Electronic Supplementary Information (ESI)

Alkali-regulated Fe_6 and Fe_{18} molecular clusters and their structural transformation

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Experiment details

All chemicals and solvents used in the syntheses were of analytical grade and used without further purification. IR spectra were recorded on ABB Bomen MB 102 series FT-IR spectrometer with KBr pellets in the frequency range of 4000-400 cm^{-1}. The elemental analyses (C, H, N contents) were determined on a Vario EL cube analyzer. Powder X-ray diffraction (PXRD) data were collected on a Rigaku SmartLab (3 kW) X-ray diffractometer with Cu Kα radiation. Thermogravimetric analyses (TGA) were performed on a Netzsch TG209 thermal analyzer from room temperature to 800 °C under N_2 atmosphere at a heating rate of 10 °C/min. All magnetization data were recorded on a Quantum Design SQUID MPMS3 magnetometer. The variable-temperature magnetization was measured with an external magnetic field of 1000 Oe in the temperature range of 3-300 K.

X-ray Crystallography

Single crystals of **Fe$_6$** and **Fe$_{18}$** with appropriate dimensions were chosen under an optical microscope and quickly coated with high vacuum grease (Dow Corning Corporation) before being mounted on a glass fiber for data collections. Single-crystal X-ray diffraction data of **Fe$_6$** and **Fe$_{18}$** were collected using a Rigaku XtaLAB MM007 CCD diffractometer with Cu Ka radiation (λ = 1.5418 Å) radiation. Structures of **Fe$_6$** and **Fe$_{18}$** were solved using SHELXS-97 (direct methods) and refined using SHELXL-97 (full-matrix least-squares on F^2).[1] Hydrogen atoms were placed in calculated positions and included as riding atoms with isotropic displacement parameters 1.2-1.5 times U_{eq} of the attached C atoms. The X-ray crystallographic coordinates for structures reported in this article have been deposited at the Cambridge Crystallographic Data Centre (CCDC). CCDC: 2080069 (**Fe$_{18}$**) and 2080070 (**Fe$_6$**). Furthermore, All structures were examined using the Addsym subroutine of PLATON[2] to assure that no additional symmetry could be applied to the models. Pertinent crystallographic data collection and refinement parameters of **Fe$_6$** and **Fe$_{18}$** are collated in Table S1. Selected bond lengths and angles are collated in Table S2.

Synthesis of $[Fe_{18}O_8(OH)_8(H_2thmmg)_{10}]$ (Fe_{18})

A mixture of H_5thmmg (17.9 mg, 0.1 mmol), $Fe(NO_3)_3$•$9H_2O$ (40.4 mg, 0.1 mmol), triethanolamine (14.9 mg, 0.1 mmol), KCl (7.5 mg, 0.1 mmol), H_2O (4 mL), CH_3OH (4 mL) were sealed in a 20 mL glass reaction bottle, and 40μL triethylamine was slowly added into the mixture while ultrasound, then transferred to a preheated oven at 65 °C for 3 days. We can obtain red-block crystals **Fe_{18}**. The crystals **Fe_{18}** were isolated by filtration, washed with H_2O and Methanol, and dried in air (yield 33.2 % based on Fe). Elemental analyses calc. (found) for **Fe_{18}**: $C_{60}H_{104}Fe_{18}N_{10}O_{66}$: C, 23.81 (23.78); H, 3.46 (3.50); N, 4.63 (4.59) %. Selected IR peaks (cm^{-1}): 3434 (s), 2964 (m), 2925 (m), 2856 (m), 2296 (w),1629 (s), 1381 (m), 1080 (m), 1043 (m), 929 (w), 879 (w), 565 (w).

Synthesis of $[Fe_6(H_2thmmg)_6]$ (Fe_6)

A mixture of H_5thmmg (17.9 mg, 0.1 mmol), $Fe(NO_3)_3$•$9H_2O$ (40.4 mg, 0.1 mmol), triethanolamine (14.9 mg, 0.1 mmol), KCl (7.5 mg, 0.1 mmol), H_2O (4 mL), CH_3OH (4 mL) were sealed in a 20 mL glass reaction bottle, and 30μL triethylamine was slowly added into the mixture while ultrasound, then transferred to a preheated oven at 65 °C for five hours. We can obtain yellow-block crystals **Fe_6**. The crystals **Fe_6** were isolated by filtration, washed with H_2O and Methanol, and dried in air (yield 24.1 % based on Fe). Elemental analyses calc. (found) for **Fe_6**: $C_{36}H_{60}Fe_6N_6O_{30}$: C, 31.1 (30.5); H, 4.3 (4.5); N, 6.0 (6.2) %. Selected IR peaks (cm^{-1}): 3434 (s), 2923 (m), 2854 (m), 2339 (w), 1635 (s), 1440 (w), 1348 (m), 1224 (w), 1153 (w), 1086 (m), 1031 (m), 804 (w), 719(w), 632(w), 588(m), 507(m).

The transformation from Fe_6 to Fe_{18}

When we obtain yellow-block crystals **Fe_6** in the solution, 10 μL triethylamine was slowly added into the mixture. Then transferred to a preheated oven at 65 °C for 3 days. We can obtain red-block crystals **Fe_{18}**.

Fig. S1: The coordinative modes of H_5thmmg ligands in Fe_6 and Fe_{18} clusters.

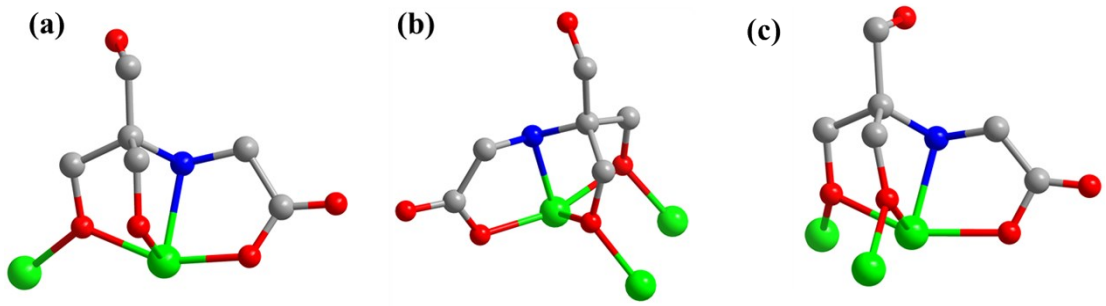

Fig. S2: The molecule packing of Fe_{18} and Fe_6 in the unit cell.

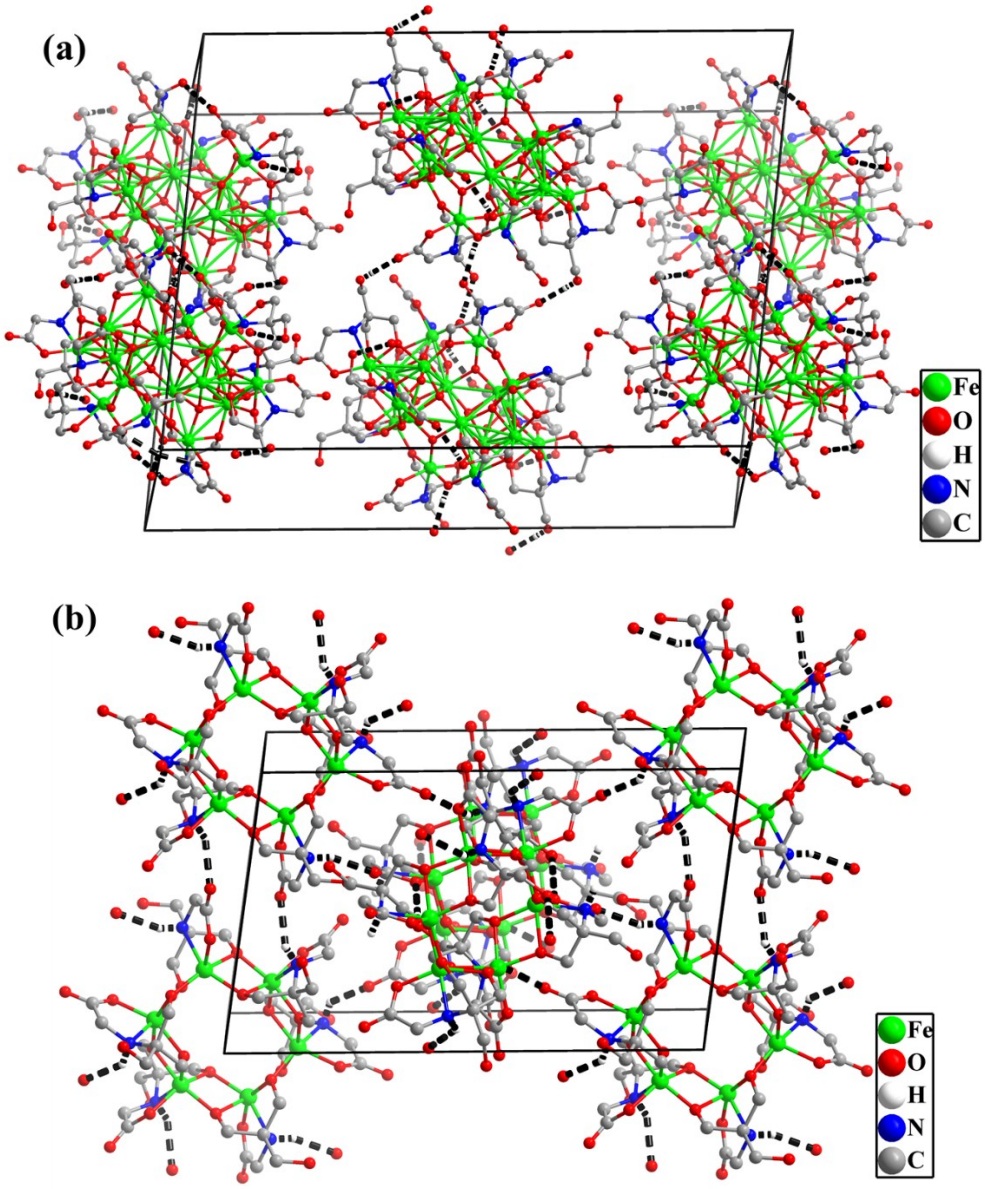

Fig. S3: The IR spectra of Fe_{18}, $Fe_{18\text{-trans}}$ and Fe_6.

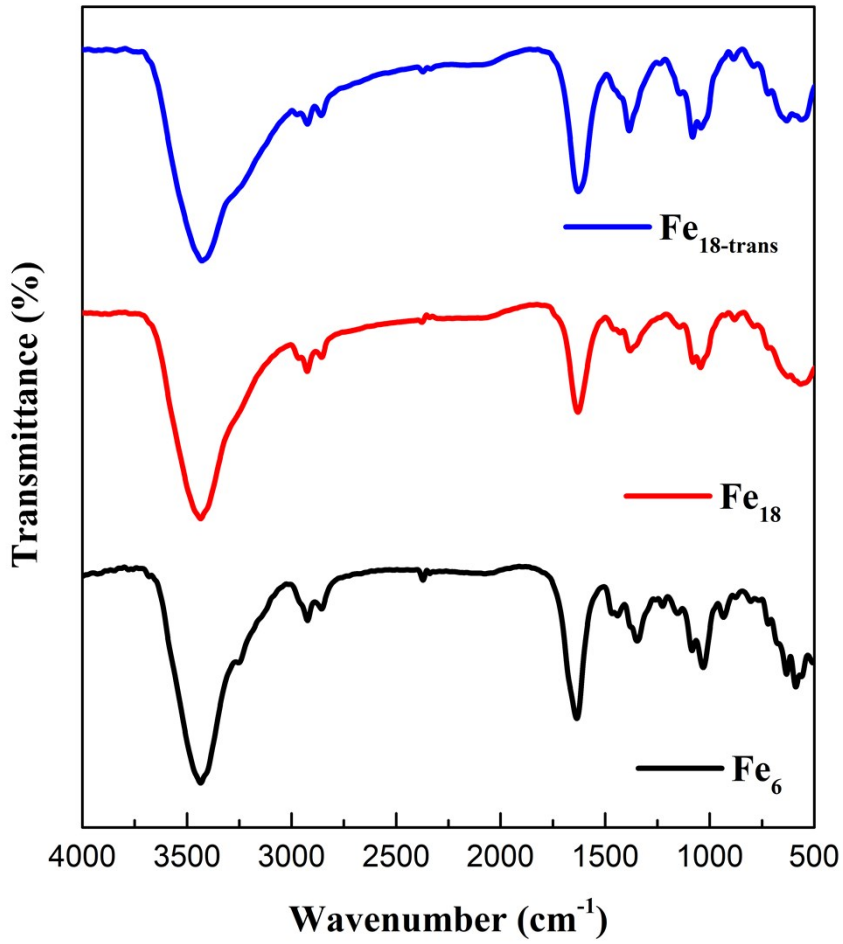

Fig. S4: The TGA of Fe_{18} and Fe_6.

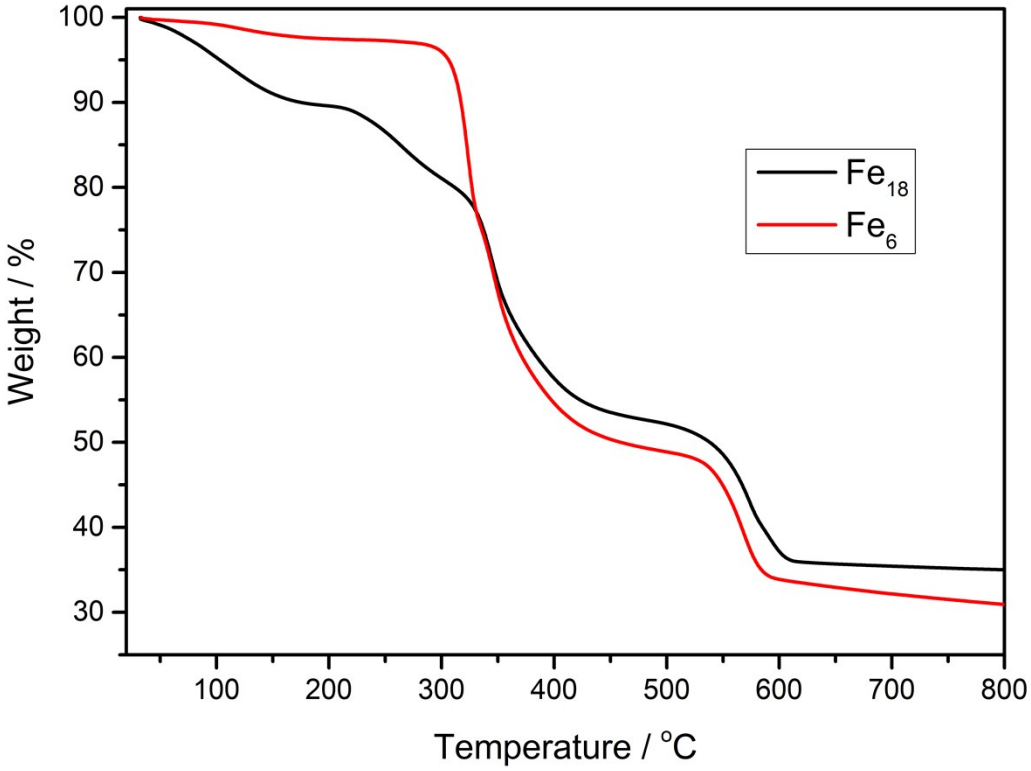

Fig. S5: Plots of the temperature dependence of χ_m^{-1} between 160-300 K for Fe_6 and Fe_{18}.

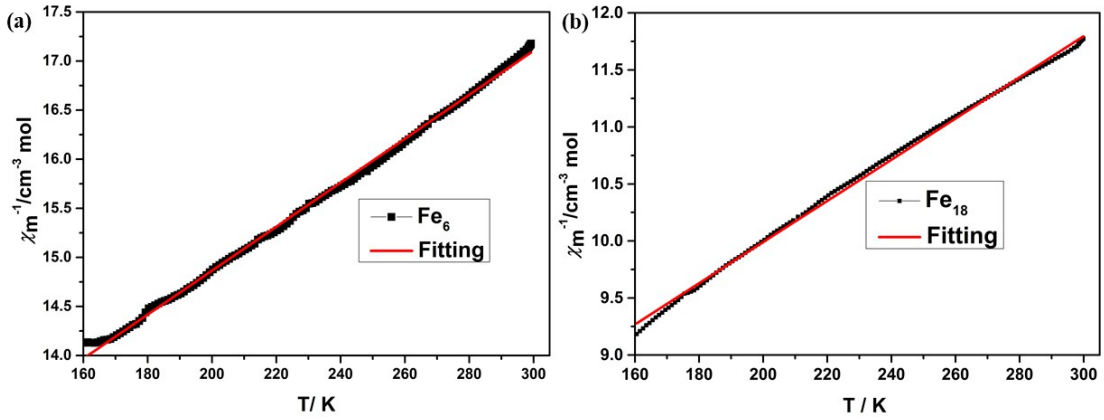

Table S1: Crystal data for Fe_{18} and Fe_6.

Identification code	**Fe_{18}**	**Fe_6**
Empirical formula	$C_{60}H_{104}Fe_{18}N_{10}O_{66}$	$C_{36}H_{60}Fe_6N_6O_{30}$
Formula weight	3026.83	1392.00
Temperature/K	117(20)	100.00(10)
Crystal system	monoclinic	triclinic
Space group	*C*2/*c*	*P*-1
a/Å	24.6598(10)	11.8437(7)
b/Å	15.4024(7)	13.2329(8)
c/Å	33.2312(13)	20.6099(10)
α/°	90	78.264(4)
β/°	97.015(4)	80.601(4)
γ/°	90	73.213(5)
Volume/Å^3	12527.4(9)	3008.9(3)
Z	4	2
ρ_{calc}g/cm^3	1.605	1.536
μ/mm^{-1}	16.963	12.060
F(000)	6120.0	1428.0
Radiation	CuKα (λ = 1.54184)	CuKα (λ = 1.54184)
2Θ range for data collection/°	7.128 to 152.644	7.074 to 147.458
Index ranges	-29 ≤ h ≤ 29, -16 ≤ k ≤ 19, -39 ≤ l ≤ 41	-14 ≤ h ≤ 14, -16 ≤ k ≤ 15, -25 ≤ l ≤ 22
Reflections collected	22743	27826
Independent reflections	11964 [R_{int} =0.0627, R_{sigma} = 0.1032]	11235 [R_{int} = 0.0645, R_{sigma} = 0.0711]
Data/restraints/parameters	11964/6/699	11235/0/709
Goodness-of-fit on F^2	1.073	1.077
Final R indexes [I>=2σ (I)]	R_1 = 0.0723, wR_2 = 0.1928	R_1 = 0.0844, wR_2 = 0.2262
Final R indexes [all data]	R_1 = 0.1002, wR_2 = 0.2154	R_1 = 0.1044, wR_2 = 0.2373
Largest diff. peak/hole / e Å^{-3}	1.32/-1.09	1.62/-0.75

Table S2: Selected bond distances (Å) and angles (°) for Fe_{18} and Fe_6.

Fe_{18}			
Fe1—O3	1.982 (5)	Fe5—O25	1.957 (5)
Fe1—O4	1.941 (5)	Fe5—O27^{i}	2.081 (5)
Fe1—O23^{i}	2.057 (5)	Fe5—O33	2.322 (5)
Fe1—O25^{i}	2.180 (5)	Fe6—O1^{i}	2.001 (5)
Fe1—O26^{i}	1.994 (5)	Fe6—O3^{i}	2.176 (5)
Fe1—O27	1.990 (5)	Fe6—O27	1.942 (5)
Fe2—N3^{i}	2.180 (7)	Fe6—O27^{i}	2.084 (5)
Fe2—O2	1.941 (5)	Fe6—O28	1.975 (5)
Fe2—O3	1.910 (5)	Fe6—O33	1.963 (5)
Fe2—O28^{i}	2.012 (5)	Fe7—O3^{i}	2.059 (5)
Fe2—O29^{i}	2.137 (5)	Fe7—O10	1.967 (5)
Fe2—O32^{i}	2.003 (5)	Fe7—O24	1.977 (5)
Fe3—N1	2.199 (7)	Fe7—O25	2.138 (5)
Fe3—O1	1.832 (5)	Fe7—O29	1.958 (5)
Fe3—O2	1.996 (5)	Fe7—O33	2.067 (5)
Fe3—O4	2.053 (5)	Fe8—N4	2.197 (6)
Fe3—O5	2.079 (5)	Fe8—O19	1.969 (5)
Fe3—O7	2.052 (5)	Fe8—O20	2.020 (5)
Fe4—N2	2.171 (6)	Fe8—O23	2.044 (5)
Fe4—O5	1.948 (5)	Fe8—O24	2.050 (5)
Fe4—O9	1.995 (5)	Fe8—O25	1.944 (5)
Fe4—O10	2.134 (5)	Fe9—N5	2.179 (6)
Fe4—O12	1.990 (6)	Fe9—O14	1.938 (6)
Fe4—O33	1.901 (5)	Fe9—O15	1.964 (5)
Fe5—O1	1.905 (5)	Fe9—O17	2.161 (6)
Fe5—O9	1.961 (5)	Fe9—O19	2.004 (5)
Fe5—O14	1.947 (6)	Fe9—O26	1.929 (5)
O3—Fe1—O23^{i}	95.7 (2)	O14—Fe5—O25	89.7 (2)
O3—Fe1—O25^{i}	81.9 (2)	O14—Fe5—O27^{i}	102.5 (2)
O3—Fe1—O26^{i}	170.5 (2)	O14—Fe5—O33	172.4 (2)
O3—Fe1—O27	85.6 (2)	O25—Fe5—O9	97.8 (2)
O4—Fe1—O3	88.9 (2)	O25—Fe5—O27^{i}	83.9 (2)
O4—Fe1—O23^{i}	94.5 (2)	O25—Fe5—O33	82.7 (2)
O4—Fe1—O25^{i}	168.3 (2)	O27^{i}—Fe5—O33	77.02 (19)
O4—Fe1—O26^{i}	100.4 (2)	O27—Fe6—O1^{i}	82.1 (2)
O4—Fe1—O27	105.9 (2)	O27—Fe6—O3^{i}	163.6 (2)
O23^{i}—Fe1—O25^{i}	79.5 (2)	O27^{i}—Fe6—O3^{i}	78.57 (19)
O26^{i}—Fe1—O23^{i}	85.8 (2)	O27—Fe6—O27^{i}	86.1 (2)
O26^{i}—Fe1—O25^{i}	89.2 (2)	O27—Fe6—O28	115.1 (2)

O27—Fe1—O23^{i}	159.6 (2)	O27—Fe6—O33	103.2 (2)
O27—Fe1—O25^{i}	80.6 (2)	O28—Fe6—O1^{i}	86.9 (2)
O27—Fe1—O26^{i}	89.8 (2)	O28—Fe6—O3^{i}	79.3 (2)
O2—Fe2—N3^{i}	112.9 (2)	O28—Fe6—O27^{i}	157.2 (2)
O2—Fe2—O28^{i}	98.4 (2)	O33—Fe6—O1^{i}	171.3 (2)
O2—Fe2—O29^{i}	171.3 (2)	O33—Fe6—O3^{i}	81.71 (19)
O2—Fe2—O32^{i}	89.8 (2)	O33—Fe6—O27^{i}	85.5 (2)
O3—Fe2—N3^{i}	149.5 (2)	O33—Fe6—O28	96.8 (2)
O3—Fe2—O2	94.6 (2)	O3^{i}—Fe7—O25	81.2 (2)
O3—Fe2—O28^{i}	85.1 (2)	O3^{i}—Fe7—O33	82.2 (2)
O3—Fe2—O29^{i}	78.1 (2)	O10—Fe7—O3^{i}	160.9 (2)
O3—Fe2—O32^{i}	114.0 (2)	O10—Fe7—O24	98.1 (2)
O28^{i}—Fe2—N3^{i}	78.3 (2)	O10—Fe7—O25	102.8 (2)
O28^{i}—Fe2—O29^{i}	85.8 (2)	O10—Fe7—O33	79.6 (2)
O29^{i}—Fe2—N3^{i}	75.3 (2)	O24—Fe7—O3^{i}	101.1 (2)
O32^{i}—Fe2—N3^{i}	80.4 (2)	O24—Fe7—O25	77.9 (2)
O32^{i}—Fe2—O28^{i}	158.6 (2)	O24—Fe7—O33	161.8 (2)
O32^{i}—Fe2—O29^{i}	88.9 (2)	O29—Fe7—O3^{i}	79.0 (2)
O1—Fe3—N1	172.2 (2)	O29—Fe7—O10	99.4 (2)
O1—Fe3—O2	91.9 (2)	O29—Fe7—O24	95.3 (2)
O1—Fe3—O4	101.5 (2)	O29—Fe7—O25	157.4 (2)
O1—Fe3—O5	96.3 (2)	O29—Fe7—O33	102.9 (2)
O1—Fe3—O7	102.5 (2)	O33—Fe7—O25	85.0 (2)
O2—Fe3—N1	95.9 (2)	O19—Fe8—N4	109.3 (2)
O2—Fe3—O4	94.6 (2)	O19—Fe8—O20	89.0 (2)
O2—Fe3—O5	171.7 (2)	O19—Fe8—O23	95.0 (2)
O2—Fe3—O7	87.6 (2)	O19—Fe8—O24	171.5 (2)
O4—Fe3—N1	78.7 (2)	O20—Fe8—N4	79.1 (2)
O4—Fe3—O5	82.2 (2)	O20—Fe8—O23	154.7 (2)
O5—Fe3—N1	76.0 (2)	O20—Fe8—O24	90.5 (2)
O7—Fe3—N1	77.1 (2)	O23—Fe8—N4	76.1 (2)
O7—Fe3—O4	155.8 (2)	O23—Fe8—O24	89.1 (2)
O7—Fe3—O5	92.1 (2)	O24—Fe8—N4	78.9 (2)
O5—Fe4—N2	95.0 (2)	O25—Fe8—N4	152.7 (2)
O5—Fe4—O9	88.6 (2)	O25—Fe8—O19	92.0 (2)
O5—Fe4—O10	168.6 (2)	O25—Fe8—O20	119.3 (2)
O5—Fe4—O12	93.0 (2)	O25—Fe8—O23	85.6 (2)
O9—Fe4—N2	79.6 (2)	O25—Fe8—O24	80.8 (2)
O9—Fe4—O10	83.6 (2)	O14—Fe9—N5	164.3 (2)
O10—Fe4—N2	75.5 (2)	O14—Fe9—O15	94.6 (2)
O12—Fe4—N2	80.9 (2)	O14—Fe9—O17	86.3 (2)
O12—Fe4—O9	160.5 (2)	O14—Fe9—O19	107.3 (2)
O12—Fe4—O10	91.6 (2)	O15—Fe9—N5	79.0 (2)

O33—Fe4—N2	152.9 (2)	O15—Fe9—O17	87.1 (2)
O33—Fe4—O5	108.6 (2)	O15—Fe9—O19	154.8 (2)
O33—Fe4—O9	87.7 (2)	O17—Fe9—N5	79.2 (2)
O33—Fe4—O10	79.4 (2)	O19—Fe9—N5	76.6 (2)
O33—Fe4—O12	110.1 (2)	O19—Fe9—O17	82.1 (2)
O1—Fe5—O9	92.7 (2)	O26—Fe9—N5	95.4 (2)
O1—Fe5—O14	100.8 (2)	O26—Fe9—O14	99.6 (2)
O1—Fe5—O25	163.1 (2)	O26—Fe9—O15	97.1 (2)
O1—Fe5—O27^{i}	80.9 (2)	O26—Fe9—O17	172.4 (2)
O1—Fe5—O33	86.7 (2)	O26—Fe9—O19	91.5 (2)
O9—Fe5—O27^{i}	154.2 (2)	O26—Fe9—O17	172.4 (2)
O9—Fe5—O33	77.7 (2)	O26—Fe9—O19	91.5 (2)
O14—Fe5—O9	103.3 (2)		
Symmetry code: (i) −x+3/2, −y+1/2, −z+1.			
Fe$_6$			
Fe1—O5	1.951 (5)	Fe4—O19	1.938 (5)
Fe1—O7^{i}	2.010 (6)	Fe4—O20	1.945 (5)
Fe1—O9^{i}	2.079 (5)	Fe4—O25^{ii}	1.983 (6)
Fe1—O10^{i}	2.026 (5)	Fe4—O26^{ii}	2.080 (5)
Fe1—O15	1.958 (5)	Fe4—O29^{ii}	2.038 (5)
Fe1—N1^{i}	2.155 (6)	Fe4—N6^{ii}	2.163 (7)
Fe2—O4	1.952 (5)	Fe5—O20^{ii}	2.004 (5)
Fe2—O10	1.972 (5)	Fe5—O22^{ii}	1.994 (5)
Fe2—O11^{i}	2.022 (5)	Fe5—O28^{ii}	2.050 (5)
Fe2—O14^{i}	1.983 (5)	Fe5—O29	1.972 (5)
Fe2—O15^{i}	2.039 (5)	Fe5—O30	1.938 (5)
Fe2—N3^{i}	2.154 (6)	Fe5—N4^{ii}	2.154 (6)
Fe3—O2^{i}	1.991 (5)	Fe3—O4^{i}	2.019(5)
Fe3—O5^{i}	2.047 (5)	Fe6—O26	1.944 (5)
Fe3—O9	1.939 (5)	Fe6—O28	1.982 (5)
Fe3—O11	1.968 (5)	Fe6—O30^{ii}	2.038 (5)
Fe3—N2^{i}	2.167 (7)	Fe6—N5^{ii}	2.169 (7)
O5—Fe1—O7^{i}	99.2 (2)	O19—Fe4—O20	99.9 (2)
O5—Fe1—O9^{i}	75.6 (2)	O19—Fe4—O25^{ii}	100.2 (2)
O5—Fe1—O10^{i}	108.6 (2)	O19—Fe4—O26^{ii}	76.3 (2)
O5—Fe1—O15	99.8 (2)	O19—Fe4—O29^{ii}	107.0 (2)
O5—Fe1—N1^{i}	151.3 (2)	O19—Fe4—N6^{ii}	154.0 (2)
O7^{i}—Fe1—O9^{i}	100.4 (2)	O20—Fe4—O25^{ii}	97.5 (2)
O7^{i}—Fe1—O10^{i}	152.1 (2)	O20—Fe4—O26^{ii}	162.1 (2)
O7^{i}—Fe1—N1^{i}	79.3 (2)	O20—Fe4—O29^{ii}	76.4 (2)
O9^{i}—Fe1—N1^{i}	76.5 (2)	O20—Fe4—N6^{ii}	105.8 (2)
O10^{i}—Fe1—O9^{i}	88.6 (2)	O25^{ii}—Fe4—O26^{ii}	100.4 (2)
O10^{i}—Fe1—N1^{i}	77.2 (2)	O25^{ii}—Fe4—O29^{ii}	152.7 (2)

O15—Fe1—O7^{i}	97.0 (2)	O25^{ii}—Fe4—N6^{ii}	80.2 (3)
O15—Fe1—O9^{i}	162.4 (2)	O26^{ii}—Fe4—N6^{ii}	78.2 (2)
O15—Fe1—O10^{i}	76.7 (2)	O29^{ii}—Fe4—O26^{ii}	87.9 (2)
O15—Fe1—N1^{i}	108.8 (2)	O29^{ii}—Fe4—N6^{ii}	76.2 (2)
O4—Fe2—O10	99.0 (2)	O20^{ii}—Fe5—O28^{ii}	88.0 (2)
O4—Fe2—O11^{i}	77.2 (2)	O20^{ii}—Fe5—N4^{ii}	77.1 (2)
O4—Fe2—O14^{i}	97.2 (2)	O22^{ii}—Fe5—O20^{ii}	101.5 (2)
O4—Fe2—O15^{i}	162.7 (2)	O22^{ii}—Fe5—O28^{ii}	152.1 (2)
O4—Fe2—N3^{i}	108.5 (2)	O22^{ii}—Fe5—N4^{ii}	79.2 (2)
O10—Fe2—O11^{i}	108.8 (2)	O28^{ii}—Fe5—N4^{ii}	77.5 (2)
O10—Fe2—O14^{i}	98.8 (2)	O29—Fe5—O20^{ii}	76.6 (2)
O10—Fe2—O15^{i}	76.1 (2)	O29—Fe5—O22^{ii}	94.7 (2)
O10—Fe2—N3^{i}	152.6 (2)	O29—Fe5—O28^{ii}	113.1 (2)
O11^{i}—Fe2—O15^{i}	88.7 (2)	O29—Fe5—N4^{ii}	151.1 (2)
O11^{i}—Fe2—N3^{i}	77.1 (2)	O30—Fe5—O20^{ii}	164.7 (2)
O14^{i}—Fe2—O11^{i}	152.4 (2)	O30—Fe5—O22^{ii}	93.7 (2)
O14^{i}—Fe2—O15^{i}	99.9 (2)	O30—Fe5—O28^{ii}	77.7 (2)
O14^{i}—Fe2—N3^{i}	79.3 (2)	O30—Fe5—O29	103.8 (2)
O15^{i}—Fe2—N3^{i}	77.3 (2)	O30—Fe5—N4^{ii}	104.8 (2)
O2^{i}—Fe3—O4^{i}	102.0 (2)	O17^{ii}—Fe6—O19^{ii}	153.3 (2)
O2^{i}—Fe3—O5^{i}	152.1 (2)	O17^{ii}—Fe6—O30^{ii}	101.3 (2)
O2^{i}—Fe3—N2^{i}	78.9 (2)	O17^{ii}—Fe6—N5^{ii}	78.7 (2)
O4^{i}—Fe3—O5^{i}	86.0 (2)	O19^{ii}—Fe6—N5^{ii}	77.6 (2)
O4^{i}—Fe3—N2^{i}	75.8 (2)	O26—Fe6—O17^{ii}	98.7 (2)
O5^{i}—Fe3—N2^{i}	77.3 (2)	O26—Fe6—O19^{ii}	77.1 (2)
O9—Fe3—O2^{i}	98.4 (2)	O26—Fe6—O28	101.7 (2)
O9—Fe3—O4^{i}	159.6 (2)	O26—Fe6—O30^{ii}	160.0 (2)
O9—Fe3—O5^{i}	76.6 (2)	O26—Fe6—N5^{ii}	107.5 (2)
O9—Fe3—O11	99.4 (2)	O28—Fe6—O17^{ii}	95.0 (2)
O9—Fe3—N2^{i}	110.1 (2)	O28—Fe6—O19^{ii}	111.6 (2)
O11—Fe3—O2^{i}	96.0 (2)	O28—Fe6—O30^{ii}	77.0 (2)
O11—Fe3—O4^{i}	76.9 (2)	O28—Fe6—N5^{ii}	150.7 (2)
O11—Fe3—O5^{i}	111.9 (2)	O30^{ii}—Fe6—O19^{ii}	84.7 (2)
O11—Fe3—N2^{i}	150.4(2)	O30^{ii}—Fe6—N5^{ii}	76.3 (2)
Symmetry codes: (i) −x+2, −y+1, −z+1; (ii) −x+1, −y+2, −z.			

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