-2}	0.4794
160	2.251	1.783×10^{-1}	7.599×10^{-2}	1.659×10^{-2}	1.726×10^{-4}	2.898×10^{-8}	9.090×10^{-12}	9.258×10^{-2}	0.5193
165	2.258	1.732×10^{-1}	7.962×10^{-2}	1.797×10^{-2}	2.077×10^{-4}	4.078×10^{-8}	1.468×10^{-11}	9.759×10^{-2}	0.5635
170	2.265	1.679×10^{-1}	8.338×10^{-2}	1.946×10^{-2}	2.482×10^{-4}	5.667×10^{-8}	2.329×10^{-11}	1.028×10^{-1}	0.6124
175	2.272	1.624×10^{-1}	8.723×10^{-2}	2.104×10^{-2}	2.942×10^{-4}	7.777×10^{-8}	3.629×10^{-11}	1.083×10^{-1}	0.6665
180	2.280	1.568×10^{-1}	9.118×10^{-2}	2.272×10^{-2}	3.460×10^{-4}	1.055×10^{-7}	5.556×10^{-11}	1.139×10^{-1}	0.7265
185	2.287	1.509×10^{-1}	9.519×10^{-2}	2.450×10^{-2}	4.039×10^{-4}	1.413×10^{-7}	8.362×10^{-11}	1.197×10^{-1}	0.7931
190	2.294	1.449×10^{-1}	9.926×10^{-2}	2.639×10^{-2}	4.680×10^{-4}	1.872×10^{-7}	1.238×10^{-10}	1.256×10^{-1}	0.8672
195	2.302	1.387×10^{-1}	1.033×10^{-1}	2.839×10^{-2}	5.383×10^{-4}	2.452×10^{-7}	1.805×10^{-10}	1.317×10^{-1}	0.9496

Table S8: continued

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
200	2.310	$1.324x10^{-1}$	$1.074x10^{-1}$	$3.050x10^{-2}$	$6.146x10^{-4}$	$3.176x10^{-7}$	$2.592x10^{-10}$	$1.379x10^{-1}$	1.041
205	2.317	$1.261x10^{-1}$	$1.115x10^{-1}$	$3.272x10^{-2}$	$6.965x10^{-4}$	$4.071x10^{-7}$	$3.667x10^{-10}$	$1.442x10^{-1}$	1.144
210	2.326	$1.196x10^{-1}$	$1.155x10^{-1}$	$3.506x10^{-2}$	$7.839x10^{-4}$	$5.165x10^{-7}$	$5.118x10^{-10}$	$1.506x10^{-1}$	1.259
215	2.334	$1.131x10^{-1}$	$1.195x10^{-1}$	$3.751x10^{-2}$	$8.760x10^{-4}$	$6.489x10^{-7}$	$7.049x10^{-10}$	$1.570x10^{-1}$	1.387
220	2.343	$1.067x10^{-1}$	$1.233x10^{-1}$	$4.009x10^{-2}$	$9.721x10^{-4}$	$8.075x10^{-7}$	$9.588x10^{-10}$	$1.634x10^{-1}$	1.532
225	2.353	$1.002x10^{-1}$	$1.270x10^{-1}$	$4.278x10^{-2}$	$1.072x10^{-3}$	$9.957x10^{-7}$	$1.289x10^{-9}$	$1.697x10^{-1}$	1.694
230	2.363	$9.378x10^{-2}$	$1.304x10^{-1}$	$4.559x10^{-2}$	$1.174x10^{-3}$	$1.217x10^{-6}$	$1.715x10^{-9}$	$1.760x10^{-1}$	1.877
235	2.373	$8.747x10^{-2}$	$1.337x10^{-1}$	$4.853x10^{-2}$	$1.277x10^{-3}$	$1.476x10^{-6}$	$2.259x10^{-9}$	$1.822x10^{-1}$	2.083
240	2.385	$8.129x10^{-2}$	$1.367x10^{-1}$	$5.159x10^{-2}$	$1.381x10^{-3}$	$1.776x10^{-6}$	$2.950x10^{-9}$	$1.883x10^{-1}$	2.317
245	2.398	$7.526x10^{-2}$	$1.395x10^{-1}$	$5.477x10^{-2}$	$1.484x10^{-3}$	$2.123x10^{-6}$	$3.820x10^{-9}$	$1.943x10^{-1}$	2.581
250	2.411	$6.943x10^{-2}$	$1.419x10^{-1}$	$5.807x10^{-2}$	$1.586x10^{-3}$	$2.519x10^{-6}$	$4.912x10^{-9}$	$2.000x10^{-1}$	2.880
255	2.426	$6.383x10^{-2}$	$1.440x10^{-1}$	$6.148x10^{-2}$	$1.685x10^{-3}$	$2.972x10^{-6}$	$6.278x10^{-9}$	$2.055x10^{-1}$	3.219
260	2.443	$5.849x10^{-2}$	$1.457x10^{-1}$	$6.499x10^{-2}$	$1.781x10^{-3}$	$3.486x10^{-6}$	$7.981x10^{-9}$	$2.107x10^{-1}$	3.603
265	2.461	$5.342x10^{-2}$	$1.471x10^{-1}$	$6.861x10^{-2}$	$1.873x10^{-3}$	$4.068x10^{-6}$	$1.010x10^{-8}$	$2.157x10^{-1}$	4.038
270	2.480	$4.867x10^{-2}$	$1.481x10^{-1}$	$7.230x10^{-2}$	$1.960x10^{-3}$	$4.725x10^{-6}$	$1.275x10^{-8}$	$2.204x10^{-1}$	4.528
275	2.502	$4.423x10^{-2}$	$1.487x10^{-1}$	$7.606x10^{-2}$	$2.042x10^{-3}$	$5.466x10^{-6}$	$1.605x10^{-8}$	$2.247x10^{-1}$	5.080
280	2.526	$4.014x10^{-2}$	$1.489x10^{-1}$	$7.984x10^{-2}$	$2.119x10^{-3}$	$6.298x10^{-6}$	$2.016x10^{-8}$	$2.287x10^{-1}$	5.698
285	2.553	$3.640x10^{-2}$	$1.488x10^{-1}$	$8.363x10^{-2}$	$2.190x10^{-3}$	$7.232x10^{-6}$	$2.532x10^{-8}$	$2.324x10^{-1}$	6.384
290	2.582	$3.302x10^{-2}$	$1.483x10^{-1}$	$8.739x10^{-2}$	$2.256x10^{-3}$	$8.280x10^{-6}$	$3.178x10^{-8}$	$2.357x10^{-1}$	7.139
295	2.614	$3.000x10^{-2}$	$1.476x10^{-1}$	$9.104x10^{-2}$	$2.317x10^{-3}$	$9.450x10^{-6}$	$3.991x10^{-8}$	$2.387x10^{-1}$	7.957
300	2.649	$2.734x10^{-2}$	$1.467x10^{-1}$	$9.454x10^{-2}$	$2.374x10^{-3}$	$1.076x10^{-5}$	$5.014x10^{-8}$	$2.413x10^{-1}$	8.825

Table S9: Aqueous speciation calculations using GEM-Selektor and the composition of the 0.59 mol/kg $DyCl_3$ experimental solution (Table 1). Listed are the calculated pH values at temperature T, the concentrations of aqueous Dy species and the combined concentration of Dy chloride complexes (Dy-Cl) used to plot Figure 8C, and the ratio of Dy chloride species to the Dy^{3+} aqua ion (Dy-Cl/Dy^{3+}) used in Figure 10.

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
25	2.172	$4.649x10^{-1}$	$1.162x10^{-1}$	$1.190x10^{-2}$	$3.608x10^{-8}$	$3.864x10^{-15}$	$8.819x10^{-22}$	$1.281x10^{-1}$	0.2756
30	2.162	$4.660x10^{-1}$	$1.141x10^{-1}$	$1.285x10^{-2}$	$5.842x10^{-8}$	$9.371x10^{-15}$	$3.402x10^{-21}$	$1.270x10^{-1}$	0.2725
35	2.154	$4.664x10^{-1}$	$1.127x10^{-1}$	$1.386x10^{-2}$	$9.302x10^{-8}$	$2.199x10^{-14}$	$1.243x10^{-20}$	$1.266x10^{-1}$	0.2714
40	2.147	$4.661x10^{-1}$	$1.119x10^{-1}$	$1.495x10^{-2}$	$1.457x10^{-7}$	$4.998x10^{-14}$	$4.309x10^{-20}$	$1.269x10^{-1}$	0.2722
45	2.142	$4.652x10^{-1}$	$1.117x10^{-1}$	$1.611x10^{-2}$	$2.248x10^{-7}$	$1.105x10^{-13}$	$1.423x10^{-19}$	$1.278x10^{-1}$	0.2747
50	2.139	$4.637x10^{-1}$	$1.119x10^{-1}$	$1.736x10^{-2}$	$3.418x10^{-7}$	$2.373x10^{-13}$	$4.480x10^{-19}$	$1.293x10^{-1}$	0.2788
55	2.137	$4.617x10^{-1}$	$1.126x10^{-1}$	$1.869x10^{-2}$	$5.125x10^{-7}$	$4.965x10^{-13}$	$1.350x10^{-18}$	$1.313x10^{-1}$	0.2843
60	2.136	$4.592x10^{-1}$	$1.137x10^{-1}$	$2.010x10^{-2}$	$7.578x10^{-7}$	$1.012x10^{-12}$	$3.898x10^{-18}$	$1.338x10^{-1}$	0.2913
65	2.136	$4.563x10^{-1}$	$1.151x10^{-1}$	$2.160x10^{-2}$	$1.105x10^{-6}$	$2.012x10^{-12}$	$1.081x10^{-17}$	$1.367x10^{-1}$	0.2996
70	2.138	$4.529x10^{-1}$	$1.169x10^{-1}$	$2.321x10^{-2}$	$1.594x10^{-6}$	$3.909x10^{-12}$	$2.884x10^{-17}$	$1.401x10^{-1}$	0.3093
75	2.140	$4.491x10^{-1}$	$1.190x10^{-1}$	$2.490x10^{-2}$	$2.270x10^{-6}$	$7.425x10^{-12}$	$7.423x10^{-17}$	$1.439x10^{-1}$	0.3204
80	2.143	$4.449x10^{-1}$	$1.214x10^{-1}$	$2.670x10^{-2}$	$3.193x10^{-6}$	$1.380x10^{-11}$	$1.845x10^{-16}$	$1.481x10^{-1}$	0.3330
85	2.146	$4.403x10^{-1}$	$1.241x10^{-1}$	$2.860x10^{-2}$	$4.442x10^{-6}$	$2.512x10^{-11}$	$4.434x10^{-16}$	$1.527x10^{-1}$	0.3469
90	2.151	$4.352x10^{-1}$	$1.271x10^{-1}$	$3.061x10^{-2}$	$6.112x10^{-6}$	$4.483x10^{-11}$	$1.032x10^{-15}$	$1.577x10^{-1}$	0.3624
95	2.156	$4.299x10^{-1}$	$1.304x10^{-1}$	$3.272x10^{-2}$	$8.319x10^{-6}$	$7.848x10^{-11}$	$2.330x10^{-15}$	$1.631x10^{-1}$	0.3795
100	2.162	$4.241x10^{-1}$	$1.339x10^{-1}$	$3.494x10^{-2}$	$1.121x10^{-5}$	$1.348x10^{-10}$	$5.108x10^{-15}$	$1.689x10^{-1}$	0.3982
105	2.169	$4.180x10^{-1}$	$1.377x10^{-1}$	$3.728x10^{-2}$	$1.498x10^{-5}$	$2.279x10^{-10}$	$1.090x10^{-14}$	$1.750x10^{-1}$	0.4187
110	2.176	$4.115x10^{-1}$	$1.418x10^{-1}$	$3.973x10^{-2}$	$1.979x10^{-5}$	$3.783x10^{-10}$	$2.259x10^{-14}$	$1.815x10^{-1}$	0.4411
115	2.183	$4.046x10^{-1}$	$1.460x10^{-1}$	$4.229x10^{-2}$	$2.592x10^{-5}$	$6.177x10^{-10}$	$4.571x10^{-14}$	$1.883x10^{-1}$	0.4654
120	2.192	$3.975x10^{-1}$	$1.505x10^{-1}$	$4.497x10^{-2}$	$3.365x10^{-5}$	$9.932x10^{-10}$	$9.020x10^{-14}$	$1.955x10^{-1}$	0.4919
125	2.200	$3.899x10^{-1}$	$1.552x10^{-1}$	$4.777x10^{-2}$	$4.332x10^{-5}$	$1.572x10^{-9}$	$1.737x10^{-13}$	$2.030x10^{-1}$	0.5206
130	2.209	$3.821x10^{-1}$	$1.602x10^{-1}$	$5.068x10^{-2}$	$5.521x10^{-5}$	$2.452x10^{-9}$	$3.270x10^{-13}$	$2.108x10^{-1}$	0.5518
135	2.219	$3.739x10^{-1}$	$1.653x10^{-1}$	$5.372x10^{-2}$	$6.986x10^{-5}$	$3.769x10^{-9}$	$6.022x10^{-13}$	$2.190x10^{-1}$	0.5857
140	2.228	$3.655x10^{-1}$	$1.706x10^{-1}$	$5.687x10^{-2}$	$8.773x10^{-5}$	$5.711x10^{-9}$	$1.085x10^{-12}$	$2.274x10^{-1}$	0.6224
145	2.239	$3.567x10^{-1}$	$1.760x10^{-1}$	$6.014x10^{-2}$	$1.092x10^{-4}$	$8.540x10^{-9}$	$1.914x10^{-12}$	$2.362x10^{-1}$	0.6621
150	2.249	$3.477x10^{-1}$	$1.817x10^{-1}$	$6.353x10^{-2}$	$1.349x10^{-4}$	$1.260x10^{-8}$	$3.311x10^{-12}$	$2.452x10^{-1}$	0.7053
155	2.260	$3.384x10^{-1}$	$1.874x10^{-1}$	$6.704x10^{-2}$	$1.654x10^{-4}$	$1.835x10^{-8}$	$5.614x10^{-12}$	$2.545x10^{-1}$	0.7521
160	2.271	$3.288x10^{-1}$	$1.933x10^{-1}$	$7.066x10^{-2}$	$2.012x10^{-4}$	$2.639x10^{-8}$	$9.340x10^{-12}$	$2.640x10^{-1}$	0.8029
165	2.282	$3.190x10^{-1}$	$1.993x10^{-1}$	$7.439x10^{-2}$	$2.429x10^{-4}$	$3.748x10^{-8}$	$1.525x10^{-11}$	$2.737x10^{-1}$	0.8579
170	2.293	$3.091x10^{-1}$	$2.054x10^{-1}$	$7.824x10^{-2}$	$2.911x10^{-4}$	$5.257x10^{-8}$	$2.446x10^{-11}$	$2.836x10^{-1}$	0.9176
175	2.305	$2.990x10^{-1}$	$2.115x10^{-1}$	$8.219x10^{-2}$	$3.462x10^{-4}$	$7.285x10^{-8}$	$3.853x10^{-11}$	$2.937x10^{-1}$	0.9824
180	2.316	$2.887x10^{-1}$	$2.177x10^{-1}$	$8.624x10^{-2}$	$4.089x10^{-4}$	$9.977x10^{-8}$	$5.968x10^{-11}$	$3.039x10^{-1}$	1.053
185	2.328	$2.783x10^{-1}$	$2.238x10^{-1}$	$9.039x10^{-2}$	$4.795x10^{-4}$	$1.351x10^{-7}$	$9.090x10^{-11}$	$3.142x10^{-1}$	1.129
190	2.340	$2.678x10^{-1}$	$2.300x10^{-1}$	$9.464x10^{-2}$	$5.583x10^{-4}$	$1.808x10^{-7}$	$1.362x10^{-10}$	$3.246x10^{-1}$	1.212
195	2.352	$2.573x10^{-1}$	$2.361x10^{-1}$	$9.898x10^{-2}$	$6.455x10^{-4}$	$2.394x10^{-7}$	$2.010x10^{-10}$	$3.350x10^{-1}$	1.302

Table S9: continued

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
200	2.365	$2.468x10^{-1}$	$2.421x10^{-1}$	$1.034x10^{-1}$	$7.413x10^{-4}$	$3.135x10^{-7}$	$2.922x10^{-10}$	$3.455x10^{-1}$	1.400
205	2.378	$2.362x10^{-1}$	$2.480x10^{-1}$	$1.079x10^{-1}$	$8.458x10^{-4}$	$4.064x10^{-7}$	$4.187x10^{-10}$	$3.559x10^{-1}$	1.507
210	2.391	$2.257x10^{-1}$	$2.538x10^{-1}$	$1.125x10^{-1}$	$9.586x10^{-4}$	$5.216x10^{-7}$	$5.917x10^{-10}$	$3.663x10^{-1}$	1.623
215	2.404	$2.153x10^{-1}$	$2.594x10^{-1}$	$1.172x10^{-1}$	$1.080x10^{-3}$	$6.630x10^{-7}$	$8.255x10^{-10}$	$3.766x10^{-1}$	1.749
220	2.418	$2.050x10^{-1}$	$2.648x10^{-1}$	$1.220x10^{-1}$	$1.209x10^{-3}$	$8.351x10^{-7}$	$1.138x10^{-9}$	$3.868x10^{-1}$	1.886
225	2.432	$1.949x10^{-1}$	$2.700x10^{-1}$	$1.268x10^{-1}$	$1.345x10^{-3}$	$1.043x10^{-6}$	$1.550x10^{-9}$	$3.968x10^{-1}$	2.036
230	2.447	$1.849x10^{-1}$	$2.749x10^{-1}$	$1.317x10^{-1}$	$1.488x10^{-3}$	$1.291x10^{-6}$	$2.090x10^{-9}$	$4.066x10^{-1}$	2.199
235	2.463	$1.751x10^{-1}$	$2.795x10^{-1}$	$1.367x10^{-1}$	$1.638x10^{-3}$	$1.587x10^{-6}$	$2.790x10^{-9}$	$4.162x10^{-1}$	2.376
240	2.479	$1.656x10^{-1}$	$2.838x10^{-1}$	$1.417x10^{-1}$	$1.793x10^{-3}$	$1.935x10^{-6}$	$3.691x10^{-9}$	$4.256x10^{-1}$	2.569
245	2.497	$1.564x10^{-1}$	$2.878x10^{-1}$	$1.468x10^{-1}$	$1.953x10^{-3}$	$2.345x10^{-6}$	$4.845x10^{-9}$	$4.346x10^{-1}$	2.779
250	2.515	$1.475x10^{-1}$	$2.914x10^{-1}$	$1.520x10^{-1}$	$2.118x10^{-3}$	$2.823x10^{-6}$	$6.312x10^{-9}$	$4.434x10^{-1}$	3.005
255	2.535	$1.390x10^{-1}$	$2.946x10^{-1}$	$1.571x10^{-1}$	$2.286x10^{-3}$	$3.378x10^{-6}$	$8.170x10^{-9}$	$4.517x10^{-1}$	3.250
260	2.556	$1.308x10^{-1}$	$2.974x10^{-1}$	$1.623x10^{-1}$	$2.456x10^{-3}$	$4.019x10^{-6}$	$1.051x10^{-8}$	$4.597x10^{-1}$	3.513
265	2.578	$1.231x10^{-1}$	$2.999x10^{-1}$	$1.674x10^{-1}$	$2.629x10^{-3}$	$4.758x10^{-6}$	$1.346x10^{-8}$	$4.672x10^{-1}$	3.794
270	2.602	$1.159x10^{-1}$	$3.019x10^{-1}$	$1.724x10^{-1}$	$2.804x10^{-3}$	$5.606x10^{-6}$	$1.715x10^{-8}$	$4.743x10^{-1}$	4.091
275	2.628	$1.092x10^{-1}$	$3.036x10^{-1}$	$1.772x10^{-1}$	$2.980x10^{-3}$	$6.573x10^{-6}$	$2.177x10^{-8}$	$4.808x10^{-1}$	4.402
280	2.656	$1.031x10^{-1}$	$3.049x10^{-1}$	$1.819x10^{-1}$	$3.158x10^{-3}$	$7.673x10^{-6}$	$2.753x10^{-8}$	$4.867x10^{-1}$	4.721
285	2.685	$9.759x10^{-2}$	$3.059x10^{-1}$	$1.862x10^{-1}$	$3.336x10^{-3}$	$8.917x10^{-6}$	$3.470x10^{-8}$	$4.921x10^{-1}$	5.042
290	2.717	$9.278x10^{-2}$	$3.067x10^{-1}$	$1.900x10^{-1}$	$3.515x10^{-3}$	$1.032x10^{-5}$	$4.358x10^{-8}$	$4.967x10^{-1}$	5.354
295	2.752	$8.874x10^{-2}$	$3.073x10^{-1}$	$1.932x10^{-1}$	$3.696x10^{-3}$	$1.187x10^{-5}$	$5.451x10^{-8}$	$5.006x10^{-1}$	5.641
300	2.789	$8.557x10^{-2}$	$3.079x10^{-1}$	$1.956x10^{-1}$	$3.878x10^{-3}$	$1.360x10^{-5}$	$6.789x10^{-8}$	$5.035x10^{-1}$	5.884

Table S10: Aqueous speciation calculations using GEM-Selektor and the composition of the 1.8 mol/kg $DyCl_3$ experimental solution (Table 1). Listed are the calculated pH values at temperature T, the concentrations of aqueous Dy species and the combined concentration of Dy chloride complexes (Dy-Cl) used to plot Figure 8D, and the ratio of Dy chloride species to the Dy^{3+} aqua ion (Dy-Cl/Dy^{3+}) used in Figure 10.

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
25	2.235	$8.877x10^{-1}$	$6.618x10^{-1}$	$2.385x10^{-1}$	$4.731x10^{-8}$	$4.579x10^{-15}$	$1.910x10^{-21}$	$9.003x10^{-1}$	1.014
30	2.216	$8.790x10^{-1}$	$6.492x10^{-1}$	$2.598x10^{-1}$	$7.411x10^{-8}$	$1.052x10^{-14}$	$6.950x10^{-21}$	$9.090x10^{-1}$	1.034
35	2.198	$8.695x10^{-1}$	$6.379x10^{-1}$	$2.805x10^{-1}$	$1.143x10^{-7}$	$2.343x10^{-14}$	$2.386x10^{-20}$	$9.185x10^{-1}$	1.056
40	2.183	$8.593x10^{-1}$	$6.281x10^{-1}$	$3.006x10^{-1}$	$1.738x10^{-7}$	$5.070x10^{-14}$	$7.769x10^{-20}$	$9.287x10^{-1}$	1.081
45	2.170	$8.485x10^{-1}$	$6.195x10^{-1}$	$3.199x10^{-1}$	$2.606x10^{-7}$	$1.069x10^{-13}$	$2.410x10^{-19}$	$9.395x10^{-1}$	1.107
50	2.159	$8.372x10^{-1}$	$6.123x10^{-1}$	$3.385x10^{-1}$	$3.857x10^{-7}$	$2.197x10^{-13}$	$7.150x10^{-19}$	$9.508x10^{-1}$	1.136
55	2.150	$8.253x10^{-1}$	$6.062x10^{-1}$	$3.565x10^{-1}$	$5.639x10^{-7}$	$4.411x10^{-13}$	$2.034x10^{-18}$	$9.627x10^{-1}$	1.166
60	2.143	$8.130x10^{-1}$	$6.013x10^{-1}$	$3.738x10^{-1}$	$8.149x10^{-7}$	$8.663x10^{-13}$	$5.568x10^{-18}$	$9.750x10^{-1}$	1.199
65	2.138	$8.002x10^{-1}$	$5.974x10^{-1}$	$3.904x10^{-1}$	$1.165x10^{-6}$	$1.666x10^{-12}$	$1.469x10^{-17}$	$9.878x10^{-1}$	1.234
70	2.134	$7.871x10^{-1}$	$5.945x10^{-1}$	$4.064x10^{-1}$	$1.647x10^{-6}$	$3.141x10^{-12}$	$3.746x10^{-17}$	1.001	1.272
75	2.133	$7.737x10^{-1}$	$5.925x10^{-1}$	$4.218x10^{-1}$	$2.304x10^{-6}$	$5.811x10^{-12}$	$9.244x10^{-17}$	1.014	1.311
80	2.133	$7.600x10^{-1}$	$5.914x10^{-1}$	$4.366x10^{-1}$	$3.193x10^{-6}$	$1.056x10^{-11}$	$2.212x10^{-16}$	1.028	1.353
85	2.134	$7.460x10^{-1}$	$5.911x10^{-1}$	$4.509x10^{-1}$	$4.382x10^{-6}$	$1.885x10^{-11}$	$5.142x10^{-16}$	1.042	1.397
90	2.137	$7.318x10^{-1}$	$5.915x10^{-1}$	$4.646x10^{-1}$	$5.960x10^{-6}$	$3.309x10^{-11}$	$1.162x10^{-15}$	1.056	1.443
95	2.142	$7.174x10^{-1}$	$5.927x10^{-1}$	$4.779x10^{-1}$	$8.034x10^{-6}$	$5.716x10^{-11}$	$2.558x10^{-15}$	1.071	1.492
100	2.148	$7.028x10^{-1}$	$5.945x10^{-1}$	$4.907x10^{-1}$	$1.074x10^{-5}$	$9.724x10^{-11}$	$5.489x10^{-15}$	1.085	1.544
105	2.155	$6.881x10^{-1}$	$5.969x10^{-1}$	$5.030x10^{-1}$	$1.423x10^{-5}$	$1.629x10^{-10}$	$1.149x10^{-14}$	1.100	1.599
110	2.163	$6.732x10^{-1}$	$5.999x10^{-1}$	$5.148x10^{-1}$	$1.871x10^{-5}$	$2.691x10^{-10}$	$2.351x10^{-14}$	1.115	1.656
115	2.172	$6.582x10^{-1}$	$6.034x10^{-1}$	$5.263x10^{-1}$	$2.440x10^{-5}$	$4.384x10^{-10}$	$4.703x10^{-14}$	1.130	1.716
120	2.182	$6.432x10^{-1}$	$6.074x10^{-1}$	$5.374x10^{-1}$	$3.158x10^{-5}$	$7.045x10^{-10}$	$9.205x10^{-14}$	1.145	1.780
125	2.193	$6.281x10^{-1}$	$6.119x10^{-1}$	$5.480x10^{-1}$	$4.058x10^{-5}$	$1.117x10^{-9}$	$1.764x10^{-13}$	1.160	1.847
130	2.205	$6.129x10^{-1}$	$6.167x10^{-1}$	$5.583x10^{-1}$	$5.176x10^{-5}$	$1.749x10^{-9}$	$3.315x10^{-13}$	1.175	1.917
135	2.218	$5.978x10^{-1}$	$6.219x10^{-1}$	$5.683x10^{-1}$	$6.554x10^{-5}$	$2.705x10^{-9}$	$6.107x10^{-13}$	1.190	1.991
140	2.232	$5.826x10^{-1}$	$6.275x10^{-1}$	$5.778x10^{-1}$	$8.244x10^{-5}$	$4.133x10^{-9}$	$1.104x10^{-12}$	1.205	2.069
145	2.246	$5.675x10^{-1}$	$6.333x10^{-1}$	$5.871x10^{-1}$	$1.030x10^{-4}$	$6.240x10^{-9}$	$1.960x10^{-12}$	1.220	2.151
150	2.262	$5.524x10^{-1}$	$6.394x10^{-1}$	$5.960x10^{-1}$	$1.278x10^{-4}$	$9.312x10^{-9}$	$3.417x10^{-12}$	1.235	2.236
155	2.277	$5.375x10^{-1}$	$6.458x10^{-1}$	$6.045x10^{-1}$	$1.575x10^{-4}$	$1.374x10^{-8}$	$5.856x10^{-12}$	1.250	2.326
160	2.294	$5.227x10^{-1}$	$6.524x10^{-1}$	$6.128x10^{-1}$	$1.929x10^{-4}$	$2.005x10^{-8}$	$9.867x10^{-12}$	1.265	2.421
165	2.311	$5.080x10^{-1}$	$6.591x10^{-1}$	$6.207x10^{-1}$	$2.348x10^{-4}$	$2.894x10^{-8}$	$1.635x10^{-11}$	1.280	2.519
170	2.328	$4.935x10^{-1}$	$6.660x10^{-1}$	$6.282x10^{-1}$	$2.840x10^{-4}$	$4.132x10^{-8}$	$2.666x10^{-11}$	1.294	2.622
175	2.346	$4.793x10^{-1}$	$6.730x10^{-1}$	$6.354x10^{-1}$	$3.413x10^{-4}$	$5.836x10^{-8}$	$4.279x10^{-11}$	1.308	2.730
180	2.364	$4.653x10^{-1}$	$6.801x10^{-1}$	$6.422x10^{-1}$	$4.078x10^{-4}$	$8.159x10^{-8}$	$6.762x10^{-11}$	1.322	2.842
185	2.383	$4.516x10^{-1}$	$6.873x10^{-1}$	$6.486x10^{-1}$	$4.844x10^{-4}$	$1.129x10^{-7}$	$1.052x10^{-10}$	1.336	2.959
190	2.402	$4.382x10^{-1}$	$6.946x10^{-1}$	$6.547x10^{-1}$	$5.715x10^{-4}$	$1.546x10^{-7}$	$1.613x10^{-10}$	1.349	3.079
195	2.422	$4.252x10^{-1}$	$7.018x10^{-1}$	$6.603x10^{-1}$	$6.712x10^{-4}$	$2.096x10^{-7}$	$2.437x10^{-10}$	1.362	3.201

Table S10: continued

T	pH	Dy^{3+}	$DyCl^{2+}$	$DyCl_2^+$	$DyOH^{2+}$	$Dy(OH)_2^+$	$Dy(OH)_3^0$	Dy-Cl complexes	Dy-Cl/ Dy^{3+}
°C		mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	mol/kg	
200	2.441	$4.126x10^{-1}$	$7.091x10^{-1}$	$6.655x10^{-1}$	$7.835x10^{-4}$	$2.813x10^{-7}$	$3.630x10^{-10}$	1.375	3.332
205	2.461	$4.004x10^{-1}$	$7.164x10^{-1}$	$6.703x10^{-1}$	$9.095x10^{-4}$	$3.741x10^{-7}$	$5.332x10^{-10}$	1.387	3.463
210	2.482	$3.887x10^{-1}$	$7.237x10^{-1}$	$6.746x10^{-1}$	$1.050x10^{-3}$	$4.928x10^{-7}$	$7.724x10^{-10}$	1.398	3.597
215	2.502	$3.775x10^{-1}$	$7.309x10^{-1}$	$6.784x10^{-1}$	$1.206x10^{-3}$	$6.433x10^{-7}$	$1.104x10^{-9}$	1.409	3.733
220	2.523	$3.669x10^{-1}$	$7.381x10^{-1}$	$6.816x10^{-1}$	$1.379x10^{-3}$	$8.321x10^{-7}$	$1.558x10^{-9}$	1.420	3.870
225	2.545	$3.569x10^{-1}$	$7.453x10^{-1}$	$6.843x10^{-1}$	$1.568x10^{-3}$	$1.067x10^{-6}$	$2.172x10^{-9}$	1.430	4.006
230	2.567	$3.475x10^{-1}$	$7.524x10^{-1}$	$6.864x10^{-1}$	$1.776x10^{-3}$	$1.357x10^{-6}$	$2.990x10^{-9}$	1.439	4.141
235	2.589	$3.388x10^{-1}$	$7.595x10^{-1}$	$6.877x10^{-1}$	$2.002x10^{-3}$	$1.712x10^{-6}$	$4.067x10^{-9}$	1.447	4.271
240	2.611	$3.309x10^{-1}$	$7.665x10^{-1}$	$6.883x10^{-1}$	$2.248x10^{-3}$	$2.142x10^{-6}$	$5.470x10^{-9}$	1.455	4.396
245	2.634	$3.239x10^{-1}$	$7.735x10^{-1}$	$6.881x10^{-1}$	$2.514x10^{-3}$	$2.660x10^{-6}$	$7.273x10^{-9}$	1.462	4.513
250	2.658	$3.178x10^{-1}$	$7.805x10^{-1}$	$6.868x10^{-1}$	$2.801x10^{-3}$	$3.278x10^{-6}$	$9.563x10^{-9}$	1.467	4.617
255	2.682	$3.128x10^{-1}$	$7.876x10^{-1}$	$6.844x10^{-1}$	$3.110x10^{-3}$	$4.011x10^{-6}$	$1.244x10^{-8}$	1.472	4.706
260	2.707	$3.091x10^{-1}$	$7.947x10^{-1}$	$6.807x10^{-1}$	$3.442x10^{-3}$	$4.872x10^{-6}$	$1.600x10^{-8}$	1.475	4.773
265	2.732	$3.068x10^{-1}$	$8.020x10^{-1}$	$6.754x10^{-1}$	$3.799x10^{-3}$	$5.875x10^{-6}$	$2.035x10^{-8}$	1.477	4.815
270	2.758	$3.063x10^{-1}$	$8.094x10^{-1}$	$6.681x10^{-1}$	$4.182x10^{-3}$	$7.032x10^{-6}$	$2.560x10^{-8}$	1.478	4.824
275	2.784	$3.078x10^{-1}$	$8.171x10^{-1}$	$6.585x10^{-1}$	$4.593x10^{-3}$	$8.355x10^{-6}$	$3.181x10^{-8}$	1.476	4.794
280	2.812	$3.120x10^{-1}$	$8.250x10^{-1}$	$6.460x10^{-1}$	$5.034x10^{-3}$	$9.850x10^{-6}$	$3.903x10^{-8}$	1.471	4.715
285	2.840	$3.194x10^{-1}$	$8.332x10^{-1}$	$6.299x10^{-1}$	$5.508x10^{-3}$	$1.152x10^{-5}$	$4.721x10^{-8}$	1.463	4.581
290	2.869	$3.311x10^{-1}$	$8.414x10^{-1}$	$6.095x10^{-1}$	$6.018x10^{-3}$	$1.335x10^{-5}$	$5.621x10^{-8}$	1.451	4.382
295	2.898	$3.485x10^{-1}$	$8.495x10^{-1}$	$5.834x10^{-1}$	$6.569x10^{-3}$	$1.531x10^{-5}$	$6.569x10^{-8}$	1.433	4.112
300	2.929	$3.736x10^{-1}$	$8.568x10^{-1}$	$5.504x10^{-1}$	$7.168x10^{-3}$	$1.737x10^{-5}$	$7.509x10^{-8}$	1.407	3.766

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