Supporting Information

$C_2O_4^{2-}$-templated cage-shaped Ln_{28} (Ln=Gd, Eu) nanoclusters with magnetocaloric effect and luminescence

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Experimental Section

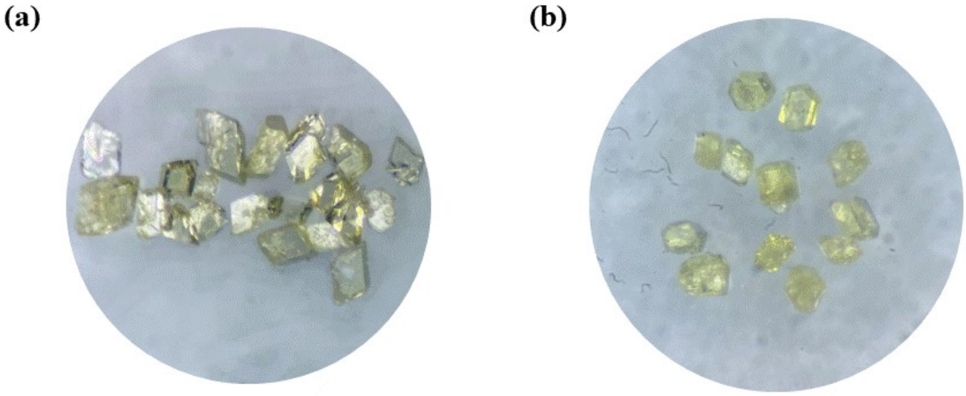


Fig. S1 The images of (a) **Gd_{28}**, (b) **Eu_{28}** under optical microscope.

Structure

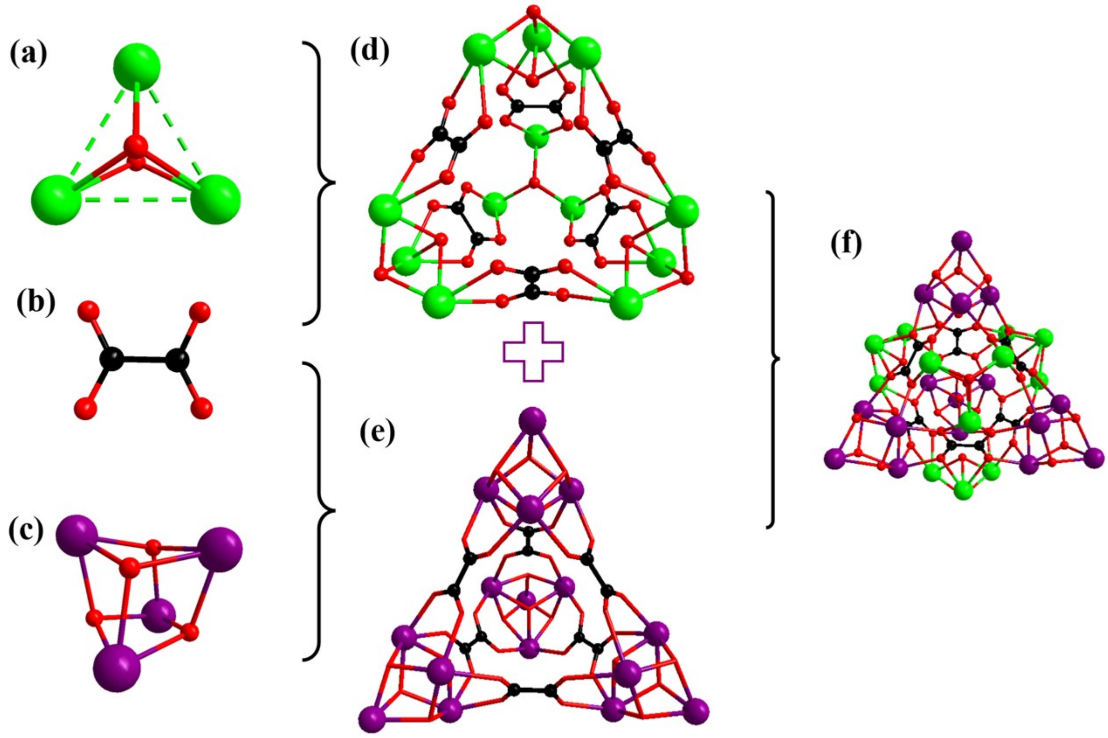


Fig. S2 Ball-and-stick view of (a) $[Gd_3(\mu_3\text{-}OH)_2]^{7+}$ unit; (b) $C_2O_4^{2-}$ anion; (c) $[Gd_4(\mu_3\text{-}OH)_4]^{8+}$ unit; (d) $[Gd_{12}(\mu_3\text{-}OH)_8(C_2O_4)_6]^{16+}$ subunit; (e) $[Gd_{16}(\mu_3\text{-}OH)_{16}(C_2O_4)_6]^{20+}$ subunit; (f) $[Gd_{28}(C_2O_4)_6(\mu_3\text{-}OH)_{36}(\mu_2\text{-}OH)_2]^{34+}$.

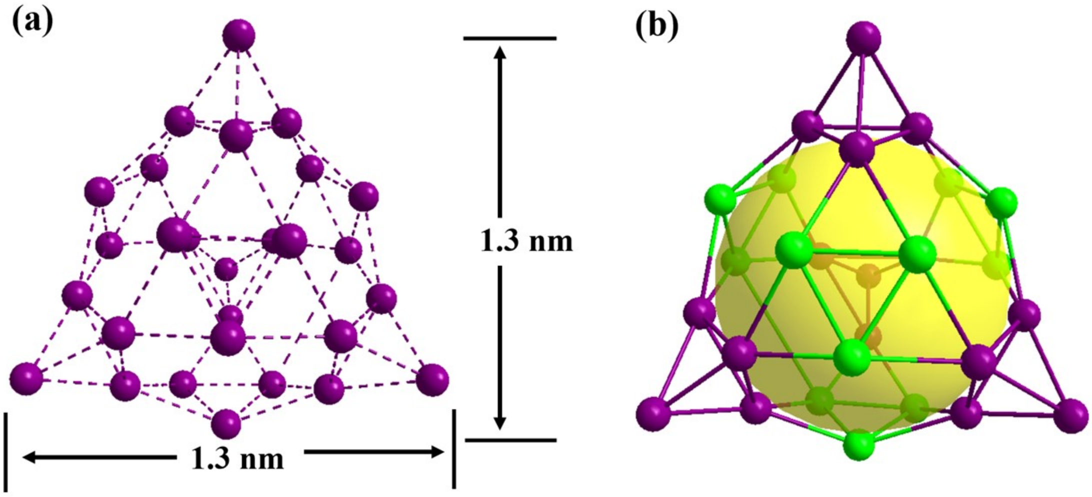


Fig. S3 The metal skeleton of $\mathbf{Gd_{28}}$.

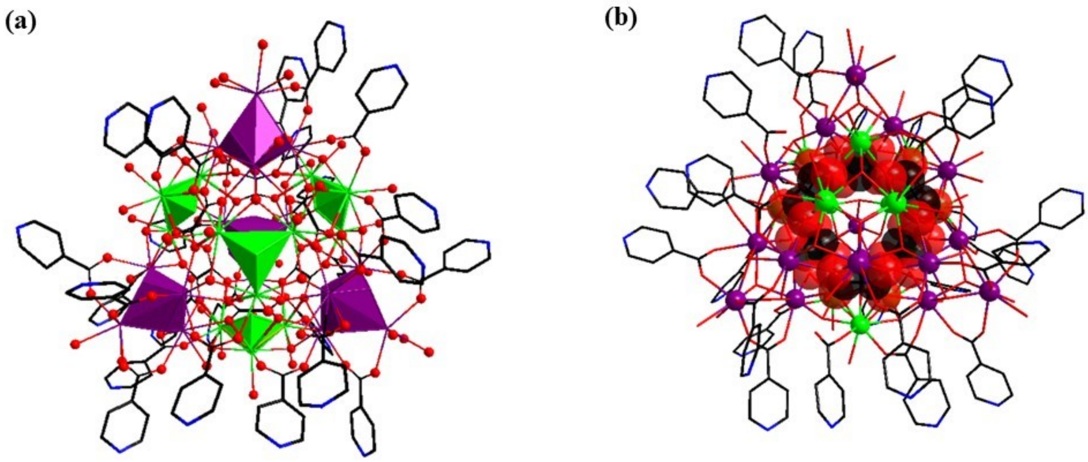
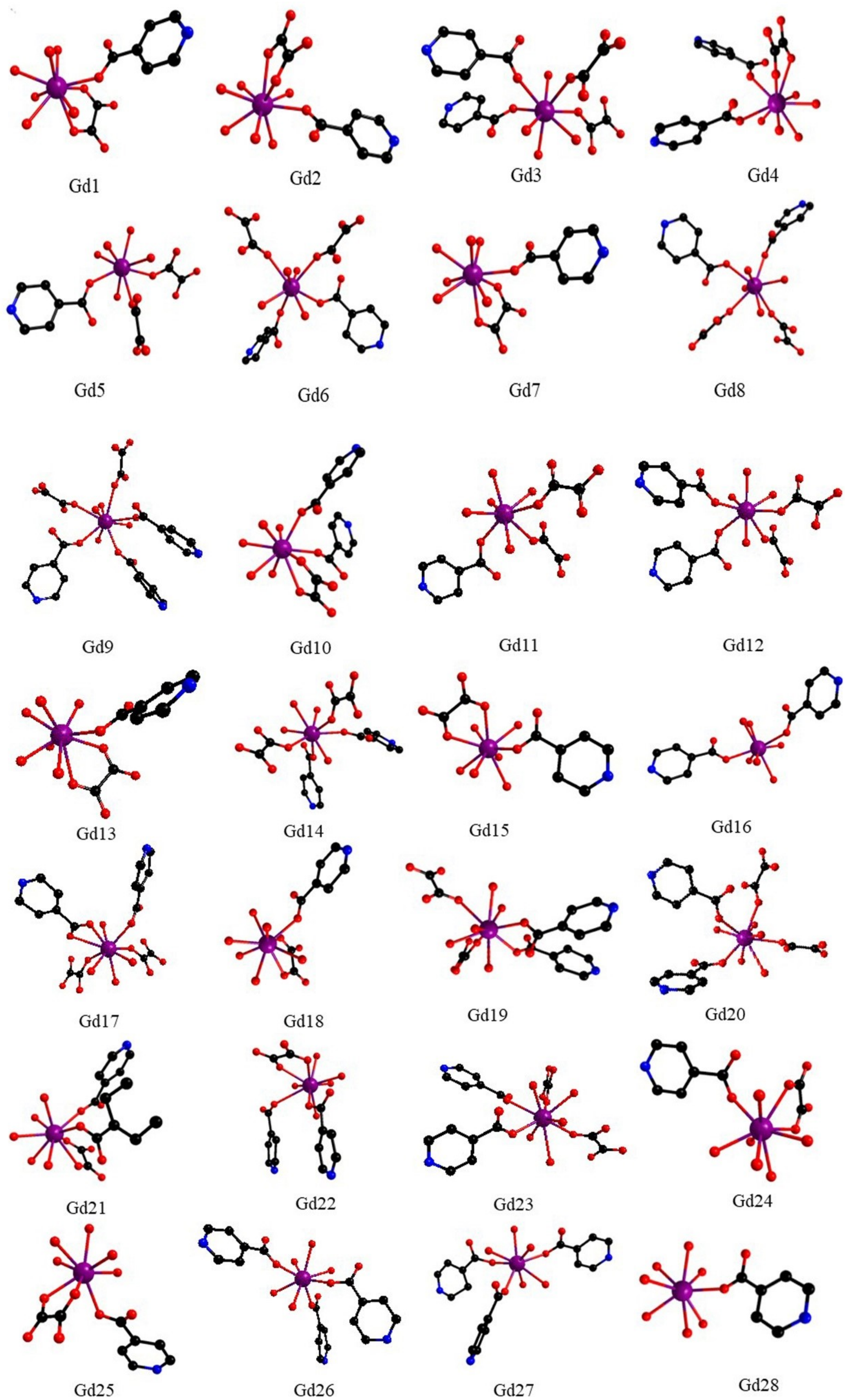


Fig. S4 Representation of $\mathbf{Gd_{28}}$ based on (a) $\mathbf{Gd_3}$ and $\mathbf{Gd_4}$ unit (b) $C_2O_4^{2-}$ anionic templates.



F

ig. S5 Coordination environments of 28 different Gd^{III} ions.

Table S1. The CShM (Continuous Shape Measures) values of eight-coordination of Gd^{III} ions in **Gd_{28}**.

	Octagon	**Heptagonal pyramid**	**Hexagonal bipyramid**	**Cube**	**Square antiprism**	**Triangular dodecahedron**	**Johnson-Gyrobifastigium J26**	**Johnson-Elongated triangular bipyramid J14**	**Johnson-Biaugmented trigonal prism J50**	**Biaugmented trigonal prism**	**Snub diphenoid J84**	**Triakis tetrahedron**	**Elongated trigonal bipyramid**
Gd16	31.57244	23.29306	16.96220	10.17181	2.45775	**0.31430**	13.56307	29.40452	2.82168	2.18982	2.49707	11.00914	25.32895
Gd8	58.09215	51.99503	46.87071	45.98718	49.82011	48.36206	47.60483	**43.10357**	45.00656	46.39710	48.22951	46.33355	47.12274
Gd12	28.99820	23.67745	16.13101	10.46344	2.14151	**0.80577**	14.23738	28.22626	2.62350	1.80749	3.22529	11.25575	23.15551
Gd3	30.70519	23.31967	14.41076	9.57658	2.70619	**0.83199**	13.80598	28.11689	2.72063	1.93110	3.45059	10.33901	22.96524
Gd14	44.78659	38.57630	43.27928	43.26096	43.54551	43.14939	38.00744	**33.10107**	38.15059	38.84904	35.47430	39.42785	40.07003
Gd6	28.76705	24.16349	16.28630	9.85993	1.92066	**0.64540**	14.86619	28.79118	3.04664	2.11437	3.47886	10.46608	23.58522
Gd26	30.23454	24.08236	16.88696	9.88705	2.18320	**0.40640**	12.79257	29.24734	2.24312	1.81316	2.10115	10.41697	24.58076
Gd27	29.55541	23.61301	15.88942	10.15675	1.91876	**1.04234**	13.37332	27.23162	1.60927	1.19297	2.81509	10.74322	22.91530
Gd28	22.69508	23.43771	16.80728	11.34599	**1.11197**	3.14065	14.44132	25.21684	2.93171	2.48548	5.11699	12.05596	20.67076
Gd11	31.37728	22.82900	15.49588	9.77657	3.18784	**0.74079**	14.99344	28.79753	3.10827	1.98741	3.95448	10.22424	23.71704
Gd5	27.85828	23.95382	15.83208	9.41022	1.73981	**0.94513**	15.27388	28.08186	2.87018	1.81651	4.02845	9.96920	22.42156
Gd22	30.96602	22.64395	13.98008	11.60034	4.85064	3.66092	9.58619	24.59086	2.85437	**1.84665**	5.21051	12.40456	19.29079
Gd25	30.19222	23.35258	17.05882	13.34342	3.60991	2.32018	12.88298	25.75615	1.75428	**1.00856**	3.16702	13.47305	21.45386

Table S2. The CShM (Continuous Shape Measures) values of nine-coordination of Gd^{III} ions in $\mathbf{Gd_{28}}$.

	Enneagon	**Octagonal pyramid**	**Heptagonal bipyramid**	**Johnson triangular cupola J3**	**Capped cube J8**	**Spherical-relaxed capped cube**	**Capped square antiprism J10 J26**	**Spherical capped square antiprismJ14**	**Tricapped trigonal prism J51trigonal prism J50**	**Spherical tricapped trigonal prism**	**Tridiminished icosahedron J63**	**Hula-hoopT**	**Muffin**
Gd23	36.67895	22.44078	19.19049	16.16553	9.69815	9.17781	1.36254	1.00447	1.78258	**0.99039**	12.83151	11.17741	1.30642
Gd20	36.06425	21.80207	18.99208	16.37753	9.12616	8.63518	1.40016	1.06496	2.37189	**0.85709**	12.19337	11.08981	1.60496
Gd9	37.03736	21.08640	17.69969	16.65952	10.21308	8.70924	2.57693	1.29014	3.16355	**0.62058**	11.90255	11.58504	1.55821
Gd4	35.94429	22.97067	17.11069	14.99922	7.84317	7.09962	1.72506	**1.27777**	2.95977	1.69789	10.92071	9.86830	1.78316
Gd19	36.67920	22.17129	17.42200	16.72087	8.69130	7.72734	2.30843	1.39112	2.80759	**0.99565**	13.26578	10.07724	1.41096
Gd21	43.02343	33.06960	31.79243	23.23462	25.04451	25.31343	**20.65182**	20.86608	20.96080	21.09692	26.97353	28.19029	21.35039
Gd13	63.28090	58.25949	56.63662	54.42950	49.61953	55.67917	**47.05175**	53.83618	48.30373	54.25396	54.55605	55.00210	53.06404
Gd24	33.82836	21.63958	16.69124	14.96467	8.84803	8.05750	1.70199	**1.06731**	3.08628	1.80431	12.69559	9.80164	1.50842
Gd10	36.17768	23.42041	17.23778	14.64841	7.58813	6.91348	1.38227	**0.97006**	3.32150	2.07949	10.70363	11.31211	1.70320
Gd7	33.55453	22.42499	18.09518	13.58033	9.77277	8.83270	1.76147	**0.91942**	1.77412	1.31445	12.61838	11.48950	1.43830
Gd15	34.84055	20.93221	17.16954	15.83306	10.65338	9.38573	2.11519	**1.21141**	3.34717	1.42565	11.05302	10.26849	1.26485
Gd18	35.39423	22.54196	19.69406	15.41679	11.25483	10.04386	1.65672	**0.70204**	3.11966	1.83977	11.70083	10.76745	1.34002
Gd2	34.73584	22.06043	17.43797	13.90846	10.02244	9.09152	1.82951	**1.14580**	1.97125	1.18065	12.01613	12.22453	1.50683
Gd1	34.34913	21.82234	17.31980	13.61914	9.42099	8.52795	1.52918	**0.89866**	1.93312	1.31582	11.98333	11.92160	1.50423
Gd17	34.25488	21.19019	16.73164	15.88556	8.48350	7.75421	2.97965	2.55570	3.43371	**2.14413**	12.77926	8.73959	2.30066

PXRD pattern

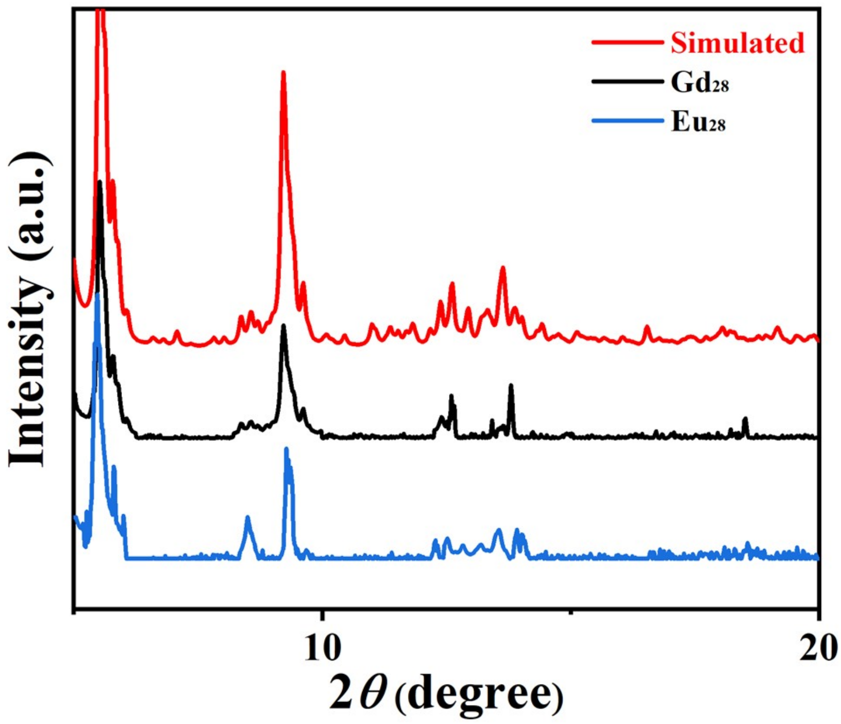


Fig. S6 The PXRD pattern of $\mathbf{Gd_{28}}$ and $\mathbf{Eu_{28}}$ at 2θ = 5-20°.

IR spectrum

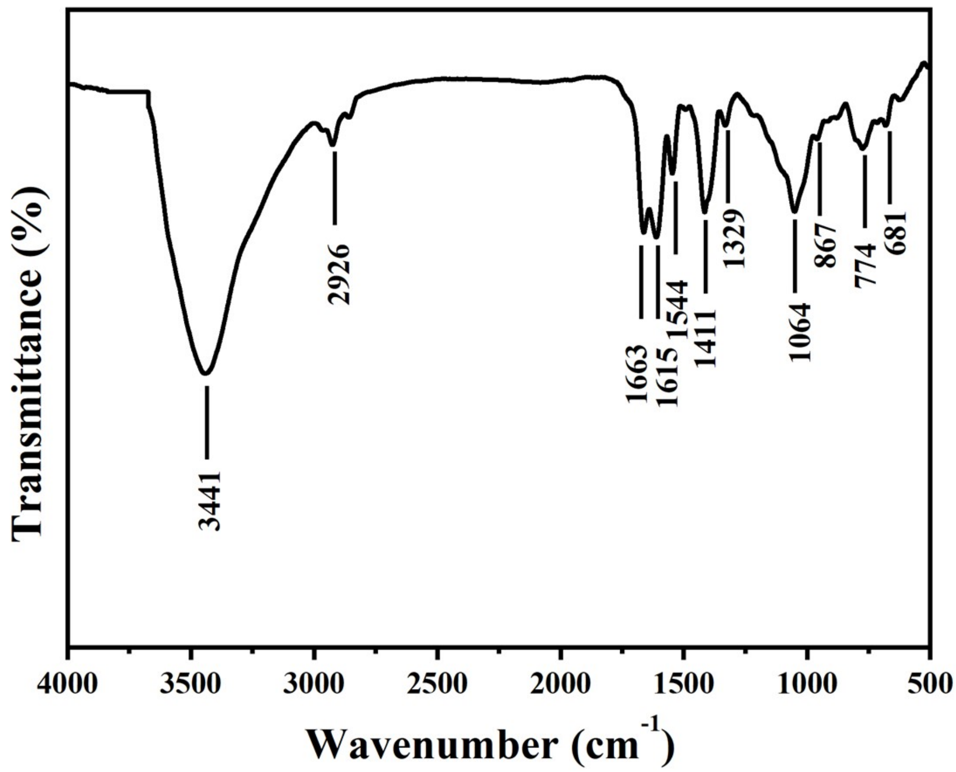


Fig. S7 The IR spectrum of **Gd_{28}**.

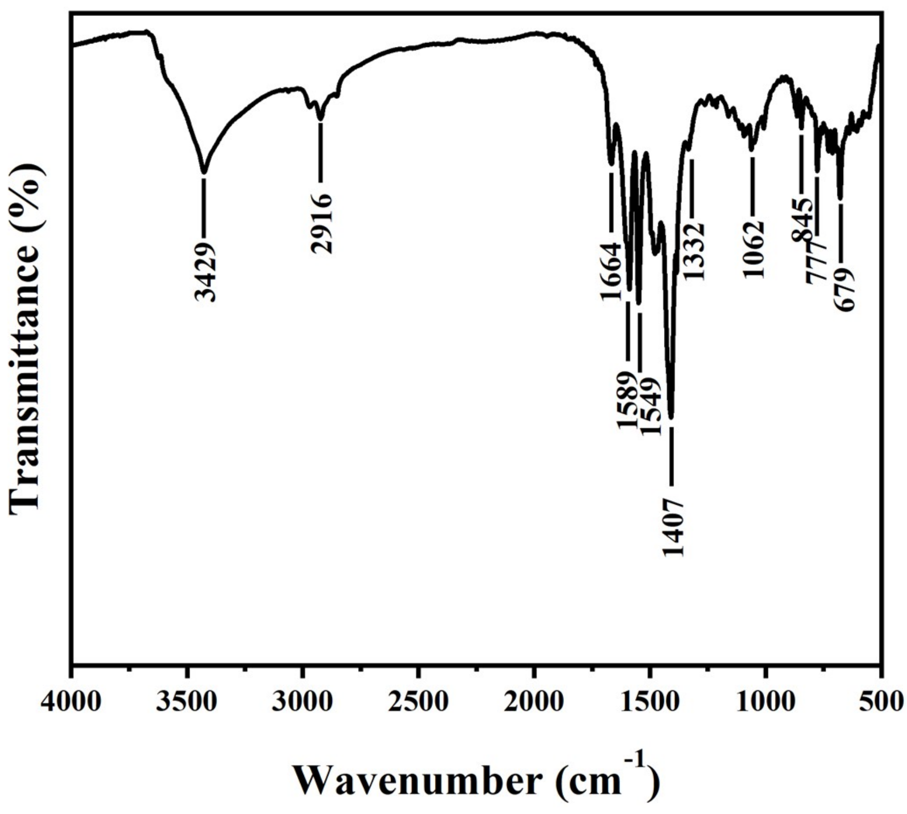


Fig. S8 The IR spectrum of **Eu_{28}**.

As presented in Fig. S7-8, the IR spectra patterns of $\mathbf{Gd_{28}}$ and $\mathbf{Eu_{28}}$ at 4000-400 cm^{-1} are almost the same. So only the infrared spectrum of $\mathbf{Gd_{28}}$ is discussed in detail. The broad and strong absorption peak in the field of 3441 cm^{-1} ascribes to the -OH of H_2O molecules and the characteristic peak at 2926 cm^{-1} belongs to the signature of *v*(C-H), while the stretching vibration peak at 1663 cm^{-1} reveal the presence of the pyridine ring. In addition, the characteristic peaks of symmetric and antisymmetric stretching vibration on $-COO^-$ groups appear at 1411 cm^{-1} and 1544 cm^{-1}. The peak at 867-681 cm^{-1} corresponds to the characteristic peak of the *v*(Gd-O). The mentioned infrared peaks above are consistent with those of lanthanide clusters previously reported in the literature.

TG

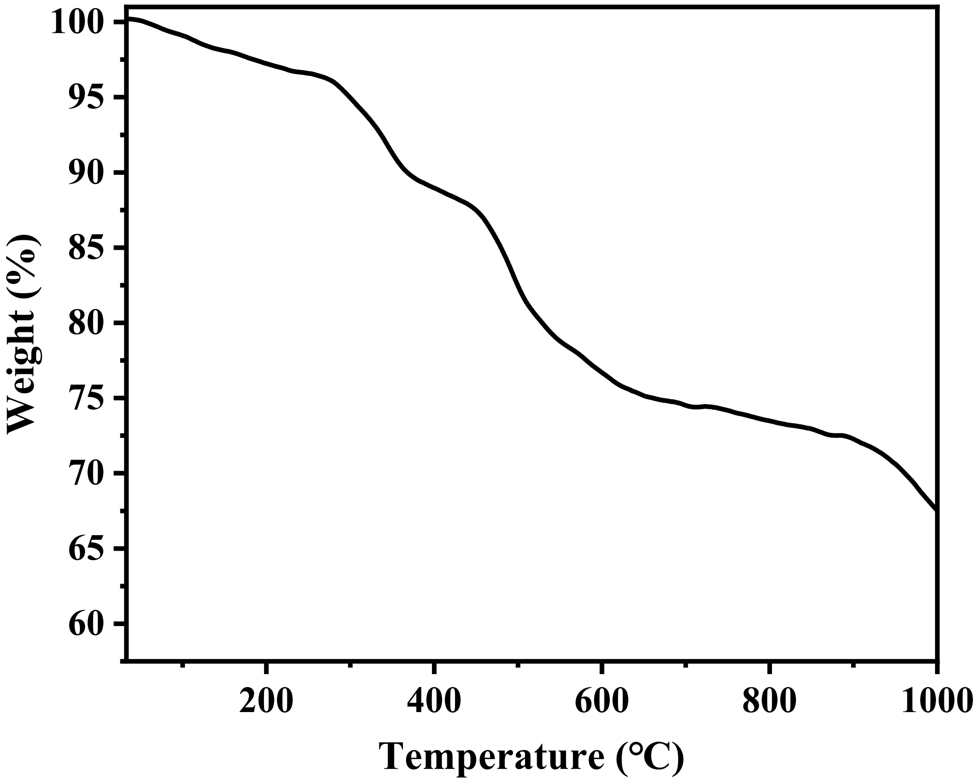


Fig. S9 TGA curve of $\mathbf{Gd_{28}}$.

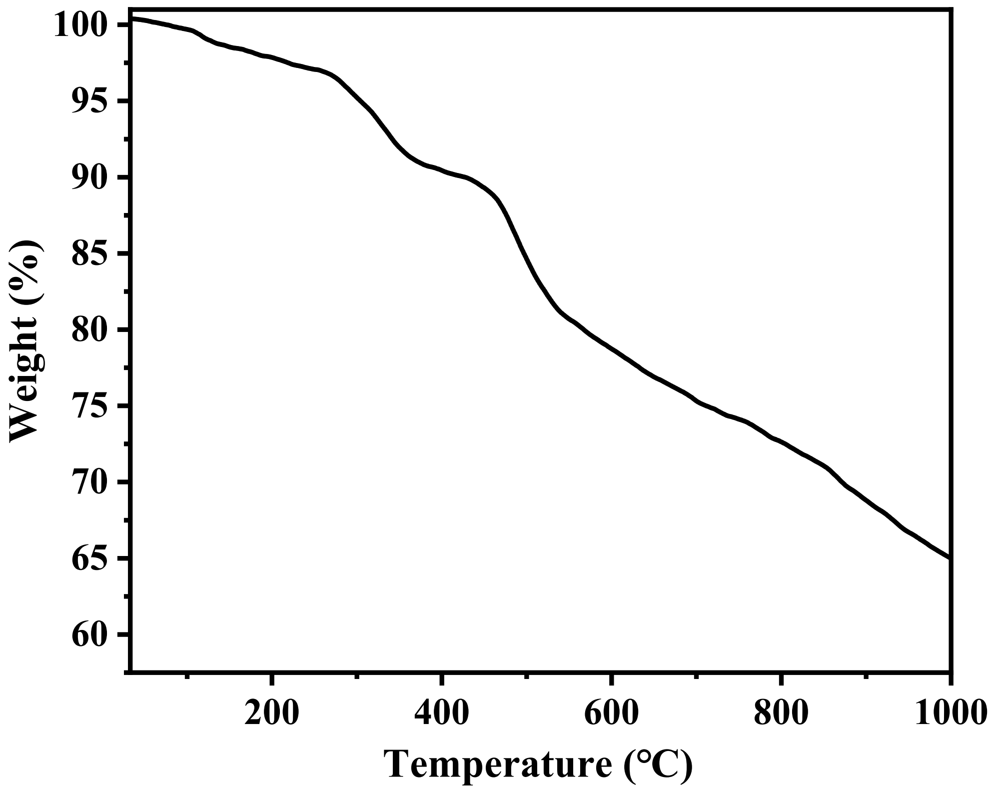


Fig. S10 TGA curve of $\mathbf{Eu_{28}}$.

EDS

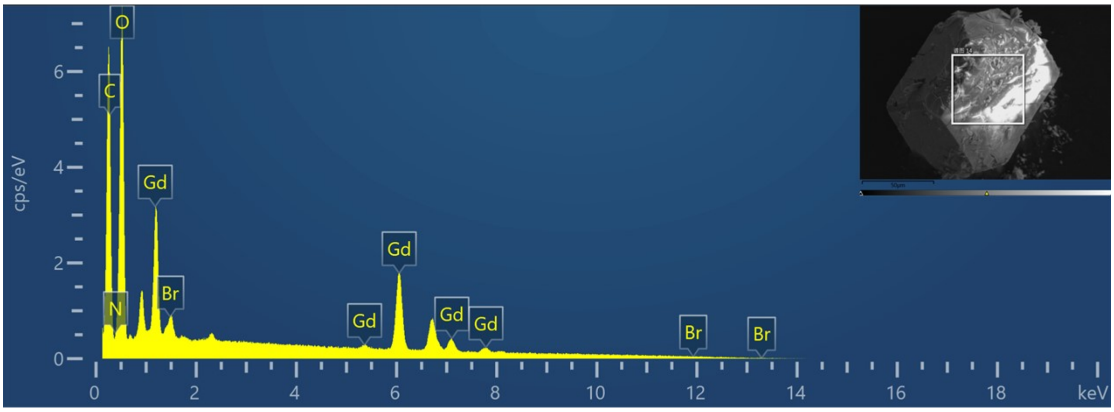


Fig. S11 The EDS measurement of compound **Gd_{28}**.

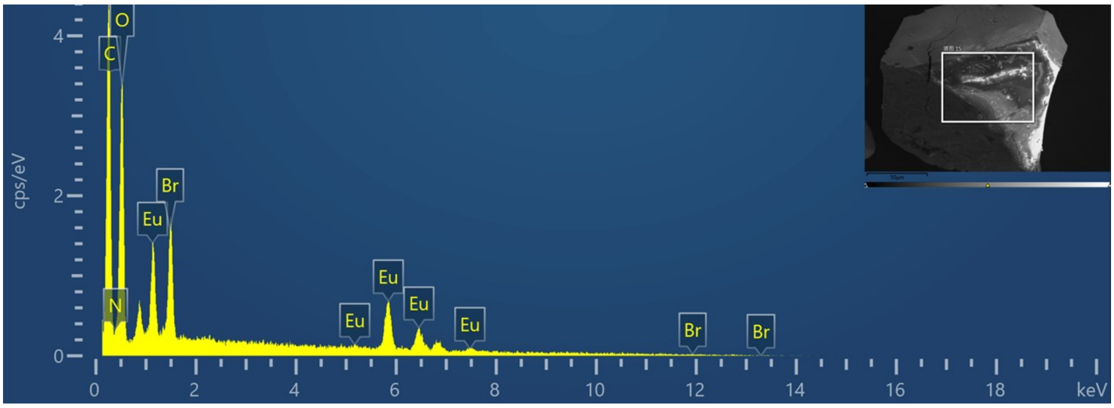


Fig. S12 The EDS measurement of compound **Eu_{28}**.

Luminescence of Eu_{28}

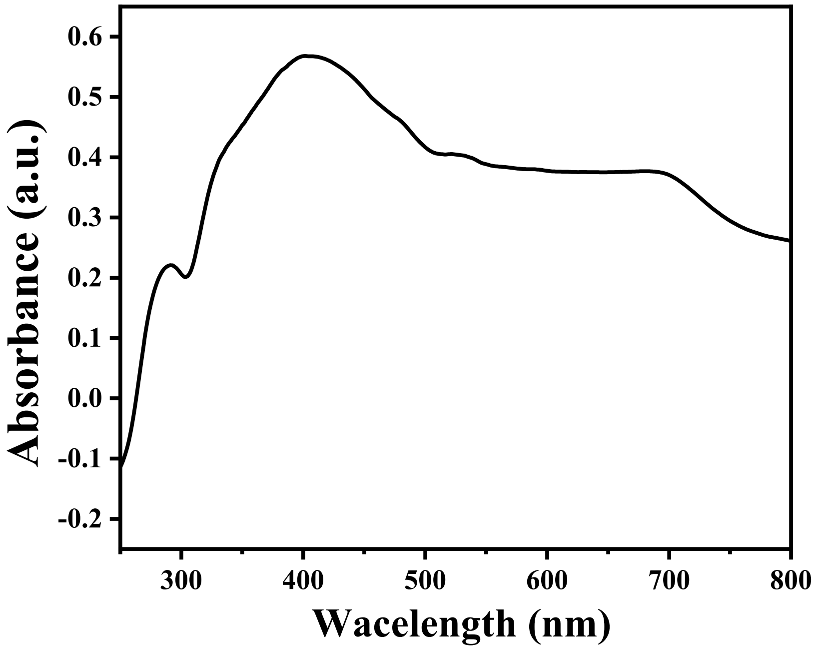


Fig. S13 The solid-state UV-absorption spectra in the wavelength range 250-800 nm for $\mathbf{Eu_{28}}$.

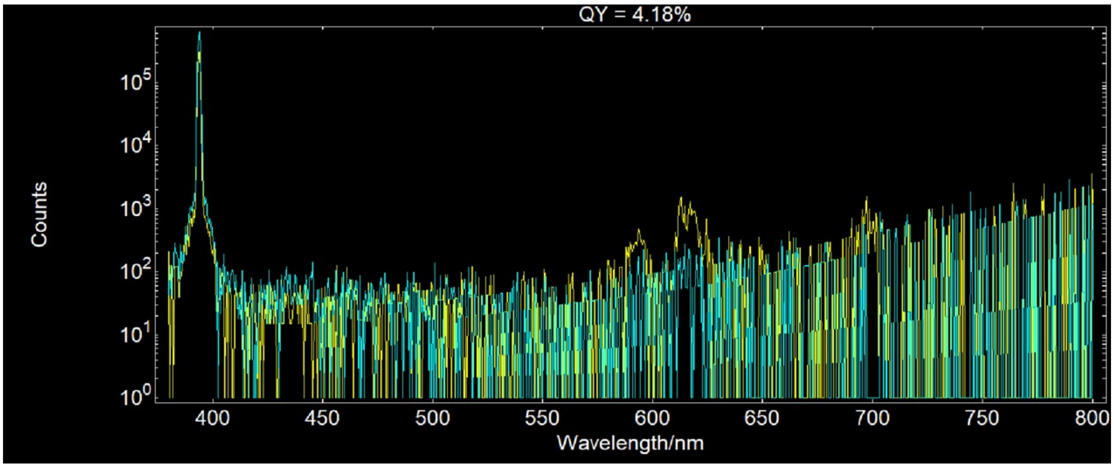


Fig. S14 The solid-state photoluminescence quantum yield for $\mathbf{Eu_{28}}$.

Magnetic property of Gd_{28}

Table S3 Summary -ΔS_m for some reported 4f and 3d-4f clusters

Compound	-ΔS_m^{max} (J kg^{-1} K^{-1})	T (K)	ΔH (T)	Ref.
{Gd_{28}}	**37.5**	**2.0**	**7.0**	**This work**
{**Gd_{27}**}	41.8	2.0	7.0	1
{**Gd_{32}**}	43.0	2.0	7.0	2
{**Gd_{36}**}	39.7	2.5	7.0	3
{**Gd_{37}**}	38.7	2.0	7.0	4
{**Gd_{38}**}	37.9	1.8	7.0	5
{**Gd_{60}**}	48.0	2.0	7.0	6
{**Gd_{104}**}	46.9	2.0	7.0	7
{**Gd_{140}**}	38.0	2.0	7.0	8
{**$Gd_{23}Ni_{20}$**}	38.1	2.0	7.0	9
{**$Gd_{36}Ni_{12}$**}	36.3	3.0	7.0	10
{**$Co^{II}_9Co^{III}Gd_{42}$**}	41.26	2.0	7.0	11
{**$Co^{II}Co^{III}_6Gd_{18}$**}	36.90	2.0	7.0	12

Table S4 Summary of the reported lanthanide clusters with isonicotinic ligand

Compound	Formula	Dimensionality	Ref.
Ln	$[Ln(IN)(CO_3)(H_2O)]$ (Ln = La, Eu)	2D	13
Ln	$[Ln(IN)_2L]$ (Ln = Eu, Tb, Er, Dy, Ho, Gd, La, L = OCH_2CH_2OH)	1D	14
Ln	L-/D-$\{Ln[IN][HIN][CH_2OCH_2O]\}_n$ (Ln = Gd, Dy)	1D	15
Ln_3	$[Ln_3(IN)_2(bdc)_{3.5}(H_2O)_3]\cdot 0.5H_2O$ (Ln = Er, Ho)	3D	16
Ln_3	$[Ln_3(IN)_3(\mu_3\text{-}OH)(HIDA)(IDA)_2]_n$ (Ln = Eu, Sm)	2D	17
Ln_4	$\{[Nd_4(ox)_4(NO_3)_2(OH)_2(H_2O)_2]\cdot 5H_2O\}n$	3D	18
Ln_8	$[Ln_8(IN)_{14}(\mu_3\text{-}OH)_8(\mu_2\text{-}OH)_2(H_2O)_8]\cdot xH_2O$ (Ln = 1-Gd, Ln = 2-Dy, Ln = 3-Eu).	0D	19
Ln_{18}	$\{[Gd_{18}(IN)_{20}(HCOO)_8(\mu_6\text{-}O)(\mu_3\text{-}OH)_{24}(H_2O)_4]\cdot 4H_2O\}n$	3D	20
	$\{[Eu_{18}(IN)_{16}(HCOO)_8(CH_3COO)_4(\mu_6\text{-}O)(\mu_3\text{-}OH)_{24}(H_2O)_4]\cdot 5H_2O\}_n$		
Ln_{26}	$[Ho_{26}(IN)_{28}(CH_3COO)_4(CO_3)_{10}(OH)_{26}(H_2O)_{18}]\cdot 20H_2O$	0D	21
	$[Er_{26}(IN)_{29}(CH_3COO)_3(CO_3)_{10}(OH)_{26}(H_2O)_{19}]\cdot 26H_2O$		
Ho_{48}	$K_2[Ho_{48}(IN)_{46}(\mu_3\text{-}OH)_{84}(\mu_4\text{-}OH)_4(\mu_5\text{-}O)_2(OAc)_4(H_2O)_{14}(CO_3)Br_2]\cdot 2HIN\cdot 20H_2O$	2D	22
Dy_{30}	$Dy_{30}I(IN)_{41}(\mu_3\text{-}OH)_{24}(\mu_3\text{-}O)_6(NO_3)_9(OH)_3(H_2O)_{38}$	0D	23
Dy_{104}	$Dy_{104}I_4(IN)_{125}(\mu_3\text{-}OH)_{80}(\mu_3\text{-}O)_{24}(NO_3)_{36}(OH)_{19}\cdot(H_2O)_{167}$		

Table S5 Selected bond lengths (Å) and bond angles (°) of main metal atoms for $\mathbf{Gd_{28}}$.

Gd(1)-O(39)	2.345(9)	Gd(15)-O(13)	2.377(9)
Gd(1)-O(27)	2.599(10)	Gd(15)-O(28)	2.376(10)
Gd(2)-O(91)	2.388(10)	Gd(16)-O(44)	2.384(9)
Gd(2)-O(3)	2.393(10)	Gd(16)-O(94)	2.392(11)
Gd(3)-O(4)	2.339(9)	Gd(17)-O(54)	2.366(10)
Gd(3)-O(16)	2.351(9)	Gd(17)-O(77)	2.767(17)
Gd(4)-O(6)	2.374(9)	Gd(18)-O(62)	2.375(9)
Gd(4)-O(32)	2.402(10)	Gd(18)-O(19)	2.580(10)
Gd(5)-O(46)	2.345(9)	Gd(19)-O(67)	2.352(10)
Gd(5)-O(16)	2.353(9)	Gd(19)-O(5W)	2.628(12)
Gd(6)-O(50)	2.333(10)	Gd(20)-O(6)	2.345(9)
Gd(6)-O(8)	2.357(9)	Gd(20)-O(87)	2.353(11)
Gd(7)-O(10W)	2.360(16)	Gd(21)-O(93)	2.362(13)
Gd(7)-O(63)	2.370(11)	Gd(21)-O(37)	2.371(9)
Gd(8)-O(33)	2.321(9)	Gd(22)-O(77)	2.905(19)
Gd(8)-O(10)	2.330(9)	Gd(22)-O(13)	2.341(10)
Gd(9)-O(45)	2.403(9)	Gd(23)-O(65)	2.354(10)
Gd(9)-O(31)	2.410(9)	Gd(23)-O(11)	2.354(10)
Gd(10)-O(5)	2.379(9)	Gd(24)-O(111)	2.317(12)
Gd(10)-O(107)	2.384(13)	Gd(24)-O(37)	2.395(10)
Gd(11)-O(46)	2.346(9)	Gd(25)-O(65)	2.312(11)
Gd(11)-O(25W)	2.367(11)	Gd(25)-O(67)	2.357(11)
Gd(12)-O(31)	2.342(10)	Gd(26)-O(75)	2.331(11)
Gd(12)-O(99)	2.367(11)	Gd(26)-O(104)	2.334(12)
Gd(13)-O(6)	2.362(9)	Gd(27)-O(103)	2.322(15)
Gd(13)-O(45)	2.363(9)	Gd(27)-O(112)	2.331(15)
Gd(14)-O(102)	2.330(13)	Gd(28)-O(95)	2.317(18)
Gd(14)-O(8)	2.357(9)	Gd(28)-O(46)	2.385(9)
O(39)-Gd(1)-O(79)	87.0(4)	O(13)-Gd(15)-O(28)	139.9(3)
O(39)-Gd(1)-O(3)	140.5(3)	O(13)-Gd(15)-O(109)	75.3(4)
O(91)-Gd(2)-O(3)	84.2(4)	O(94)-Gd(16)-O(83)	144.6(4)
O(91)-Gd(2)-O(12)	141.7(4)	O(11)-Gd(16)-O(83)	97.6(4)
O(4)-Gd(3)-O(16)	74.2(3)	O(73)-Gd(17)-O(43)	121.2(4)
O(4)-Gd(3)-O(5)	143.8(3)	O(13)-Gd(17)-O(43)	141.7(3)
O(6)-Gd(4)-O(32)	90.3(3)	O(62)-Gd(18)-O(11W)	69.9(4)
O(6)-Gd(4)-O(5)	139.4(3)	O(62)-Gd(18)-O(72)	128.7(5)
O(46)-Gd(5)-O(16)	71.1(3)	O(67)-Gd(19)-O(49)	125.4(3)
O(46)-Gd(5)-O(39)	143.6(3)	O(71)-Gd(19)-O(49)	145.9(4)
O(50)-Gd(6)-O(8)	72.0(3)	O(6)-Gd(20)-O(87)	87.2(4)
O(50)-Gd(6)-O(85)	68.8(4)	O(6)-Gd(20)-O(54)	130.2(3)
O(63)-Gd(7)-O(35)	144.7(4)	O(37)-Gd(21)-O(67)	137.1(3)
O(1)-Gd(7)-O(35)	77.2(3)	O(66)-Gd(21)-O(67)	70.0(4)

O(33)-Gd(8)-O(10)	73.3(3)	O(97)-Gd(22)-O(40)	82.9(5)
O(33)-Gd(8)-O(1)	145.0(3)	O(13)-Gd(22)-O(40)	140.9(3)
O(45)-Gd(9)-O(31)	145.2(3)	O(11)-Gd(23)-O(43)	74.0(3)
O(45)-Gd(9)-O(33)	144.4(3)	O(65)-Gd(23)-O(92)	84.9(4)
O(5)-Gd(10)-O(29W)	75.0(3)	O(111)-Gd(24)-O(37)	82.0(4)
O(107)-Gd(10)-O(29W)	77.8(4)	O(111)-Gd(24)-O(65)	134.5(4)
O(25W)-Gd(11)-O(4)	123.0(3)	O(65)-Gd(25)-O(67)	141.5(4)
O(28)-Gd(11)-O(4)	139.1(3)	O(65)-Gd(25)-O(81)	95.1(4)
O(31)-Gd(12)-O(99)	95.2(4)	O(75)-Gd(26)-O(104)	143.5(4)
O(31)-Gd(12)-O(37)	143.5(3)	O(75)-Gd(26)-O(31)	91.4(4)
O(6)-Gd(13)-O(59)	128.9(3)	O(50)-Gd(27)-O(8)	68.7(3)
O(45)-Gd(13)-O(59)	67.6(3)	O(114)-Gd(27)-O(8)	141.7(4)
O(102)-Gd(14)-O(49)	144.9(4)	O(46)-Gd(28)-O(16)	68.8(3)
O(8)-Gd(14)-O(49)	68.9(3)	O(8W)-Gd(28)-O(16)	140.8(8)

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