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Structural Studies on the Rare Earth Carboxylates

10. The Crystal and Molecular Structure of Monoclinic Trisodium Tris-(pyridine-2,6-dicarboxylato)ytterbate(III) 13-Hydrate

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The crystal and molecular structure of $Na_3[Yb(C_7H_3NO_4)_3].13H_2O$ has been determined from three-dimensional X-ray intensity data collected with the Weissenberg multi-film technique. Four formula units crystallize in a monoclinic cell with the dimensions $a = 9.729(5)$ Å, $b = 19.245(15)$ Å, $c = 18.175(12)$ Å, and $\beta = 91.40(6)°$. The space group is $P2_1/c$. The elements Ho – Tm form isomorphous compounds. These are stable but the ytterbium compound is metastable and passes into an orthorhombic phase when stored in the mother liquor. The investigated monoclinic phase contains mononuclear tris(pyridine-2,6-dicarboxylato) complexes. In these the lanthanoid ion is surrounded by six carboxylate oxygen atoms and three nitrogen atoms which form a tri-capped trigonal prism with Yb – O and Yb – N bond distances in the ranges 2.34 – 2.43 Å and 2.50 – 2.53 Å, respectively. The lanthanoid complexes are held together in columns parallel with the *c* axis by the sodium ions. These columns are connected by hydrogen bonds *via* water molecules. Three of the thirteen water molecules have not been located but are assumed to be occluded in the fairly large cavities in the structure.

The tridentate ligands pyridine-2,6-dicarboxylate (or dipicolinate) and oxydiacetate are similar and form mononuclear complexes of the composition $[ML_3]^{3-}$ with the trivalent lanthanoid ions. The present investigation is part of a systematic study of some crystal structures of the sodium salts formed with these complexes. The structures of the lanthanoid oxydiacetates $Na_3[M(C_4H_4O_5)_3].2NaClO_4.6H_2O$, M = Ce – Lu, and of the orthorhombic ytterbium dipicolinate $Na_3[Yb(C_7H_3NO_4)_3].14H_2O$, denoted ORTYBDIPIC, have been reported earlier.[1–3]

All the structures in the oxydiacetate series are isomorphous, but with the dipicolinate ion several phases are formed. The elements Ce – Dy give compounds with almost identical triclinic structures, but hexagonal, monoclinic, and orthorhombic phases are obtained for the elements Ho – Lu.

Which phase is formed depends upon the method of preparation and the lanthanoid ion used.

This paper is a report of the crystal and molecular structure of the monoclinic ytterbium compound trisodium tris(dipicolinato)ytterbate 13-hydrate, $Na_3[Yb(C_7H_3NO_4)_3].13H_2O$, subsequently denoted MONYBDIPIC. The same phase is formed by the elements Ho – Tm. The variation of the unit cell dimensions in this series is also studied. Two following papers will deal with the hexagonal and triclinic lanthanoid dipicolinate phases represented by $Na_3[Yb(C_7H_3NO_4)_3].NaClO_4.10H_2O$ and $Na_3[Nd(C_7H_3NO_4)_3].15H_2O$, respectively.

EXPERIMENTAL

The monoclinic lanthanoid dipicolinates $Na_3[M(C_7H_3NO_4)_3].13H_2O$, M = Ho – Yb, were prepared by mixing water solutions of the lanthanoid nitrates and disodium dipicolinate in the molar ratio 1 : 3. The pH of the resulting solutions had values near 7. Slow evaporation at room temperature gave prismatic crystals, which were stored in the mother liquor to prevent efflorescing. The ytterbium compound MONYBDIPIC was analysed for Yb, N, C, H, and H_2O as described for ORTYBDIPIC.[2] The relative amounts found are compared with those calculated for $Na_3[Yb(C_7H_3NO_4)_3].13H_2O$, F. W. 971.5.

	Yb	N	C	H	H_2O
Found	17.9	4.5	27.2	3.5	23.8 (%)
Calc.	17.8	4.3	26.0	3.6	24.1 (%)

In the structure determination only 10 H_2O per Yb were located.

The holmium, erbium, and thulium compounds prepared as described above gave the same powder pattern as MONYBDIPIC.

MONYBDIPIC is a metastable ytterbium dipicolinate phase. Despite this, several samples prepared using either ytterbium nitrate and disodium dipicolinate or ytterbium hydroxide, dipicolinic acid, and sodium hydroxide resulted in MONYBDIPIC. These crystals were kept in the mother liquor for 1/2 – 2 years. Powder photographs then revealed that they all had been transformed to ORTYBDIPIC. Using both methods of preparation only the monoclinic phase has been obtained with holmium, erbium, and thulium. With lutetium only the orthorhombic phase was obtained.

Powder photographs were taken as described for ORTYBDIPIC in a Guinier-Hägg focusing camera using Cu$K\alpha$-radiation ($\lambda = 1.54178$ Å) for the erbium and ytterbium compounds. These films were used for determination of unit cell dimensions.

The intensity data of MONYBDIPIC were recorded with integrated Weissenberg multi-film technique. Ni-filtered Cu-radiation was used. Two freshly prepared crystals were coated with canada balsam and mounted along the a and c axes, respectively. Both crystals were prismatic with the approximate dimensions $0.07 \times 0.07 \times 0.15$ mm³. They were elongated in the mounting direction. After about 25 days the crystal had decomposed. The relative intensities of the reflexions $0kl - 7kl$ from crystal 1 were measured visually by comparison with a calibrated scale, and those of the reflexions $hk0 - hk10$ from crystal 2 by a flying spot integrating microdensitometer (manufactured by Joyce, Loeble and Co., Gateshead, England). The majority of the reflexions were too weak to be measured. 2300 independent intensities, representing about 35 % of the possible number in the recorded reciprocal region, were used in the refinement of the structure.

The intensity data were corrected for Lorentz and polarization effects. The linear absorption coefficient, μ, is 65 cm^{-1}. Because of this low value and the small crystals used, no absorption corrections were applied.

Table 1. X-Ray powder data: observed and calculated values of $10^5 \times \sin^2 \theta$ for the compounds $Na_3[M(C_7H_3NO_4)_3].13H_2O$, M = Er and Yb. The observed powder intensities of the ytterbium compound are also given.

h	k	l	Er obs	Er calc	Yb obs	Yb calc	I_{obs} Yb
0	1	1	330	338	339	341	m
1	0	0	610	624	622	628	m
0	2	0	630	636	638	642	s
0	0	2	715	716	722	720	w
1	1	0	776	783	784	789	w
0	2	1	815	815	820	822	vw
0	1	2	872	875	882	881	vs
1	2	0	1257	1260	1277	1270	s
1	0	-2	1306	1303	1322	1310	w
1	2	1	1460	1458	1479	1469	s
1	1	-2		1462		1471	
1	1	2	1535	1536	1554	1547	m
0	3	1	1615	1610	1627	1624	w
0	3	2	2169	2147	2187	2164	w
1	3	-1	2236	2216	2249	2233	w
0	2	3		2247		2262	
1	3	1	2258	2252	2287	2272	vw
2	0	0	2463	2496	2493	2513	vvw
0	4	0	2494	2544	2532	2567	vvw
2	1	0	2658	2655	2691	2674	w
0	4	1	2750	2723	2775	2747	w
1	3	-2		2734		2754	
1	2	-3	2845	2816	2862	2833	s
0	0	4		2864		2881	
2	1	1	2894	2871	2921	2892	m
1	2	3		2927		2948	
2	0	-2	3153	3138	3181	3157	m
1	4	0		3168		3196	
1	4	-1	3333	3329	3356	3357	vvw
2	2	1		3348		3374	
1	0	-4	3411	3414	3433	3433	m
2	1	2	3459	3445	3493	3471	m
0	2	4	-	3500		3523	
1	3	-3	3612	3611	3643	3636	m
2	2	-2	3767	3774	3805	3799	vw
2	2	2	3892	3922	3926	3952	vvw
1	4	2		3921		3954	
1	2	-4	3968	4050	3995	4075	w
2	3	-1	4110	4070	4144	4099	s
2	3	1		4143		4176	
1	2	4	4213	4198	4242	4227	vw
1	5	-1	4763	4760	4796	4797	m
1	5	1	4822	4797	4853	4838	vvw
0	2	5	5076	5111	5127	5143	vw
1	5	-2	5317	5279	5352	5322	m
1	1	5		5351		5386	
2	3	-3	5436	5428	5491	5463	s
0	5	3	5571	5587	5615	5632	w
3	1	-1	5905	5899	5938	5939	w
0	3	5		5906		5945	
3	1	1	6018	6010	6043	6053	w
1	6	0	6355	6349	6414	6405	vw
3	2	-1		6376		6420	
2	4	-3	6562	6541	6603	6587	vvw
1	6	1		6546		6604	
2	3	-4	6660	6644	6688	6686	w
2	5	1	-	6688	6776	6743	w
2	4	3	6742	6762		6815	
0	5	4	6872	6840	6897	6892	w
3	2	-2		6558		6903	
1	6	2	7127	7102	7174	7163	vvw
2	5	-2		7114		7169	
3	1	-3	7232	7220	7275	7264	m
2	2	-5	7466	7422	7512	7466	w
3	3	2	7872	7875	7925	7934	vw
0	3	6		7876		7926	

h	k	l	Er obs	Er calc	Yb obs	Yb calc	I_{obs} Yb
1	6	3	8029	8015	-	8082	
3	2	3		8030	8096	8090	m
2	4	4	-	8053		8114	
3	4	0	8173	8161	8217	8223	w
1	7	-1	8578	8577	8640	8652	m
2	3	5		8588		8650	
2	0	-6	8741	8719	8789	8766	w
2	1	-6	8873	8878	-	8926	
2	6	-2	-	8863	8950	8934	m
3	2	-4		8895		8949	
3	4	2	-	8988	9073	9058	vvw
2	6	2	9020	9011		9086	
2	4	-5	9333	9331	9397	9391	vvw
3	2	4		9339		9407	
2	5	4	9510	9484	9548	9559	m
1	4	-6		9502		9563	
3	5	0	9602	9592	9667	9667	vw
3	4	-3		9606		9671	
3	5	1	9875	9826	9914	9904	m
1	2	-7		9902	-	9959	
1	7	3	10132	10083	10186	10168	w
3	3	4		10134		10210	
4	1	-1	10284	10249	10331	10318	w
2	7	0		10288		10376	
0	8	2	10901	10893	11051	10989	vvw
1	6	5		10916		11001	
1	8	1	10991	10999	-	11097	
3	2	5		11005		11115	
2	1	-7	-	11168	11229	11229	w
1	5	6		11155		11236	
0	4	7	11340	11316	11388	11390	vvw
3	6	0		11341		11432	
4	3	0	11432	11416	11505	11498	w
0	0	8		11457		11523	
4	3	-1	11541	11521	11617	11602	m
4	1	-3		11533		11606	
2	4	6	-	11707	11791	11792	w
3	0	-6	11767	11728		11793	
0	8	3		11788	-	11889	
3	1	-6	11901	11887	11958	11954	vvw
1	0	-8		11933	-	11999	
3	6	2	12205	12168	12266	12267	w
0	6	6		12169		12258	
4	0	-4	-	12553	12636	12629	w
1	2	-8	12603	12569		12640	
4	4	-1		12634	-	12725	
0	5	7	12754	12747	12825	12834	vvw
1	6	6	-	12904	13002	13001	vw
3	4	5	12948	12913		13011	
2	3	7		12958	-	13047	
2	7	-4	13029	13004	13125	13104	vvw
3	2	6		13030		13123	
4	4	-2	13114	13098	13206	13189	vw
3	6	3		13119		13224	
1	5	-7	13246	13242	13330	13328	vw
4	3	3		13249		13348	
0	9	2	13637	13597	-	13717	
3	7	1		13643		13755	
1	9	1	-	13702	13830	13824	vvw
1	8	4		13739		13855	
4	4	-3	-	13919	14005	14013	w
4	5	-1	14098	14066	14180	14169	w
2	4	7		14071		14171	
3	7	-3	14870	14853	14957	14966	w
2	2	8		14885		14984	

UNIT CELL AND SPACE GROUP

MONYBDIPIC, and thus the isomorphous Ho – Tm dipicolinates, crystallize in the Laue class $2/m$. The systematically absent reflexions are $h0l$: $l \neq 2n$, and $0k0$: $k \neq 2n$ which is only compatible with space group $P2_1/c$ (No. 14).

Preliminary unit cell dimensions were obtained from Weissenberg and oscillation photographs of MONYBDIPIC. They were improved for the erbium and ytterbium compounds by a series of least-squares treatments of the powder data minimizing $\sum w(\sin^2 \theta_o - \sin^2 \theta_c)^2$ with weights $w = 1/\sin^2 2\theta_o$. The observed powder patterns are given in Table 1. The following unit cell dimensions with estimated standard deviations were obtained for $Na_3[M(C_7H_3NO_4)_3].13H_2O$, M = Er and Yb.

	a/Å	b/Å	c/Å	β/°	V/Å^3
Er	9.762(7)	19.332(19)	18.227(16)	91.46(8)	3439(6)
Yb	9.729(5)	19.245(15)	18.175(12)	91.50(6)	3402(4)

For MONYBDIPIC the density 1.9 g/cm^3 was estimated by flotation. With four formula units in the cell the calculated density is 1.90 g/cm^3.

DETERMINATION AND REFINEMENT OF THE STRUCTURE

The position of the ytterbium ion in MONYBDIPIC was determined from the Harker vectors in a three-dimensional Patterson synthesis computed using the reflexions from crystal 2. After a least-squares refinement of the preliminary ytterbium parameters and the inter-layer scale factors, a three-dimensional difference synthesis was calculated. The nonhydrogen atoms of the ligands were located by fitting a model of the tris(dipicolinato)ytterbate complex found in ORTYBDIPIC [2] to the peaks in the obtained electron density maps. The position of the three sodium ions and of eight water oxygen atoms, coordinated to the sodium ions, were also deduced.

A series of least-squares refinements using the intensity material obtained from crystal 1 was computed. Because of the large number of parameters the diagonal approximation described in Ref. 2 was used in the preliminary refinements. After eight cycles the discrepancy indices $R = \sum||F_o| - |F_c||/\sum|F_o|$ and $wR = [\sum w(|F_o| - |F_c|)^2/\sum w|F_o|^2]^{\frac{1}{2}}$ had decreased from $R = 0.30$, $wR = 0.33$, to $R = 0.17$, $wR = 0.20$. A second difference synthesis indicated that the ytterbium ion vibrates anisotropically. Three cycles of full-matrix least-squares refinements of only the inter-layer scale factors and the ytterbium parameters using anisotropic temperature factors gave $R = 0.14$, $wR = 0.17$. From the second difference synthesis it was also possible to deduce the positions of the ninth and tenth water oxygen atoms.

The intensity data from crystal 1 and 2 were brought together. 218 structural parameters including anisotropic temperature factors for the ytterbium ion were simultaneosuly improved with full-matrix least-squares refinements. After seven cycles the discrepancy indices had converged to $R = 0.102$, $wR = 0.093$. In the seventh cycle the shifts in the parameters were less than 1/3 of the estimated standard deviations except for some temperature factors which had shifts in the range 50 – 60 %.

The isotopic temperature factors, B, obtained for Na(3) in the refinement is 7.8 Å^2 compared with 3.2 and 4.4 Å^2 for Na(1) and Na(2), respectively. The water oxygen atoms O(19) – O(22) have also large values of B, in the range 8.2 – 10.4 Å^2. The intensity data from crystals 1 and 2 were obtained by two different methods of measurement (*cf.* p. 986). The values of the temperature factors might depend upon the method used. As the data from crystal 1 gave values similar to those obtained when the whole body of data was used, this dependence cannot be too significant. However, there are at least two other plausible interpretations of the large temperature factors.

Table 2. Positional parameters and isotropic temperature factors in MONYBDIPIC with estimated standard deviations. The space group is $P2_1/c$ (No. 14).[4]

Atom	x	y	z	$B/Å^2$
Yb	0.0537(01)	0.21748(5)	0.10954(5)	–
N(1)	0.0662(15)	0.1709(07)	0.2397(07)	1.6(2)
C(1)	–0.1171(29)	0.2570(13)	0.2595(14)	4.7(5)
C(2)	–0.0157(22)	0.2006(08)	0.2921(10)	2.4(4)
C(3)	0.0244(27)	0.1877(12)	0.3667(13)	4.1(5)
C(4)	0.1188(23)	0.1408(10)	0.3826(11)	3.4(4)
C(5)	0.1950(23)	0.1065(11)	0.3293(12)	3.6(4)
C(6)	0.1578(21)	0.1236(09)	0.2542(10)	2.2(3)
C(7)	0.2267(22)	0.0937(10)	0.1887(11)	2.9(4)
O(1)	–0.0973(15)	0.2670(06)	0.1929(07)	2.5(2)
O(2)	–0.1821(18)	0.2916(07)	0.3066(09)	4.2(3)
O(3)	0.1968(13)	0.1202(06)	0.1237(06)	2.3(2)
O(4)	0.3170(21)	0.0474(10)	0.2016(10)	5.6(4)
N(2)	0.0310(15)	0.1453(06)	–0.0045(07)	1.4(2)
C(8)	0.2361(24)	0.2047(10)	–0.0376(12)	3.3(4)
C(9)	0.1217(22)	0.1503(10)	–0.0555(10)	2.7(4)
C(10)	0.1273(23)	0.1145(11)	–0.1178(11)	3.1(4)
C(11)	0.0191(22)	0.0631(10)	–0.1285(10)	2.6(4)
C(12)	–0.0871(26)	0.0594(11)	–0.0752(12)	3.5(4)
C(13)	–0.0683(20)	0.1031(08)	–0.0144(09)	1.7(3)
C(14)	–0.1695(22)	0.1001(10)	0.0510(11)	2.9(4)
O(5)	0.2230(16)	0.2420(07)	0.0214(08)	3.2(3)
O(6)	0.3348(16)	0.2087(07)	–0.0783(08)	3.4(3)
O(7)	–0.1317(16)	0.1390(07)	0.1048(07)	3.4(3)
O(8)	–0.2802(16)	0.0672(07)	0.0431(08)	3.4(3)
N(3)	0.0924(29)	0.3459(14)	0.1188(14)	7.1(6)
C(15)	–0.1096(26)	0.3461(11)	0.0356(12)	3.7(4)
C(16)	–0.0120(25)	0.3840(11)	0.0815(12)	3.4(4)
C(17)	–0.0057(33)	0.4550(15)	0.0900(17)	6.0(6)
C(18)	0.0761(34)	0.4914(16)	0.1383(17)	6.4(7)
C(19)	0.1835(27)	0.4462(11)	0.1775(13)	4.0(4)
C(20)	0.1807(28)	0.3742(12)	0.1580(13)	4.2(5)
C(21)	0.2781(26)	0.3206(12)	0.1948(12)	3.4(4)
O(9)	–0.1040(15)	0.2775(07)	0.0331(07)	3.2(3)
O(10)	–0.1974(20)	0.3819(09)	–0.0046(10)	5.7(4)
O(11)	0.2548(16)	0.2571(07)	0.1802(08)	3.1(3)
O(12)	0.3852(21)	0.3428(10)	0.2323(11)	5.9(4)
Na(1)	0.5383(09)	0.0510(04)	0.1334(04)	–
Na(2)	0.5298(10)	0.1339(04)	–0.0314(05)	–
Na(3)	0.5123(15)	0.3068(07)	–0.0924(08)	–
O(13)	0.4981(18)	–0.0783(08)	0.1498(09)	4.8(4)
O(14)	0.7034(21)	0.0887(10)	0.2331(11)	6.5(5)
O(15)	0.4998(17)	0.1785(08)	0.1006(08)	4.4(3)
O(16)	0.3756(15)	0.0547(07)	0.0248(07)	2.9(2)
O(17)	0.6866(17)	0.2249(07)	–0.0599(08)	3.9(3)
O(18)	0.4280(22)	0.3489(10)	0.0170(11)	6.2(4)
O(19)	0.6283(28)	0.4043(13)	–0.1231(13)	–
O(20)	0.5006(30)	0.2930(13)	–0.2108(14)	–
O(21)	0.6268(22)	0.2984(13)	0.1438(13)	–
O(22)	0.7079(27)	0.4124(13)	0.2507(15)	–

Table 3. Anisotropic temperature factor parameters $\beta_{ij} \times 10^4$ with estimated standard deviations. The expression used is $\exp[-(h^2\beta_{11}+hk\beta_{12}+\cdots)]$. Root-mean-square components R_i along principal axes of the ellipsoids of thermal vibration calculated from the values of β_{ij} are also given.

Atom	β_{11}	β_{22}	β_{33}	β_{12}	β_{13}	β_{23}	R_1/Å	R_2/Å	R_3/Å
Yb	53(02)	23(00)	19(00)	−6(01)	19(01)	−6(01)	0.141	0.181	0.214
Na(1)	98(12)	21(02)	24(02)	3(08)	16(10)	−3(04)	0.187	0.203	0.225
Na(2)	134(15)	28(03)	31(03)	8(10)	12(11)	4(05)	0.219	0.230	0.259
Na(3)	216(22)	54(05)	71(06)	63(16)	15(20)	40(09)	0.210	0.283	0.444
O(19)	309(45)	68(11)	55(09)	−87(37)	19(36)	13(17)	0.284	0.327	0.425
O(20)	329(51)	61(11)	60(10)	−3(36)	68(40)	−27(16)	0.276	0.355	0.412
O(21)	136(31)	91(13)	56(09)	−16(31)	−70(29)	−14(18)	0.205	0.341	0.416
O(22)	255(44)	69(11)	82(13)	42(36)	91(41)	8(20)	0.286	0.370	0.410

Firstly, they, as well as the somewhat large shifts mentioned above, might depend on the incomplete data set used (*cf.* Ref. 2). Secondly, there might be a slight disorder of Na(3) and O(19) − O(22) causing their temperature factors to be large. The latter interpretation was chosen (*cf.* Discussion, p. 1002) and three cycles of least-squares refinement of the parameters of Yb, Na(1) − Na(3), and O(19) − O(22) were computed using anisotropic temperature factors. In this way one might obtain some conception of the range in which these atoms could be located. Na(1) and Na(2) were included in the refinement for comparison. The result is shown in Fig. 3. The discrepancy indices converged to $R=0.099$ and $w\mathrm{R}=0.091$.

Table 2 gives the final atomic positions and the isotropic temperature factors with estimated standard deviations. The anisotropic temperature factors are given in Table 3 together with the root-mean-square components along the principal axes of the ellipsoids of "thermal vibration". A three-dimensional difference synthesis based upon the parameters of Tables 2 and 3 showed only small spurious peaks above the background. The highest one, about 2 e/Å^3, was situated at the ytterbium position.

In the least-squares refinements $\sum w(|F_o|-|F_c|)^2$ were minimized. The weights w were calculated according to the expression $w=1/(a+|F_o|+c|F_o|^2+ +d|F_o|^3)$. An analysis of the weighting scheme suggested suitable values for a, c, and d. In the last cycle of refinement the values $a=35$, $c=0.015$, and $d=0.0002$ were used. Reflexions not obeying the condition $A \leq |F_o|/|F_c| \leq B$ were given zero weight. In the earlier stages of the refinement the values $B=1/A=3$ were used but when the intensities from crystals 1 and 2 had been brought together these values were changed to $B=1/A=1.25$.

The atomic scattering factors used in the calculations were taken from the *International Tables* [5] (Na^+, O, N, and C) and from Cromer *et al.*[6] (Yb). Observed and calculated structure factors are compared in Table 4.

Selected interatomic distances and angles in the structure are given in Table 5. The standard deviations are calculated from the estimated standard deviations of the atomic coordinates and the unit cell dimensions.

Table 4. Observed and calculated structure factors of MONYBDIPIC. The 247 reflexions not obeying the condition $0.80 \leq |F_o|/|F_c| \leq 1.25$ are denoted by asterisks.

h	k	l	$|F_o|$	$|F_c|$
0	0	-22	43	46
0	0	-16	53	26 *
0	0	-14	179	157
0	0	-12	122	111
0	0	-10	179	163
0	0	-8	250	217
0	0	-6	96	99
0	0	-4	246	215
0	0	-2	145	107 *
0	-1	-22	39	35
0	-1	-16	137	118
0	-1	-13	57	55
0	-1	-12	188	175
0	-1	-11	51	46
0	-1	-10	146	126
0	-1	-9	40	45
0	-1	-8	117	105
0	-1	-6	174	175
0	-1	-4	21	22
0	-1	-3	31	29
0	-1	-2	338	291
0	-2	-18	68	72
0	-2	-15	57	69
0	-2	-14	156	137
0	-2	-12	42	45
0	-2	-11	92	90
0	-2	-10	191	173
0	-2	-9	41	27 *
0	-2	-8	60	71
0	-2	-7	208	197
0	-2	-6	178	139 *
0	-2	-5	78	83
0	-2	-4	96	120 *
0	-2	-3	30	45 *
0	-2	-2	29	47 *
0	-2	-1	97	99
0	-3	-20	60	60
0	-3	-19	54	46
0	-3	-17	53	57
0	-3	-16	70	81
0	-3	-15	57	55
0	-3	-13	69	72
0	-3	-12	170	147
0	-3	-11	52	46
0	-3	-10	71	82
0	-3	-9	102	84
0	-3	-6	242	213
0	-3	-5	184	189
0	-3	-4	44	47
0	-3	-3	30	26
0	-3	-2	132	137
0	-3	-1	112	83 *
0	-4	21	53	50
0	-4	-17	69	58
0	-4	-14	61	67
0	-4	-13	70	71
0	-4	-11	63	70
0	-4	-10	106	96
0	-4	-9	66	55
0	-4	-8	43	36
0	-4	-7	352	304
0	-4	-6	56	60
0	-4	-5	154	114 *
0	-4	-4	104	153
0	-4	-3	145	137
0	-4	-2	28	40 *
0	-4	-1	79	75
0	-5	-19	54	53
0	-5	-17	69	63
0	-5	-15	63	67
0	-5	-13	59	62
0	-5	-12	122	114
0	-5	-11	89	78
0	-5	-10	55	66
0	-5	-9	102	114
0	-6	-21	49	55
0	-5	-5	212	213
0	-5	-4	34	28
0	-5	-2	128	144
0	-5	-1	30	69 *
0	-6	-17	63	67
0	-6	-13	60	49
0	-6	-11	170	173
0	-6	-9	45	40
0	-6	-7	147	161
0	-6	-4	44	41
0	-6	-3	146	167
0	-6	-2	44	15 *
0	-6	-1	65	97 *
0	-6	0	164	176
0	-7	-19	51	43
0	-7	-17	57	50
0	-7	-15	49	49
0	-7	-13	90	84
0	-7	-9	133	132
0	-7	-5	172	180
0	-7	-3	70	95 *
0	-7	-1	243	236
0	-8	-21	52	50
0	-8	-19	47	39
0	-8	-17	90	75
0	-8	-15	70	74
0	-8	-13	48	47
0	-8	-11	151	139
0	-8	-7	154	148
0	-8	-6	46	49
0	-8	-5	37	38
0	-8	-4	62	65
0	-8	-3	150	150
0	-8	-2	58	53
0	-8	-1	117	109
0	-8	0	30	31
0	-9	-19	60	58
0	-9	-17	50	55
0	-9	-15	64	56
0	-9	-13	104	100
0	-9	-9	129	130
0	-9	-5	163	173
0	-9	-4	45	46
0	-9	-3	48	45
0	-9	-1	99	106
0	-10	-17	46	46
0	-10	-15	63	58
0	-10	-11	101	96
0	-10	-7	164	174
0	-10	-6	56	62
0	-10	-5	82	99
0	-10	-4	95	103
0	-10	-3	75	85
0	-10	-1	67	67
0	-11	-19	42	36
0	-11	-13	64	66
0	-11	-9	103	103
0	-11	-6	140	148
0	-11	-5	134	141
0	-11	-2	41	44
0	-11	-1	150	147
0	-12	-17	47	35 *
0	-12	-14	48	52
0	-12	-12	49	49
0	-12	-11	69	74
0	-12	-10	74	81
0	-12	-8	60	66
0	-12	-7	114	102
0	-12	-6	47	50
0	-12	-5	43	47
0	-12	-4	140	135
0	-12	-2	68	62
0	-13	-16	55	55
0	-13	-12	58	65
0	-13	-10	63	62
0	-13	-6	122	140
0	-13	-5	63	59
0	-13	-2	88	97
0	-13	-1	81	57 *
0	-14	-14	59	59
0	-14	-12	58	51
0	-14	-10	59	60
0	-14	-8	58	63
0	-14	-6	56	55
0	-14	-4	87	94
0	-14	-2	58	63
0	-14	0	124	135
0	-15	-16	68	63
0	-15	-12	61	63
0	-15	-10	58	55
0	-15	-8	64	66
0	-15	-6	81	83
0	-15	-4	48	46
0	-15	-2	61	65
0	-16	-14	63	51
0	-16	-10	62	63
0	-16	-8	49	36 *
0	-16	-6	54	51
0	-16	-4	63	58
0	-16	0	74	72
0	-17	-16	40	41
0	-17	-12	51	47
0	-18	-14	43	40
0	-18	-10	46	33 *
0	-18	-6	48	39
0	-18	-4	63	65
0	-19	-12	47	35 *
0	-19	-8	42	32 *
0	-19	-2	62	57
0	-20	-11	37	32
0	-21	-9	41	38
0	-21	-8	44	31 *
0	-21	-5	60	57
0	-21	-3	46	46
0	-22	-7	52	50
0	-23	-5	37	36
1	6	-21	74	71
1	4	-21	43	35
1	5	-20	45	48
1	3	-20	45	47
1	2	-20	49	50
1	1	-20	49	54
1	0	-20	49	58
1	11	-19	52	58
1	9	-19	63	65
1	7	-19	56	63
1	5	-19	57	59
1	3	-19	44	45
1	14	-18	42	47
1	13	-18	39	35
1	12	-18	43	42
1	2	-18	60	66
1	1	-18	55	52
1	0	-18	103	116
1	17	-16	41	43
1	15	-16	46	47
1	13	-16	58	60
1	4	-16	67	67
1	3	-16	47	49
1	0	-16	99	30 *
1	11	-15	54	49
1	10	-15	55	52
1	9	-15	88	91
1	7	-15	47	88 *
1	5	-15	88	100
1	3	-15	60	59
1	2	-15	60	56
1	18	-14	45	47
1	16	-14	63	71
1	14	-14	61	66
1	12	-14	65	73
1	10	-14	47	62 *
1	6	-14	46	48
1	4	-14	106	96
1	2	-14	129	127
1	0	-14	144	134
1	12	-13	61	65
1	11	-13	47	43
1	10	-13	86	83
1	9	-13	95	95
1	8	-13	71	69
1	7	-13	106	90
1	6	-13	58	59
1	5	-13	74	73
1	2	-13	55	56
1	17	-12	53	49
1	15	-12	59	65
1	13	-12	101	93
1	11	-12	56	62
1	10	-12	46	33 *
1	6	-12	56	42 *
1	5	-12	65	68
1	3	-12	128	124
1	2	-12	57	54
1	1	-12	145	145
1	12	-11	47	49
1	11	-11	76	89
1	10	-11	116	100
1	9	-11	57	67
1	8	-11	66	66
1	7	-11	90	86
1	6	-11	63	69
1	5	-11	105	88
1	4	-11	141	131
1	3	-11	42	46
1	2	-11	35	25 *
1	1	-11	53	49
1	18	-10	57	49
1	16	-10	77	74
1	14	-10	79	88
1	12	-10	86	96
1	11	-10	45	48
1	8	-10	41	36
1	6	-10	83	74
1	5	-10	78	66
1	4	-10	79	75
1	3	-10	71	70
1	2	-10	144	132
1	0	-10	202	191
1	21	-9	47	43
1	13	-9	50	52
1	12	-9	42	39
1	11	-9	103	104
1	10	-9	71	70
1	9	-9	158	150
1	8	-9	110	98
1	7	-9	137	127
1	6	-9	86	88
1	5	-9	139	133
1	4	-9	53	50
1	3	-9	163	172
1	1	-9	64	59
1	19	-8	57	57
1	17	-8	65	74
1	15	-8	73	72
1	13	-8	93	108
1	11	-8	84	83
1	8	-8	77	73
1	5	-8	78	76
1	3	-8	196	174
1	2	-8	136	104 *
1	1	-8	218	194
1	0	-8	86	99
1	22	-7	50	44
1	20	-7	49	50
1	14	-7	46	46
1	12	-7	96	100
1	10	-7	153	143
1	9	-7	49	29 *
1	8	-7	206	195
1	6	-7	181	164
1	5	-7	34	38
1	4	-7	188	159
1	2	-7	117	108
1	20	-6	53	42 *
1	18	-6	60	58
1	16	-6	73	68
1	15	-6	60	65
1	14	-6	97	97
1	12	-6	78	75
1	7	-6	46	49
1	6	-6	88	85
1	5	-6	144	114 *
1	4	-6	47	40
1	2	-6	37	60 *
1	1	-6	133	125
1	0	-6	169	142
1	23	-5	38	31
1	21	-5	58	53
1	19	-5	49	44
1	17	-5	56	43 *
1	13	-5	43	41
1	11	-5	99	92
1	9	-5	200	190
1	7	-5	251	232
1	6	-5	38	32
1	5	-5	128	139
1	3	-5	114	126
1	2	-5	46	45
1	1	-5	35	35
1	18	-4	51	36 *
1	17	-4	51	53
1	16	-4	61	59
1	15	-4	59	70
1	14	-4	103	118
1	13	-4	94	88
1	12	-4	94	107
1	11	-4	142	141
1	9	-4	44	41
1	7	-4	114	97
1	5	-4	39	44
1	4	-4	45	47
1	3	-4	83	85
1	2	-4	130	118
1	1	-4	254	260
1	0	-4	336	316
1	22	-3	57	53
1	18	-3	43	38
1	14	-3	47	54
1	12	-3	74	85
1	10	-3	169	182
1	9	-3	72	72
1	8	-3	289	256
1	7	-3	63	62
1	6	-3	151	144
1	4	-3	113	128
1	3	-3	76	68
1	19	-2	60	64
1	18	-2	56	42 *
1	17	-2	67	73
1	16	-2	60	63
1	15	-2	115	104
1	14	-2	43	39
1	13	-2	105	89
1	11	-2	121	118
1	7	-2	128	106
1	6	-2	97	85
1	3	-2	107	84 *
1	2	-2	65	52 *
1	1	-2	292	293
1	23	-1	43	29 *
1	21	-1	48	48
1	19	-1	46	50
1	17	-1	47	42
1	15	-1	41	21 *
1	13	-1	53	52
1	11	-1	85	86
1	9	-1	216	215
1	7	-1	184	207
1	6	-1	27	38 *
1	5	-1	163	155
1	4	-1	85	87
1	3	-1	124	172 *
1	2	-1	29	16 *
1	3	0	51	69 *
1	4	0	76	111 *
1	5	0	28	30
1	6	0	78	73
1	7	0	41	41
1	8	0	114	116
1	9	0	67	57
1	10	0	72	64
1	12	0	98	99
1	14	0	100	101
1	16	0	108	97
1	18	0	67	73
1	22	1	63	56
1	20	1	44	44
1	18	1	47	39
1	13	1	53	50
1	12	1	39	42
1	10	1	133	139
1	9	1	59	57
1	8	1	184	186
1	6	1	95	108
1	5	1	64	69

Table 4. Continued.

h	k	l	$\lvert F_o \rvert$	$\lvert F_c \rvert$	
1	4	1	241	268	
1	3	1	81	105	*
1	19	2	46	33	*
1	17	2	67	65	
1	15	2	174	155	
1	13	2	114	108	
1	9	2	38	52	*
1	7	2	106	103	
1	5	2	150	143	
1	4	2	101	94	
1	3	2	315	317	
1	2	2	71	69	
1	23	3	38	38	
1	21	3	49	54	
1	13	3	45	42	
1	12	3	104	96	
1	11	3	119	106	
1	10	3	163	175	
1	9	3	107	157	
1	8	3	176	161	
1	7	3	122	150	
1	6	3	109	114	
1	5	3	166	179	
1	4	3	191	194	
1	3	3	91	105	
1	1	3	68	92	*
1	20	4	47	50	
1	18	4	66	66	
1	16	4	66	75	
1	14	4	113	120	
1	12	4	103	109	
1	11	4	35	11	*
1	10	4	36	41	
1	8	4	74	71	
1	7	4	82	74	
1	6	4	46	48	
1	4	4	91	106	
1	3	4	107	98	
1	2	4	168	129	*
1	1	4	50	45	
1	0	4	87	95	
1	23	5	35	34	
1	21	5	45	36	
1	13	5	56	52	
1	11	5	153	146	
1	10	5	62	47	*
1	9	5	163	159	
1	8	5	159	177	
1	7	5	185	184	
1	6	5	136	129	
1	5	5	108	108	
1	4	5	108	126	
1	2	5	66	55	
1	1	5	64	46	*
1	19	6	44	42	
1	17	6	73	73	
1	15	6	123	123	
1	13	6	112	106	
1	11	6	89	121	*
1	10	6	82	78	
1	9	6	68	67	
1	6	6	28	26	
1	4	6	60	56	
1	3	6	169	156	
1	1	6	124	135	
1	0	6	103	97	
1	22	7	41	36	
1	21	7	44	31	*
1	20	7	45	39	
1	14	7	55	44	
1	12	7	88	96	
1	10	7	114	123	
1	9	7	91	84	
1	8	7	109	106	
1	7	7	44	52	
1	6	7	192	167	
1	4	7	154	155	
1	3	7	57	64	
1	18	8	48	52	
1	17	8	50	45	
1	16	8	73	75	
1	14	8	87	88	
1	12	8	68	76	
1	10	8	77	79	
1	9	8	39	41	
1	5	8	62	82	*
1	4	8	90	92	
1	3	8	53	45	
1	2	8	230	189	
1	1	8	79	76	
1	0	8	190	180	
1	21	9	43	41	
1	19	9	45	44	
1	13	9	55	62	
1	11	9	90	94	
1	9	9	136	142	
1	8	9	56	59	
1	7	9	170	151	
1	5	9	112	106	
1	4	9	35	45	*
1	3	9	97	100	
1	2	9	101	91	
1	1	9	71	70	
1	19	10	41	30	*
1	17	10	44	47	
1	16	10	50	47	
1	15	10	79	82	
1	14	10	79	76	
1	13	10	47	51	
1	12	10	60	61	
1	6	10	79	81	
1	5	10	71	74	
1	4	10	69	68	
1	3	10	135	127	
1	2	10	.73	70	
1	1	10	128	120	
1	0	10	157	130	
1	12	11	61	65	
1	10	11	120	106	
1	8	11	101	106	
1	6	11	88	80	
1	4	11	87	85	
1	2	11	42	45	
1	17	12	45	41	
1	16	12	44	44	
1	15	12	58	60	
1	14	12	50	46	
1	13	12	88	72	
1	10	12	60	64	
1	5	12	91	80	
1	4	12	89	83	
1	3	12	108	106	
1	2	12	134	119	
1	1	12	153	137	
1	0	12	122	124	
1	19	13	36	37	
1	11	13	47	37	*
1	9	13	110	120	
1	7	13	143	131	
1	5	13	57	60	
1	3	13	43	43	
1	1	13	43	35	
1	18	14	37	43	
1	17	14	42	31	*
1	16	14	48	52	
1	14	14	47	49	
1	12	14	46	36	*
1	6	14	55	50	
1	4	14	93	83	
1	3	14	45	51	
1	2	14	118	101	
1	1	14	63	70	
1	0	14	124	121	
1	10	15	66	63	
1	8	15	86	82	
1	7	15	47	50	
1	6	15	101	106	
1	5	15	47	46	
1	4	15	79	82	
1	15	16	42	44	
1	13	16	53	58	
1	11	16	44	39	
1	5	16	67	70	
1	3	16	88	79	
1	1	16	105	95	
1	0	16	47	49	
1	12	17	53	46	
1	11	17	45	44	
1	10	17	44	45	
1	9	17	44	46	
1	8	17	45	50	
1	7	17	50	56	
1	6	17	60	62	
1	5	17	66	74	
1	3	17	47	55	
1	12	18	55	56	
1	2	18	50	55	
1	0	18	60	62	
1	11	19	52	52	
1	9	19	44	43	
1	8	19	41	35	
1	6	19	58	59	
1	4	19	44	47	
1	3	19	43	31	*
1	5	20	45	37	
1	3	20	53	54	
1	1	20	45	36	
1	7	21	48	52	
1	6	21	46	47	
1	2	22	56	55	
2	2	-22	41	19	*
2	0	-22	45	19	*
2	8	-21	59	61	
2	6	-21	40	43	
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2	2	-20	72	74	
2	0	-20	51	61	
2	9	-19	65	73	
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2	5	-19	55	52	
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2	5	-18	46	46	
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2	0	-18	74	57	*
2	10	-17	71	72	
2	8	-17	88	82	
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2	4	-17	69	76	
2	1	-17	45	23	*
2	14	-16	55	23	*
2	13	-16	55	48	
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2	3	-16	54	56	
2	2	-16	64	70	
2	1	-16	111	113	
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2	4	-14	67	61	
2	2	-14	106	99	
2	0	-14	61	62	
2	12	-13	49	50	
2	10	-13	76	78	
2	8	-13	142	130	
2	6	-13	120	120	
2	5	-13	46	43	
2	4	-13	87	87	
2	19	-12	41	37	
2	17	-12	47	47	
2	15	-12	72	77	
2	13	-12	69	84	
2	11	-12	58	68	
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2	3	-12	92	86	
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2	11	-11	58	54	
2	9	-11	129	141	
2	8	-11	86	75	
2	7	-11	113	114	
2	6	-11	120	126	
2	5	-11	117	109	
2	4	-11	97	81	
2	3	-11	123	108	
2	2	-11	52	49	
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2	18	-10	47	49	
2	16	-10	67	76	
2	14	-10	58	70	
2	12	-10	63	72	
2	10	-10	88	87	
2	7	-10	71	52	*
2	6	-10	53	54	
2	4	-10	131	128	
2	2	-10	224	192	
2	0	-10	176	159	
2	13	-9	49	45	
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2	8	-9	126	129	
2	7	-9	86	78	
2	6	-9	99	91	
2	5	-9	93	85	
2	4	-9	136	134	
2	3	-9	131	85	*
2	2	-9	131	111	
2	19	-8	60	59	
2	17	-8	62	57	
2	15	-8	64	70	
2	13	-8	91	96	
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2	5	-8	95	98	
2	3	-8	100	104	
2	1	-8	304	272	
2	0	-8	132	112	
2	22	-7	38	33	
2	12	-7	59	66	
2	10	-7	89	92	
2	9	-7	40	36	
2	8	-7	152	129	
2	6	-7	164	145	
2	5	-7	92	90	
2	4	-7	93	92	
2	2	-7	142	129	
2	1	-7	56	49	
2	18	-6	73	82	
2	16	-6	70	72	
2	14	-6	57	60	
2	13	-6	46	39	
2	12	-6	104	101	
2	6	-6	30	20	*
2	5	-6	83	79	
2	4	-6	213	187	
2	2	-6	275	263	
2	1	-6	109	89	
2	0	-6	390	347	
2	23	-5	35	36	
2	19	-5	46	50	
2	13	-5	59	58	
2	11	-5	88	99	
2	9	-5	143	127	
2	8	-5	134	116	
2	7	-5	301	264	
2	6	-5	115	121	
2	5	-5	170	149	
2	2	-5	84	76	
2	1	-5	41	6	*
2	19	-4	47	42	
2	17	-4	102	93	
2	15	-4	81	75	
2	14	-4	60	59	
2	13	-4	85	90	
2	12	-4	55	50	
2	11	-4	108	94	
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2	9	-4	42	44	
2	8	-4	93	84	
2	6	-4	66	75	
2	5	-4	168	142	
2	4	-4	93	85	
2	3	-4	117	138	
2	2	-4	48	66	*
2	1	-4	477	396	
2	0	-4	395	320	
2	22	-3	49	41	
2	20	-3	46	42	
2	18	-3	49	41	
2	12	-3	42	41	
2	10	-3	87	97	
2	9	-3	78	75	
2	8	-3	217	210	
2	6	-3	279	267	
2	4	-3	167	142	
2	2	-3	86	62	*
2	19	-2	52	48	
2	18	-2	49	46	
2	17	-2	106	103	
2	16	-2	63	63	
2	15	-2	61	62	
2	14	-2	90	84	
2	13	-2	73	76	
2	12	-2	96	99	
2	11	-2	118	102	
2	10	-2	56	61	
2	9	-2	34	35	
2	8	-2	32	32	
2	7	-2	27	2	*
2	6	-2	27	29	
2	5	-2	53	46	
2	4	-2	74	77	
2	3	-2	39	42	
2	2	-2	78	86	
2	1	-2	123	110	
2	0	-2	249	234	
2	23	-1	47	46	
2	21	-1	47	44	
2	19	-1	48	40	
2	14	-1	45	41	
2	13	-1	61	66	
2	11	-1	129	126	
2	10	-1	43	45	
2	9	-1	141	142	
2	7	-1	129	154	
2	6	-1	156	151	
2	5	-1	91	103	
2	4	-1	132	114	
2	3	-1	206	186	
2	1	-1	57	77	*
2	22	0	42	16	*
2	20	0	54	23	*
2	18	0	49	51	
2	17	0	49	31	*
2	16	0	75	68	
2	15	0	61	52	
2	14	0	97	107	
2	13	0	56	59	
2	12	0	95	97	
2	11	0	75	74	
2	8	0	94	90	
2	7	0	40	42	
2	6	0	106	100	
2	4	0	84	95	
2	1	0	115	152	*
2	12	1	101	106	
2	10	1	144	157	
2	8	1	176	171	
2	7	1	56	56	
2	6	1	297	278	
2	5	1	64	62	
2	4	1	396	387	
2	3	1	80	78	
2	2	1	66	84	*
2	19	2	47	36	*
2	18	2	53	42	*
2	17	2	102	88	
2	15	2	102	105	
2	14	2	59	56	

Table 4. Continued.

h	k	l	$\lvert F_o \rvert$	$\lvert F_c \rvert$	
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2	6	2	27	38	*
2	5	2	95	107	
2	4	2	252	251	
2	3	2	374	343	
2	2	2	126	140	
2	23	3	38	34	
2	21	3	47	49	
2	19	3	47	49	
2	13	3	62	55	
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2	9	3	72	85	
2	8	3	60	56	
2	7	3	342	298	
2	6	3	114	110	
2	5	3	177	167	
2	4	3	220	198	
2	2	3	72	68	
2	18	4	63	66	
2	16	4	69	54	*
2	14	4	72	69	
2	12	4	72	70	
2	10	4	82	73	
2	8	4	102	108	
2	6	4	167	152	
2	4	4	267	235	
2	3	4	40	21	*
2	2	4	84	114	*
2	1	4	108	103	
2	0	4	285	238	
2	21	5	46	36	*
2	20	5	46	45	
2	13	5	59	68	
2	12	5	56	61	
2	11	5	49	50	
2	10	5	115	124	
2	9	5	57	54	
2	8	5	128	117	
2	7	5	171	165	
2	6	5	167	162	
2	5	5	175	180	
2	3	5	86	68	*
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2	17	6	82	80	
2	15	6	75	73	
2	14	6	57	60	
2	13	6	71	78	
2	11	6	60	58	
2	5	6	142	123	
2	3	6	97	102	
2	1	6	274	242	
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2	6	7	97	96	
2	5	7	56	58	
2	4	7	116	116	
2	3	7	77	88	
2	2	7	126	126	
2	1	7	195	168	
2	18	8	65	63	
2	16	8	96	95	
2	14	8	105	105	
2	12	8	61	70	
2	11	8	45	42	
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2	6	8	57	60	
2	4	8	156	147	
2	2	8	234	191	
2	0	8	354	311	
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2	11	9	60	67	
2	10	9	45	40	
2	9	9	117	108	
2	8	9	59	66	
2	7	9	136	147	
2	6	9	92	82	
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2	9	11	47	51	
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2	7	11	58	66	
2	6	11	139	128	
2	5	11	43	53	
2	4	11	98	102	
2	16	12	55	56	
2	14	12	52	60	
2	12	12	49	65	*
2	10	12	49	58	
2	6	12	46	42	
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2	3	12	62	62	
2	2	12	120	113	
2	0	12	89	79	
2	13	13	48	36	*
2	11	13	53	54	
2	9	13	105	105	
2	7	13	91	89	
2	5	13	119	119	
2	3	13	87	82	
2	17	14	42	31	*
2	16	14	46	45	
2	15	14	51	43	
2	13	14	50	54	
2	5	14	48	46	
2	4	14	52	47	
2	3	14	67	66	
2	2	14	79	93	
2	1	14	90	77	
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2	9	17	54	51	
2	7	17	72	79	
2	5	17	74	85	
2	4	18	47	51	
2	2	18	67	73	
2	1	18	52	50	
2	0	18	47	43	
2	10	19	43	43	
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2	6	19	61	66	
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3	8	-19	49	36	*
3	6	-19	46	28	*
3	5	-18	57	61	
3	1	-18	59	109	*
3	0	-18	119	26	*
3	8	-17	68	76	
3	6	-17	65	72	
3	4	-17	51	51	
3	2	-17	56	55	
3	14	-16	47	34	*
3	12	-16	54	48	
3	6	-16	47	44	
3	4	-16	68	68	
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3	11	-15	54	56	
3	9	-15	73	79	
3	7	-15	108	115	
3	5	-15	118	119	
3	3	-15	67	64	
3	16	-14	44	38	
3	14	-14	50	48	
3	9	-14	52	39	*
3	5	-14	61	65	
3	4	-14	106	88	
3	1	-14	71	81	
3	12	-13	79	86	
3	10	-13	99	92	
3	8	-13	121	119	
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3	17	-12	50	44	
3	16	-12	50	55	
3	15	-12	53	45	
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3	0	-12	65	65	
3	11	-11	65	90	*
3	9	-11	108	126	
3	8	-11	70	66	
3	7	-11	141	130	
3	6	-11	47	39	
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3	7	-9	68	70	
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3	5	-9	53	58	
3	4	-9	57	54	
3	3	-9	84	79	
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3	2	-8	83	85	
3	1	-8	281	256	
3	0	-8	73	74	
3	14	-7	52	53	
3	11	-7	47	50	
3	10	-7	46	51	
3	9	-7	111	110	
3	8	-7	140	129	
3	7	-7	162	168	
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3	14	-6	86	97	
3	12	-6	62	70	
3	10	-6	84	81	
3	8	-6	36	38	
3	6	-6	131	132	
3	4	-6	226	203	
3	3	-6	79	59	*
3	2	-6	312	223	*
3	1	-6	145	119	
3	0	-6	234	229	
3	17	-5	62	56	
3	13	-5	58	66	
3	12	-5	47	47	
3	11	-5	49	49	
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3	6	-5	79	80	
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3	4	-5	62	61	
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3	2	-5	27	27	
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3	17	-4	67	52	*
3	15	-4	73	83	
3	13	-4	104	107	
3	11	-4	99	97	
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3	4	-4	56	54	
3	3	-4	227	214	
3	2	-4	151	93	*
3	1	-4	140	137	
3	0	-4	143	100	*
3	14	-3	50	51	
3	12	-3	59	56	
3	11	-3	43	49	
3	10	-3	123	124	
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3	6	-3	69	65	
3	5	-3	104	99	
3	4	-3	155	143	
3	3	-3	88	86	
3	2	-3	36	37	
3	1	-3	47	71	*
3	18	-2	67	69	
3	16	-2	80	87	
3	14	-2	122	123	
3	12	-2	97	97	
3	11	-2	55	57	
3	10	-2	90	94	
3	9	-2	53	53	
3	8	-2	54	49	
3	6	-2	79	77	
3	4	-2	212	181	
3	3	-2	51	68	*
3	2	-2	107	121	
3	1	-2	56	76	*
3	0	-2	106	111	
3	13	-1	90	88	
3	11	-1	142	128	
3	9	-1	140	153	
3	8	-1	80	80	
3	7	-1	252	251	
3	6	-1	108	98	
3	5	-1	227	260	
3	4	-1	87	94	
3	3	-1	131	131	
3	2	-1	47	50	
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3	18	0	47	40	
3	17	0	68	65	
3	16	0	67	67	
3	15	0	125	118	
3	14	0	70	62	
3	13	0	98	97	
3	11	0	50	54	
3	10	0	47	39	
3	9	0	57	55	
3	8	0	96	88	
3	7	0	63	52	
3	6	0	34	50	*
3	5	0	37	21	*
3	4	0	165	177	
3	3	0	79	67	
3	2	0	52	59	
3	1	0	83	110	*
3	14	1	54	56	
3	12	1	105	105	
3	10	1	107	103	
3	8	1	227	211	
3	6	1	276	260	
3	5	1	31	30	
3	4	1	116	121	
3	3	1	58	57	
3	2	1	33	22	*
3	16	2	74	71	
3	15	2	66	68	
3	14	2	64	72	
3	13	2	67	71	
3	10	2	62	81	*
3	8	2	42	31	*
3	5	2	102	110	
3	3	2	100	116	
3	21	3	50	46	
3	19	3	49	46	
3	13	3	48	51	
3	11	3	121	116	
3	9	3	167	152	
3	8	3	78	63	
3	7	3	180	177	
3	6	3	52	56	
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3	16	4	52	48	
3	15	4	98	85	
3	14	4	65	71	
3	12	4	88	82	
3	11	4	84	82	
3	10	4	54	61	
3	9	4	43	43	
3	7	4	69	58	
3	6	4	106	104	
3	5	4	108	93	
3	4	4	195	188	
3	18	5	50	42	
3	10	5	126	122	
3	9	5	53	51	
3	8	5	234	213	
3	7	5	90	95	
3	6	5	140	112	*
3	5	5	83	84	
3	4	5	165	136	
3	2	5	64	60	
3	17	6	73	82	
3	16	6	57	43	*
3	15	6	112	100	
3	11	6	72	73	
3	9	6	55	50	
3	7	6	82	87	
3	5	6	117	113	
3	4	6	138	119	
3	3	6	255	240	
3	2	6	175	147	
3	1	6	207	194	
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3	11	7	57	51	
3	10	7	46	47	
3	9	7	112	105	
3	8	7	99	100	
3	7	7	62	56	
3	6	7	39	41	

Table 4. Continued.

h	k	l	$\|F_o\|$	$\|F_c\|$
3	5	7	114	102
3	4	7	60	62
3	3	7	85	86
3	2	7	34	31
3	1	7	36	35
3	18	8	49	47
3	16	8	86	94
3	14	8	74	80
3	10	8	48	52
3	6	8	41	46
3	5	8	39	27 *
3	4	8	143	138
3	3	8	37	25 *
3	2	8	217	193
3	1	8	89	86
3	0	8	162	154
3	10	9	49	50
3	9	9	52	59
3	8	9	117	115
3	7	9	63	65
3	6	9	131	110
3	5	9	42	53 *
3	4	9	113	106
3	2	9	43	40
3	17	10	49	54
3	15	10	66	88 *
3	13	10	108	104
3	11	10	61	74
3	5	10	53	57
3	3	10	151	130
3	2	10	60	54 *
3	1	10	155	134
3	10	11	73	104 *
3	9	11	66	70
3	8	11	94	93
3	7	11	110	93
3	6	11	111	114
3	5	11	105	86
3	4	11	104	86
3	3	11	45	36
3	2	11	45	44
3	16	12	50	44
3	14	12	59	64
3	12	12	67	72
3	10	12	52	57
3	4	12	66	74
3	2	12	73	73
3	0	12	89	81
3	11	13	62	68
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3	8	13	67	63
3	7	13	107	106
3	6	13	72	59
3	5	13	85	83
3	4	13	50	53
3	17	14	42	40
3	15	14	50	52
3	3	14	79	84
3	1	14	60	63
3	10	15	50	53
3	8	15	67	72
3	6	15	96	100
3	5	15	52	52
3	4	15	52	52
3	5	16	52	51
3	4	16	57	58
3	2	16	86	92
3	10	17	50	3 *
3	9	17	54	58
3	7	17	63	71
3	3	17	51	47
3	4	18	46	49
3	2	18	49	52
3	0	18	70	49 *
3	1	20	49	43
3	0	20	49	41
4	4	-20	58	46
4	0	-20	72	77
4	9	-19	49	35 *
4	8	-19	57	52
4	6	-19	56	48
4	4	-19	56	42 *
4	3	-18	60	65
4	1	-18	60	61
4	10	-17	61	27 *
4	9	-17	61	67
4	7	-17	55	39 *
4	4	-17	61	55
4	4	-16	63	57
4	2	-16	119	130
4	0	-16	119	119
4	9	-15	73	79
4	7	-15	82	92
4	5	-15	107	110
4	4	-15	64	64
4	3	-15	76	76
4	5	-14	58	59
4	4	-14	63	54
4	3	-14	129	126
4	2	-14	89	51 *
4	1	-14	89	89
4	10	-13	76	78
4	6	-13	118	121
4	4	-13	74	85
4	16	-12	67	64
4	10	-12	59	46 *
4	5	-12	60	53
4	2	-12	90	94
4	1	-12	75	70
4	0	-12	82	78
4	11	-11	69	65
4	9	-11	117	117
4	7	-11	101	110
4	6	-11	121	4 *
4	3	-11	56	54
4	16	-10	60	60
4	15	-10	61	67
4	13	-10	83	93
4	12	-10	64	60
4	10	-10	62	61
4	4	-10	110	102
4	3	-10	99	94
4	2	-10	159	138
4	1	-10	167	167
4	0	-10	137	137
4	12	-9	75	88
4	10	-9	86	81
4	8	-9	126	131
4	6	-9	188	184
4	5	-9	57	29 *
4	4	-9	142	126
4	3	-9	50	45
4	2	-9	69	66
4	17	-8	60	65
4	15	-8	69	71
4	13	-8	82	96
4	13	-7	63	49 *
4	5	-8	83	92
4	3	-8	66	70
4	2	-8	116	108
4	1	-8	196	175
4	13	-7	58	49
4	11	-7	60	56
4	9	-7	113	106
4	6	-7	89	81
4	7	-7	134	130
4	5	-7	131	125
4	4	-7	53	44
4	3	-7	61	70
4	1	-7	38	19 *
4	18	-6	60	62
4	16	-6	69	81
4	14	-6	98	100
4	12	-6	93	92
4	10	-6	66	61
4	8	-6	51	51
4	6	-6	46	41
4	4	-6	185	152
4	3	-6	89	90
4	2	-6	147	149
4	1	-6	53	49
4	0	-6	182	183
4	12	-5	71	70
4	11	-5	62	57
4	10	-5	65	66
4	8	-5	156	172
4	7	-5	122	106
4	6	-5	162	146
4	5	-5	83	80
4	4	-5	99	92
4	17	-4	68	75
4	15	-4	96	101
4	13	-4	94	100
4	11	-4	56	57
4	10	-4	53	44
4	9	-4	54	72 *
4	8	-4	51	59
4	6	-4	45	41
4	5	-4	177	171
4	3	-4	168	161
4	2	-4	76	53 *
4	1	-4	234	228
4	12	-3	53	44
4	11	-3	92	92
4	10	-3	56	73 *
4	9	-3	148	153
4	8	-3	132	137
4	7	-3	143	133
4	6	-3	216	213
4	5	-3	199	172
4	4	-3	81	83
4	3	-3	147	149
4	2	-3	62	67
4	1	-3	24	3 *
4	18	-2	62	62
4	16	-2	76	77
4	14	-2	73	83
4	12	-2	96	89
4	6	-2	38	38
4	5	-2	58	65
4	4	-2	144	155
4	3	-2	28	27
4	2	-2	161	171
4	13	-1	77	87
4	12	-1	74	81
4	11	-1	119	122
4	10	-1	95	90
4	9	-1	61	63
4	8	-1	57	54
4	7	-1	134	149
4	6	-1	44	50
4	5	-1	135	142
4	4	-1	30	28
4	3	-1	83	97
4	2	-1	36	43
4	17	0	69	67
4	15	0	82	91
4	13	0	100	112
4	11	0	76	81
4	10	0	51	44
4	9	0	43	41
4	8	0	57	61
4	7	0	80	81
4	6	0	44	51
4	5	0	52	57
4	4	0	55	55
4	2	0	23	34 *
4	1	0	133	178 *
4	18	1	62	58
4	12	1	74	87
4	11	1	54	55
4	10	1	123	127
4	8	1	123	129
4	7	1	114	90 *
4	6	1	188	207
4	5	1	103	91
4	4	1	212	217
4	3	1	27	2 *
4	18	2	67	66
4	14	2	88	81
4	12	2	74	70
4	11	2	54	47
4	10	2	73	63
4	8	2	64	63
4	7	2	85	90
4	6	2	65	65
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4	3	2	34	32
4	11	3	78	81
4	10	3	62	60
4	9	3	142	125
4	8	3	78	77
4	7	3	209	207
4	6	3	82	71
4	5	3	98	98
4	4	3	41	52 *
4	3	3	53	58
4	2	3	38	40
4	18	4	60	58
4	17	4	63	57
4	16	4	64	49 *
4	15	4	64	79
4	13	4	73	71
4	11	4	56	47
4	10	4	49	29 *
4	9	4	46	39
4	8	4	48	46
4	6	4	86	83
4	5	4	113	115
4	4	4	118	122
4	3	4	137	138
4	2	4	84	87
4	11	5	58	51
4	10	5	122	119
4	8	5	150	168
4	6	5	135	133
4	4	5	81	80
4	2	5	36	37
4	17	6	67	74
4	16	6	63	54
4	15	6	64	53
4	14	6	107	94
4	13	6	63	61
4	11	6	64	71
4	6	6	47	49
4	4	6	72	64
4	3	6	115	111
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4	1	6	144	158
4	11	7	78	83
4	9	7	86	88
4	7	7	165	154
4	5	7	152	153
4	4	7	46	55
4	3	7	124	127
4	1	7	72	82
4	14	8	90	86
4	12	8	63	63
4	5	8	60	59
4	4	8	100	92
4	3	8	89	87
4	2	8	124	115
4	1	8	78	59 *
4	0	8	147	134
4	12	9	76	77
4	10	9	73	80
4	8	9	76	72
4	6	9	159	149
4	4	9	126	123
4	13	10	64	68
4	11	10	64	60
4	5	10	73	68
4	4	10	55	49
4	3	10	101	100
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4	9	11	89	82
4	8	11	88	88
4	7	11	103	111
4	5	11	84	85
4	4	11	58	56
4	3	11	57	53
4	1	11	56	51
4	14	12	60	57
4	12	12	63	64
4	10	12	64	60
4	4	12	79	74
4	2	12	84	85
4	0	12	91	75
4	10	13	75	68
4	9	13	69	58
4	8	13	64	64
4	7	13	69	61
4	6	13	82	84
4	5	13	63	52
4	4	13	63	45 *
4	3	14	64	61
4	1	14	89	90
4	8	15	62	52
4	7	15	63	58
4	4	16	63	53
4	2	16	82	90
4	0	16	75	80
4	2	18	61	31 *
5	1	-20	51	28 *
5	5	-19	54	24 *
5	8	-17	63	23 *
5	7	-17	63	63
5	5	-17	74	68
5	4	-16	68	71
5	0	-16	106	98
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5	6	-15	98	91
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5	13	-14	63	59
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5	3	-14	122	112
5	0	-14	152	24 *
5	10	-13	63	70
5	8	-13	122	112
5	6	-13	122	102
5	4	-13	58	40 *
5	14	-12	62	67
5	12	-12	62	65
5	4	-12	68	60
5	2	-12	127	117
5	0	-12	178	172
5	11	-11	76	63
5	9	-11	122	114
5	7	-11	119	109
5	6	-11	67	61
5	5	-11	115	97
5	3	-11	99	87
5	14	-10	62	52
5	13	-10	75	92
5	5	-10	69	70
5	3	-10	87	73
5	2	-10	78	55 *
5	0	-10	110	67 *
5	15	-8	63	66
5	14	-8	70	62
5	13	-8	64	64
5	12	-8	70	72
5	10	-8	73	67
5	4	-8	86	75
5	3	-8	127	112
5	2	-8	93	86
5	1	-8	91	103
5	0	-8	129	115
5	11	-7	74	62
5	9	-7	111	127
5	7	-7	191	169
5	5	-7	176	163
5	3	-7	129	119
5	2	-7	76	76
5	14	-6	77	80
5	12	-6	68	67
5	10	-6	59	48
5	4	-6	120	119
5	3	-6	51	50
5	2	-6	89	88
5	1	-6	63	58
5	12	-5	88	107
5	10	-5	123	140
5	8	-5	122	126
5	6	-5	157	148
5	4	-5	112	122
5	2	-5	81	76
5	15	-4	70	66
5	14	-4	64	64

Table 4. Continued.

h	k	l	$\|F_o\|$	$\|F_c\|$	
5	13	-4	82	87	
5	12	-4	67	60	
5	11	-4	91	95	
5	7	-4	47	42	
5	6	-4	41	34	
5	5	-4	97	92	
5	3	-4	115	119	
5	2	-4	99	92	
5	1	-4	211	204	
5	0	-4	124	126	
5	11	-3	82	96	
5	10	-3	55	57	
5	9	-3	122	143	
5	8	-3	58	56	
5	7	-3	139	154	
5	5	-3	109	112	
5	4	-3	43	40	
5	3	-3	110	104	
5	2	-3	70	70	
5	1	-3	53	55	
5	14	-2	64	65	
5	10	-2	54	56	
5	8	-2	52	44	
5	6	-2	58	63	
5	4	-2	102	111	
5	3	-2	77	74	
5	2	-2	180	194	
5	12	-1	60	59	
5	10	-1	83	84	
5	9	-1	85	87	
5	8	-1	90	94	
5	7	-1	44	61	*
5	6	-1	112	147	*
5	5	-1	85	94	
5	3	-1	55	71	*
5	2	-1	30	32	
5	1	-1	19	37	*
5	17	0	63	62	
5	15	0	108	108	
5	11	0	74	58	*
5	9	0	60	39	*
5	7	0	44	41	
5	5	0	51	74	*
5	4	0	54	50	
5	3	0	202	243	
5	2	0	43	56	*
5	14	1	64	53	
5	11	1	57	44	*
5	10	1	107	103	
5	9	1	100	90	
5	7	1	84	84	
5	6	1	62	62	
5	5	1	40	38	
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5	3	1	112	106	
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5	12	2	85	89	
5	10	2	84	87	
5	7	2	56	45	*
5	4	2	66	85	*
5	11	3	83	87	
5	10	3	61	46	*
5	9	3	154	175	
5	8	3	70	69	
5	7	3	72	79	
5	6	3	61	62	
5	5	3	76	86	
5	4	3	53	63	
5	2	3	38	41	
5	15	4	91	92	
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5	8	5	101	100	
5	7	5	96	94	
5	6	5	80	77	
5	5	5	89	87	
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5	16	6	62	56	
5	14	6	83	90	
5	12	6	82	80	
5	11	6	56	44	*
5	10	6	60	55	
5	5	6	48	49	
5	4	6	88	93	
5	3	6	45	43	
5	13	7	64	56	
5	11	7	81	92	
5	9	7	77	79	
5	7	7	125	129	
5	5	7	150	149	
5	4	7	64	56	
5	3	7	91	89	
5	1	7	53	46	
5	15	8	62	62	
5	14	8	64	59	
5	13	8	70	58	
5	12	8	64	54	
5	4	8	68	69	

h	k	l	$\|F_o\|$	$\|F_c\|$	
5	3	8	98	85	
5	2	8	96	89	
5	1	8	117	125	
5	0	8	112	118	
5	12	9	64	66	
5	10	9	98	100	
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6	4	-19	63	72	
6	4	-18	67	34	*
6	2	-18	63	43	*
6	1	-18	68	61	
6	9	-17	67	61	
6	7	-17	66	65	
6	5	-17	69	60	
6	2	-16	79	83	
6	0	-16	78	97	
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6	12	-12	68	70	
6	10	-12	62	49	*
6	4	-12	89	90	
6	2	-12	146	129	
6	0	-12	150	144	
6	11	-11	68	52	*
6	10	-11	69	56	
6	9	-11	69	71	
6	8	-11	69	51	*
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6	5	-11	96	90	
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6	1	-10	128	115	
6	0	-10	88	82	
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6	9	-9	89	96	
6	8	-9	67	74	
6	7	-9	66	58	
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6	5	-9	63	66	
6	4	-9	80	86	
6	3	-9	79	78	
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6	10	-8	81	74	
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6	6	-7	65	50	*
6	5	-7	134	132	
6	3	-7	115	110	
6	12	-6	69	70	
6	11	-6	68	62	

h	k	l	$\|F_o\|$	$\|F_c\|$	
6	10	-6	61	38	*
6	9	-6	70	64	
6	5	-6	70	62	
6	4	-6	52	43	
6	3	-6	111	90	
6	2	-6	103	94	
6	1	-6	93	96	
6	0	-6	130	118	
6	12	-5	69	78	
6	10	-5	109	111	
6	8	-5	145	139	
6	6	-5	150	136	
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6	14	-4	69	70	
6	11	-4	72	71	
6	9	-4	56	42	*
6	5	-4	62	63	
6	4	-4	84	81	
6	3	-4	65	66	
6	2	-4	78	80	
6	1	-4	84	98	
6	0	-4	63	64	
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6	9	-3	112	109	
6	7	-3	108	117	
6	5	-3	98	120	
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6	1	-2	141	171	
6	12	-1	73	99	*
6	10	-1	95	102	
6	8	-1	102	121	
6	6	-1	120	127	
6	4	-1	54	61	
6	3	-1	55	67	
6	15	0	69	65	
6	13	0	89	93	
6	5	0	110	111	
6	3	0	77	83	
6	2	0	57	82	*
6	11	1	65	45	*
6	10	1	74	72	
6	9	1	99	100	
6	6	1	56	57	
6	5	1	93	95	
6	4	1	42	45	
6	3	1	52	73	*
6	18	2	69	53	*
6	16	2	67	65	
6	14	2	90	97	
6	12	2	104	103	
6	6	2	82	73	
6	4	2	63	74	
6	3	2	48	53	
6	12	3	68	51	*
6	11	3	66	63	
6	10	3	82	86	
6	9	3	60	55	
6	8	3	113	120	
6	7	3	101	93	
6	6	3	129	119	
6	5	3	61	63	
6	4	3	73	71	
6	2	3	38	37	
6	15	4	68	62	
6	13	4	98	104	
6	11	4	86	82	
6	3	4	98	99	
6	5	4	70	69	
6	11	5	88	82	
6	10	5	85	90	
6	9	5	82	90	
6	8	5	72	76	
6	7	5	75	82	
6	6	5	93	99	
6	5	5	104	101	
6	4	5	99	80	
6	3	5	90	82	
6	14	6	68	74	
6	12	6	82	88	
6	4	6	64	43	*
6	3	6	67	71	
6	2	6	72	72	
6	1	6	50	67	*
6	11	7	82	77	
6	10	7	62	52	
6	9	7	72	72	
6	8	7	59	40	*
6	7	7	75	63	
6	6	7	79	75	
6	5	7	83	81	
6	4	7	62	51	
6	3	7	72	74	
6	2	7	50	48	
6	15	8	68	53	*

h	k	l	$\|F_o\|$	$\|F_c\|$	
6	13	8	74	70	
6	11	8	69	55	*
6	3	8	111	106	
6	1	8	118	123	
6	10	9	90	97	
6	8	9	88	83	
6	7	9	87	76	
6	6	9	66	59	
6	5	9	65	51	*
6	4	9	82	82	
6	2	9	80	83	
6	6	10	80	68	
6	3	10	85	72	
6	2	10	77	71	
6	0	10	138	132	
6	9	11	75	82	
6	7	11	82	90	
6	4	12	69	59	
6	3	12	63	55	
6	2	12	69	63	
6	1	12	89	81	
6	0	12	69	70	
6	8	13	67	59	
6	6	13	75	70	
6	1	14	69	70	
6	0	14	75	75	
7	3	-18	55	20	*
7	2	-18	61	58	
7	0	-18	62	70	
7	3	-16	65	47	*
7	2	-16	65	44	*
7	0	-16	71	63	
7	3	-14	66	52	*
7	1	-14	89	88	
7	9	-13	84	80	
7	8	-13	69	20	*
7	5	-13	75	75	
7	4	-12	70	66	
7	2	-12	107	93	
7	1	-12	104	47	*
7	0	-12	117	110	
7	10	-11	68	74	
7	7	-11	64	33	*
7	4	-11	69	72	
7	3	-11	63	43	*
7	2	-11	75	69	
7	13	-10	71	73	
7	5	-10	68	67	
7	3	-10	113	92	
7	11	-9	69	65	
7	10	-9	70	68	
7	9	-9	70	58	
7	8	-9	69	53	*
7	7	-9	115	92	
7	6	-9	68	56	
7	5	-9	86	82	
7	4	-9	78	72	
7	2	-9	64	60	
7	4	-8	119	96	
7	2	-8	137	116	
7	0	-8	158	135	
7	9	-7	81	86	
7	8	-7	80	76	
7	7	-7	78	86	
7	6	-7	125	117	
7	5	-7	61	62	
7	4	-7	60	52	
7	3	-7	75	66	
7	2	-7	57	54	
7	13	-6	69	74	
7	11	-6	70	69	
7	6	-6	61	51	
7	5	-6	76	74	
7	3	-6	103	98	
7	1	-6	162	140	
7	10	-5	96	102	
7	8	-5	144	138	
7	7	-5	61	47	*
7	6	-5	83	95	
7	5	-5	56	42	*
7	4	-5	75	78	
7	2	-5	81	74	
7	14	-4	69	80	
7	2	-4	132	138	
7	1	-4	64	75	
7	0	-4	150	153	
7	11	-3	68	61	
7	9	-3	107	118	
7	7	-3	89	105	
7	5	-3	84	97	
7	3	-3	91	89	
7	15	-2	68	76	
7	3	-2	99	130	*
7	10	-1	66	73	
7	8	-1	84	100	
7	7	-1	51	42	
7	6	-1	86	97	
7	4	-1	84	111	*
7	10	0	65	58	
7	6	0	51	35	*
7	5	0	56	52	
7	4	0	79	83	
7	3	0	82	91	

Table 4. Continued.

h	k	l	$\|F_o\|$	$\|F_c\|$	h	k	l	$\|F_o\|$	$\|F_c\|$	h	k	l	$\|F_o\|$	$\|F_c\|$	h	k	l	$\|F_o\|$	$\|F_c\|$
7	2	0	77	115 *	6	17	0	50	54	9	9	1	64	62	-10	10	3	7[illegible]	64
7	11	1	68	65	6	4	0	75	84	9	7	1	67	63	-10	8	3	78	74
7	9	1	106	112	6	1	0	240	222	-11	1	2	52	54	-10	6	3	63	67
7	7	1	94	100	7	19	0	54	36 *	-11	0	2	56	73 *	-10	4	3	62	65
7	5	1	108	110	7	18	0	47	40	-10	12	2	45	26 *	-9	9	3	54	51
7	3	1	69	107	7	17	0	55	37 *	-10	0	2	37	62 *	-9	8	3	56	74 *
7	14	2	69	62	7	15	0	53	50	-9	15	2	57	55	-9	6	3	93	99
7	12	2	62	80	7	13	0	65	57	-9	13	2	67	64	-9	5	3	74	72
7	4	2	64	73	7	0	0	116	133	-9	3	2	71	63	-9	4	3	74	70
7	3	2	58	60	8	18	0	55	51	-8	15	2	67	72	-9	2	3	67	66
7	10	3	103	102	8	17	0	51	22 *	-8	13	2	72	79	-8	11	3	56	56
7	8	3	104	102	8	16	0	60	57	-8	11	2	65	59	-8	9	3	56	64
7	6	3	105	97	8	14	0	51	49	-8	0	2	100	37 *	-8	8	3	63	70
7	4	3	95	102	8	13	0	57	40 *	-7	17	2	52	47	-8	7	3	103	102
7	2	3	50	55	8	12	0	51	64 *	-7	16	2	56	51	-8	5	3	149	130
7	5	4	55	50	8	10	0	53	56	-7	14	2	57	63	-[illegible]	2	3	83	76
7	4	4	47	43	8	4	0	61	64	-7	2	2	105	97	[illegible]	12	3	75	68
7	3	4	96	101	8	3	0	53	40 *	-7	0	2	81	79	4	1	3	77	80
7	9	5	95	97	8	2	0	61	76 *	-6	14	2	67	63	5	3	3	118	122
7	7	5	105	104	8	1	0	83	70	-6	12	2	59	56	6	1	3	58	60
7	6	5	60	61	8	0	0	69	93 *	-6	3	2	144	155	6	8	3	100	109
7	5	5	81	87	9	16	0	46	43	-6	1	2	167	171	8	6	3	108	110
7	4	5	55	61	9	14	0	48	47	-6	0	2	208	178	9	10	3	58	57
7	3	5	53	50	9	12	0	51	43	-5	16	2	63	74	-10	3	4	76	40 *
7	4	6	70	66	9	2	0	66	80	-5	0	2	200	214	-10	2	4	78	73
7	2	6	109	112	9	1	0	54	21 *	-4	0	2	193	187	-10	0	4	58	57
7	0	6	59	58	9	0	0	65	99	-3	15	2	66	56	-9	4	4	90	90
7	10	7	70	57	10	14	0	50	45	-2	10	2	58	61	-9	2	4	94	103
7	8	7	88	68	10	12	0	45	43	-1	4	2	74	69	-9	0	4	84	81
7	6	7	71	61	10	4	0	54	59	0	17	2	114	96	-8	14	4	79	79
7	5	7	63	59	10	2	0	75	76	0	9	2	74	57 *	-8	12	4	67	72
7	4	7	67	67	10	0	0	69	65	1	11	2	125	119	-8	5	4	83	59 *
7	3	8	63	90	11	4	0	62	39 *	1	1	2	236	261	-8	4	4	100	110
7	1	8	118	125	11	2	0	61	66	1	0	2	73	75	-8	2	4	114	109
7	10	9	69	61	11	0	0	69	54 *	2	11	2	69	71	-8	0	4	74	77
7	8	9	70	64	12	2	0	42	31 *	2	1	2	126	129	-7	16	4	66	63
7	7	9	69	46 *	12	1	0	42	54 *	3	2	2	109	91	-7	12	4	71	51 *
7	5	9	68	55	12	0	0	53	44	3	1	2	119	125	-7	11	4	62	47 *
7	4	9	73	63	-9	10	1	63	49 *	3	0	2	61	106 *	-7	10	4	62	53
7	3	9	66	59	-9	8	1	76	71	4	16	2	95	100	-7	4	4	67	70
7	2	9	72	74	-9	6	1	78	72	4	2	2	89	100	-6	17	4	58	42 *
7	2	10	88	96	-8	12	1	57	56	4	1	2	57	55	-6	16	4	57	51
7	0	10	88	99	-8	10	1	78	72	4	0	2	172	160	3	3	4	255	249
7	0	14	68	71	-8	6	1	72	73	5	16	2	96	91	3	2	4	235	203
0	18	0	100	86	-6	2	1	101	114	5	2	2	241	236	3	1	4	100	161
0	12	0	180	170	-5	4	1	123	146	5	0	2	219	209	3	0	4	150	186
1	20	0	50	31 *	-4	1	1	68	73	6	2	2	92	97	4	1	4	163	174
1	2	0	248	250	1	2	1	95	84	6	0	2	212	211	4	0	4	116	110
1	1	0	124	137	4	10	1	143	127	7	2	2	140	139	5	3	4	102	101
1	0	0	126	362 *	4	2	1	109	127	7	1	2	73	88	5	1	4	106	100
2	19	0	42	38	5	8	1	157	153	7	0	2	137	139	5	0	4	103	64 *
2	2	0	165	177	5	1	1	66	66	8	5	2	59	61	6	1	4	81	89
2	0	0	217	244	6	12	1	55	59	8	4	2	91	76	7	11	4	67	53
4	19	0	50	32 *	6	7	1	145	140	8	2	2	93	92	7	1	4	101	114
4	3	0	156	184	6	2	1	128	105	8	1	2	9[illegible]	89	8	5	4	76	69
4	0	0	96	88	8	11	1	66	66	8	0	2	58	54	8	3	4	83	84
5	21	0	44	22 *	8	9	1	104	105	9	1	2	87	95	8	2	4	75	66
5	19	0	51	33 *	8	7	1	114	109	-11	8	3	54	55	9	4	4	75	76
5	1	0	213	216	8	5	1	103	109	-11	6	3	60	56	9	2	4	106	106
5	0	0	93	96	8	3	1	67	62	-11	4	3	63	60	9	0	4	91	84
6	19	0	50	39 *															

The computations were performed on the computers CDC 3600 in Uppsala and UNIVAC 1108 in Lund using the programs CELSIUS, DRF, LALS, DISTAN, PLANE, ORFFE, and ORTEP.[7]

DESCRIPTION OF THE STRUCTURE

The structure of MONYBDIPIC is built up of the mononuclear tris-(dipicolinato)ytterbate complex and a unit of six connected water and carboxylate oxygen polyhedra around sodium ions. The lanthanoid complexes are located in layers around $x=0$ as is illustrated in Fig. 1. The ligand atoms are designated in Fig. 2. The layer in Fig. 1 alternates with layers around $x=1/2$ containing the sodium unit. This building block is shown in Fig. 3. The pairs of dipicolinate complexes at the symmetry centers (0,0,0) and (0,1/2,1/2) are held together by the sodium ions in columns around the lines $y=z=0$ and $y=z=1/2$, respectively. The columns are connected by hydrogen bonds O(14) – O(19) (Fig. 4).

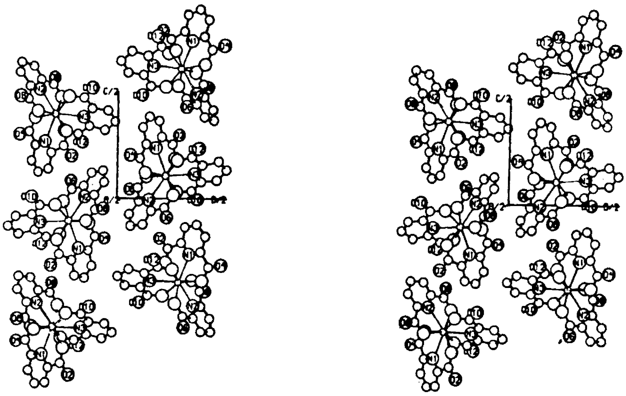

Fig. 1. A stereoscopic pair of drawings showing the layer around $x=0$ in MONYBDIPIC containing the tris(dipicolinato) complexes. Figs. 1, 3, and 4 are drawn with the program ORTEP, written by C. K. Johnson, Oak Ridge.

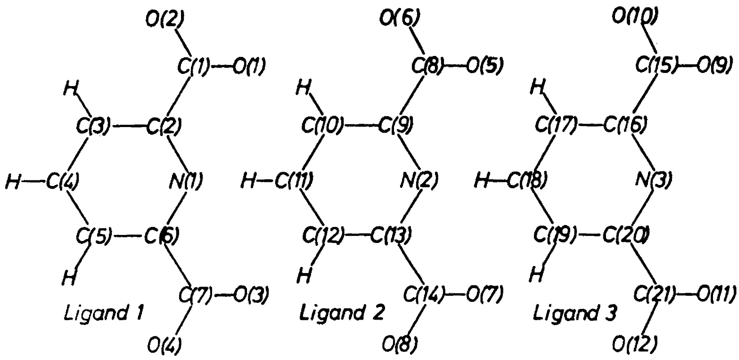

Fig. 2. Designation of the atoms in the three different dipicolinate ligands in MONYB-DIPIC.

Symmetry related sites in the structure are designated below by superscipts (i) – (ix) in the following way:

(i) $1+x,y,z$	(ii) $\bar{x},\bar{y},\bar{z}$	(iii) $1-x,\bar{y},\bar{z}$
(iv) $\bar{x},-1/2+y,1/2-z$	(v) $1-x,1/2+y,1/2-z$	(vi) $1-x,-1/2+y,1/2-z$
(vii) $x,1/2-y,1/2+z$	(viii) $x,1/2-y,-1/2+z$	(ix) $1+x,1/2-y,-1/2+z$

were x,y,z are coordinates of the "crystal-chemical" unit given in Table 2.

In a following paper dealing with the triclinic neodymium dipicolinate compound $Na_3[Nd(C_7H_3NO_4)_3].15H_2O$ all the four investigated lanthanoid dipicolinate phases are compared. Only a few references are made to the previously published structure of the orthorhombic phase ORTYBDIPIC [2] in the discussion below.

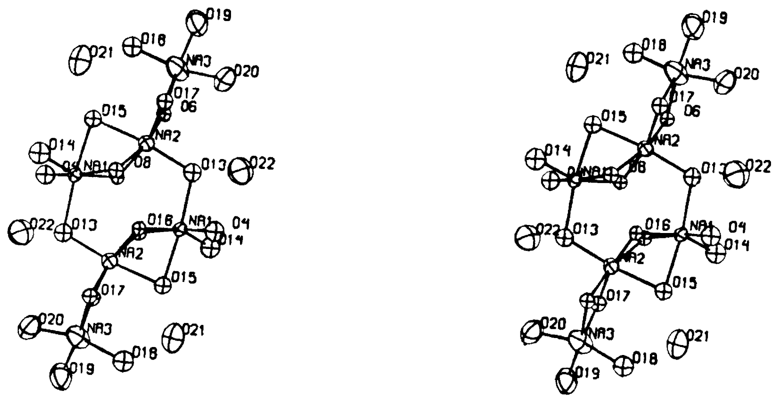

Fig. 3. A stereoscopic pair of drawings showing the unit of six connected water and carboxylate oxygen polyhedra around the sodium ions together with the non-coordinated water oxygen atoms O(21) and O(22). The atoms are represented by "thermal ellipsoids" scaled to include 50 % of the probability distribution.

The ytterbium coordination polyhedron. The tris(dipicolinato)ytterbate ion in MONYBDIPIC has almost the same structure as the corresponding complex in ORTYBDIPIC. The coordination polyhedron is a distorted tricapped trigonal prism. The carboxylate oxygens O(1), O(3), O(5), O(7), O(9), and O(11) are at the corners of the prism and the nitrogen atoms N(1), N(2), and N(3) in the equatorial plane. Selected distances in the coordination

Table 5. Selected interatomic distances (Å) and angles (°) with estimated standard deviations in MONYBDIPIC.

A. The ytterbium coordination polyhedron

Distance		Distance	
Yb – O(1)	2.34(1)	O(3) – N(1)	2.68(2)
Yb – O(3)	2.34(1)	O(3) – N(2)	2.84(2)
Yb – O(5)	2.38(2)	O(5) – O(9)	3.27(2)
Yb – O(7)	2.35(2)	O(5) – O(11)	2.91(2)
Yb – O(9)	2.35(1)	O(5) – N(2)	2.67(2)
Yb – O(11)	2.43(2)	O(5) – N(3)	2.98(3)
Yb – N(1)	2.53(1)	O(7) – O(9)	2.98(2)
Yb – N(2)	2.50(1)	O(7) – N(1)	3.14(2)
Yb – N(3)	2.51(3)	O(7) – N(2)	2.58(2)
O(1) – O(7)	2.95(2)	O(9) – N(2)	2.95(2)
O(1) – O(9)	2.91(2)	O(9) – N(3)	2.77(3)
O(1) – O(11)	3.44(2)	O(11) – N(1)	2.72(2)
O(1) – N(1)	2.57(2)	O(11) – N(3)	2.56(3)
O(1) – N(3)	2.77(3)	N(1) – N(2)	4.47(2)
O(3) – O(5)	3.01(2)	N(1) – N(3)	4.03(3)
O(3) – O(7)	3.23(2)	N(2) – N(3)	4.50(3)
O(3) – O(11)	2.88(2)		

Table 5. Continued.

B. Ligand No. 1

Distance		Angle	
N(1) – C(2)	1.38(2)	C(2) – N(1) – C(6)	124(1)
C(2) – C(3)	1.42(3)	N(1) – C(2) – C(3)	116(2)
C(3) – C(4)	1.32(3)	C(2) – C(3) – C(4)	120(2)
C(4) – C(5)	1.40(3)	C(3) – C(4) – C(5)	123(2)
C(5) – C(6)	1.44(3)	C(4) – C(5) – C(6)	115(2)
C(6) – N(1)	1.30(2)	C(5) – C(6) – N(1)	120(2)
C(1) – C(2)	1.57(3)	N(1) – C(2) – C(1)	113(2)
C(6) – C(7)	1.50(3)	C(3) – C(2) – C(1)	129(2)
C(1) – O(1)	1.25(3)	C(2) – C(1) – O(1)	111(2)
C(1) – O(2)	1.27(3)	C(2) – C(1) – O(2)	115(2)
C(7) – O(3)	1.31(2)	O(1) – C(1) – O(2)	132(2)
C(7) – O(4)	1.27(3)	N(1) – C(6) – C(7)	115(2)
N(1) – C(4)	2.70(2)	C(5) – C(6) – C(7)	124(2)
O(1) – O(2)	2.30(2)	C(6) – C(7) – O(3)	118(2)
O(3) – O(4)	2.29(2)	C(6) – C(7) – O(4)	116(2)
		O(3) – C(7) – O(4)	125(2)

C. Ligand No. 2

Distance		Angle	
N(2) – C(9)	1.30(2)	C(9) – N(2) – C(13)	118(1)
C(9) – C(10)	1.33(3)	N(2) – C(9) – C(10)	128(2)
C(10) – C(11)	1.45(3)	C(9) – C(10) – C(11)	115(2)
C(11) – C(12)	1.44(3)	C(10) – C(11) – C(12)	118(2)
C(12) – C(13)	1.40(3)	C(11) – C(12) – C(13)	115(2)
C(13) – N(2)	1.27(2)	C(12) – C(13) – N(2)	125(2)
C(8) – C(9)	1.56(3)	N(2) – C(9) – C(8)	113(2)
C(13) – C(14)	1.56(3)	C(10) – C(9) – C(8)	119(2)
C(8) – O(5)	1.30(3)	C(9) – C(8) – O(5)	117(2)
C(8) – O(6)	1.23(3)	C(9) – C(8) – O(6)	119(2)
C(14) – O(7)	1.28(2)	O(5) – C(8) – O(6)	124(2)
C(14) – O(8)	1.26(3)	N(2) – C(13) – C(14)	114(1)
N(2) – C(11)	2.75(2)	C(12) – C(13) – C(14)	120(2)
O(5) – O(6)	2.23(2)	C(13) – C(14) – O(7)	113(2)
O(7) – O(8)	2.27(2)	C(13) – C(14) – O(8)	119(2)
		O(7) – C(14) – O(8)	128(2)

D. Ligand No. 3

Distance		Angle	
N(3) – C(16)	1.41(4)	C(16) – N(3) – C(20)	122(2)
C(16) – C(17)	1.38(4)	N(3) – C(16) – C(17)	116(2)
C(17) – C(18)	1.36(4)	C(16) – C(17) – C(18)	127(3)
C(18) – C(19)	1.52(4)	C(17) – C(18) – C(19)	113(3)
C(19) – C(20)	1.43(3)	C(18) – C(19) – C(20)	115(2)
C(20) – N(3)	1.23(4)	C(19) – C(20) – N(3)	126(3)
C(15) – C(16)	1.44(3)	N(3) – C(16) – C(15)	118(2)

Table 5. Continued.

C(20) – C(21)	1.54(3)	C(17) – C(16) – C(15)	126(2)
C(15) – O(9)	1.32(3)	C(16) – C(15) – O(9)	120(2)
C(15) – O(10)	1.31(3)	C(16) – C(15) – O(10)	118(2)
C(21) – O(11)	1.27(3)	O(9) – C(15) – O(10)	122(2)
C(21) – O(12)	1.30(3)	N(3) – C(20) – C(21)	112(2)
N(3) – C(18)	2.83(4)	C(19) – C(20) – C(21)	122(2)
O(9) – O(10)	2.30(2)	C(20) – C(21) – O(11)	117(2)
O(11) – O(12)	2.27(2)	C(20) – C(21) – O(12)	119(2)
		O(11) – C(21) – O(12)	124(2)

E. The sodium coordination

Distance		Distance	
Na(1) – O(4)	2.51(2)	Na(2) – O(16)	2.39(2)
Na(1) – O(8^{i})	2.46(2)	Na(2) – O(17)	2.39(2)
Na(1) – O(13)	2.54(2)	Na(3) – O(6)	2.58(2)
Na(1) – O(14)	2.37(2)	Na(3) – O(17)	2.38(2)
Na(1) – O(15)	2.55(2)	Na(3) – O(18)	2.32(3)
Na(1) – O(16)	2.50(2)	Na(3) – O(19)	2.27(3)
Na(2) – O(6)	2.51(2)	Na(3) – O(20)	2.17(3)
Na(2) – O(8^{i})	2.60(2)	Na(1) – Na(2)	3.39(1)
Na(2) – O(13^{iii})	2.41(2)	Na(1) – Na(2^{iii})	4.05(1)
Na(2) – O(15)	2.57(2)	Na(2) – Na(3)	3.51(2)

F. Possible hydrogen bond distances

Distance		Distance	
O(13) – O(12^{vi})	2.84(3)	O(17) – O(9^{i})	2.80(2)
O(13) – O(22^{vi})	2.74(3)	O(18) – O(5)	2.87(3)
O(14) – O(7^{i})	2.89(2)	O(18) – O(21)	3.12(3)
O(14) – O(19^{vii})	2.91(3)	O(19) – O(10^{i})	2.74(3)
O(15) – O(3)	3.19(2)	O(20) – O(12^{viii})	3.02(3)
O(15) – O(11)	3.20(2)	O(21) – O(1^{i})	2.87(3)
O(15) – O(21)	2.72(3)	O(21) – O(12)	3.01(3)
O(16) – O(3)	2.83(2)	O(21) – O(22)	3.02(4)
O(16) – O(8^{ii})	2.80(2)	O(22) – O(2^{i})	2.74(3)
O(17) – O(2^{ix})	2.79(2)	O(22) – O(4^{v})	2.75(3)

polyhedron are given in Table 5 A. The metal – oxygen bond distances are in the range 2.34 – 2.43 Å with an average of 2.36 Å. The metal – nitrogen distances are almost equal, their average being 2.51 Å.

The triangles O(1)O(7)O(9), O(3)O(5)O(11), and N(1)N(2)N(3) deviate slightly from being equilateral. The two triangular faces of the prism are somewhat tilted; the perpendicular distances from O(3), O(5), and O(11) to the plane formed by O(1), O(7), and O(9) are 3.20, 3.24, and 3.43 Å, respectively. The ytterbium ion occupies a position only 0.07 Å from the plane formed by the equatorial nitrogen atoms. There are twelve independent distances in the coordination polyhedron between adjacent atoms not belonging to the same ligand. They are in the range 2.72 – 3.14 Å with an average of 2.92 Å. As in ORTYBDIPIC this indicates van der Waals contacts in the coordination polyhedron.

The ligands. Each of the three dipicolinate ions in MONYBDIPIC acts as a tridentate ligand forming two five-membered rings with the ytterbium ion. The bond angles Yb – O – C are in the range 120 – 133° and the bond angles Yb – N – C in the range 112 – 125°. As is seen in Table 5, B – D, the corresponding bond distances and angles in the three ligands are not significantly different from one another or from data given in the literature.[2,8–10] The carbon and nitrogen atoms are coplanar within 0.08, 0.04, and 0.07 Å for the ligands 1, 2, and 3, respectively (*cf.* Table 6). The carboxylate groups of ligands 2 and 3 are twisted out of the ligand planes, but in ligand 1 the C – COO groups seem to be bent. The ytterbium ion is well out of the planes of ligands 1 and 3 but lies rather near the plane of ligand 2.

Table 6. The deviations (in Å) from the least-squares planes through the seven carbon atoms and the nitrogen atom of each ligand.

Atom	Distance	Atom	Distance	Atom	Distance
N(1)	– 0.03	N(2)	– 0.01	N(3)	0.01
C(1)	– 0.03	C(8)	0.00	C(15)	– 0.04
C(2)	0.08	C(9)	0.01	C(16)	– 0.01
C(3)	– 0.03	C(10)	0.01	C(17)	0.07
C(4)	0.00	C(11)	– 0.04	C(18)	– 0.03
C(5)	– 0.01	C(12)	0.03	C(19)	– 0.04
C(6)	0.02	C(13)	0.02	C(20)	0.05
C(7)	0.00	C(14)	– 0.02	C(21)	– 0.01
O(1)	– 0.28	O(5)	0.09	O(9)	– 0.13
O(2)	– 0.09	O(6)	– 0.13	O(10)	0.09
O(3)	– 0.16	O(7)	– 0.14	O(11)	– 0.09
O(4)	0.04	O(8)	0.18	O(12)	0.16
Yb	– 0.57	Yb	– 0.08	Yb	– 0.40

The packing of the complex ions. In Fig. 1 the packing of the tris(dipicolinato) complexes within one layer is shown. The shortest carbon – carbon packing distances are C(4) – C(18^{iv}) and C(5) – C(18^{iv}). They are 3.46 ± 0.04 Å and 3.51 ± 0.04 Å, respectively. The separation distance along the *a* axis between the layers of complex ions is about 3.5 Å.

As in ORTYBDIPIC the large mononuclear complex makes all ytterbium – ytterbium distances very long. The shortest distance, Yb – Yb^{vii}, is 9.16 Å which is somewhat less than the shortest distance in ORTYBDIPIC, 9.76 Å. In both compounds the ytterbium ions can be treated as completely isolated from each other.

The coordination around sodium. As is seen in Fig. 2 the sodium ions Na(1) and Na(2) are surrounded by distorted octahedra of oxygen atoms while Na(3) is surrounded by a distorted trigonal bipyramid, O(17), O(18), and O(20), forming its equatorial triangle. Na(1) is connected to Na(2) by sharing the face O(8)O(15)O(16) and to Na(2^{iii}) by sharing the corner O(13). Na(2) is bridged to Na(1) and Na(1^{iii}) and to Na(3), to the latter by sharing the edge O(6)O(17). The sodium – oxygen bond distances and the sodium – sodium distances within the unit of six connected oxygen polyhedra around the sodium ions are given in Table 5 E. The bond distances are in the range 2.17 – 2.60 Å with an average of 2.44 Å. The 29 different oxygen – oxygen "contact" distances along the edges of the polyhedra are in the interval 2.93 – 4.35 Å. Three of these distances are less than 3.20 Å, *viz.*, O(4) – O(13) (3.15 ± 0.03 Å), O(15) – O(16) (2.99 ± 0.02 Å), and O(19) – O(20) (2.93 ± 0.04 Å). The O – Na – O bond angles with adjacent oxygen atoms have values between 73 and 109°.

Possible hydrogen bonds. According to the chemical analyses the asymmetric unit of MONYBDIPIC contains thirteen water molecules. Thus, the hydrogen bonding system in the structure must be rather intricate. As neither the hydrogen nor three of the thirteen water oxygen atoms have been located only some of the possibilities of hydrogen bonding in the structure could be outlined. In Table 5 F the possible hydrogen bond distances less than 3.20 Å are given and the most probable bond scheme among the known oxygen atoms is shown in Fig. 4. This choice has been based upon the following considerations.

(i) Hydrogen bonds between two oxygen atoms in the same coordination polyhedron around sodium are not probable on energetic grounds. The hydrogen atom will be located too near the sodium ion. No case is known of such a bond between water molecules of the same metal coordination polyhedron.[11]

(ii) Donor angles O···O(H_2O)···O between 76 and 136° are accepted in this structure. The average value is 110°. Although the angle H – O – H in the water molecule should deviate less than a few degrees from the water vapor value, 104.5°, hydrogen bonds in crystals often show large deviations from linearity.[11]

(iii) The acceptor angles C – O···O(H_2O) lie in the wide but quite normal interval 100 – 155°.[12]

As was the case in ORTYBDIPIC[2] and in the lanthanoid oxydiacetate compounds $Na_3[M(C_4H_4O_5)_3].2NaClO_4.6H_2O$,[1,3] carboxylate oxygens coordinated to the lanthanoid ion might also be hydrogen bonded to a water molecule. This behaviour is most probable for O(1), O(3), O(5), and O(9).

Experience shows that the 2300 measured independent intensities should suffice to determine even the positions of the three missing water atoms. Since they are not found in the crystal structure determination, they probably occupy disordered positions in the structure. As is seen in Fig. 4 there are cavities around the symmetry centers (1/2,0,1/2) and (1/2,1/2,0) in which at least some of the twelve missing water molecules per unit cell might be occluded.[13] It is very probable that these disordered water molecules are hydrogen bonded to the rest of the structure. In this way they might induce a slight disorder in some atoms of it. The large thermic parameters of the water oxygen atoms O(19) – O(22) and the sodium ion Na(3), which are illustrated in Fig. 3, might thus be caused by such a disorder.

The unit cell dimensions. The unit cell dimensions of the erbium and ytterbium compounds are given above (p. 988). Powder photographs taken with $CrK\alpha_1$ radiation of the holmium, erbium, and thulium compounds show that

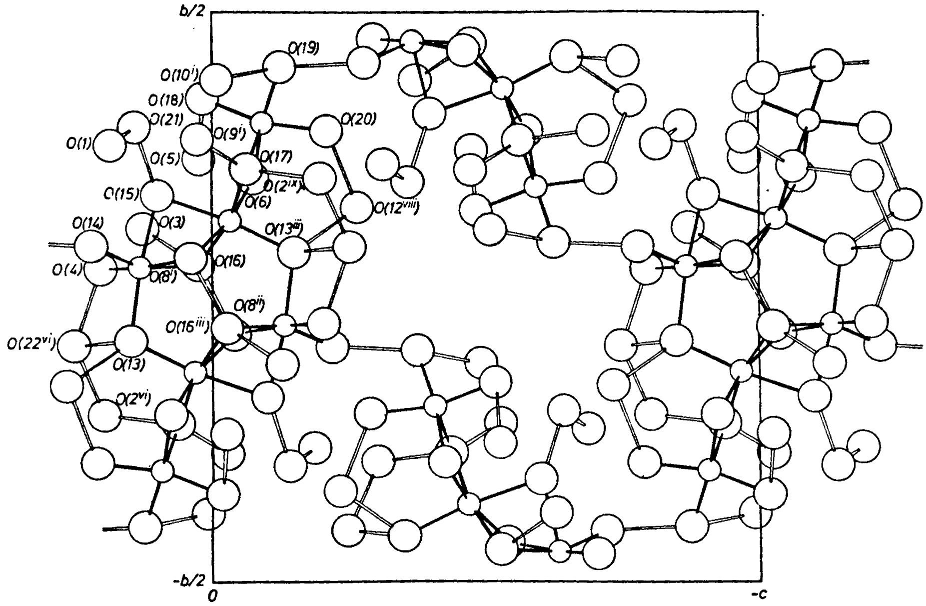

Fig. 4. A projection of MONYBDIPIC along a^* showing the layer around $x = 1/2$. The sodium – oxygen bonds are filled, hydrogen bonds open. Only the most probable hydrogen bonds among those listed in Table 5 F are included.

these three dipicolinates have the same unit cell dimensions within the limits of error but there is a clear-cut decrease of 1.1 % in volume when going from the presumably stable thulium compound to the unstable MONYBDIPIC. In view of other investigations in the present series [3,14] one possible explanation of this fact is that the sodium coordination and the hydrogen bond system (including the disordered water molecules) in the stable monoclinic dipicolinate compounds obstruct the contraction imposed on the structure by the shrinking lanthanoid ion. When the decrease in the radius of the central ion has made a change in the unit cell dimensions inevitable, *i.e.*, at ytterbium, it is the orthorhombic structure ORTYBDIPIC which becomes the most stable one. ORTYBDIPIC contains one more water molecule per formula unit than MONYBDIPIC and the molar volume has increased 53.3 Å^3, *i.e.*, 6.3 %.

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