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# Further Refinement of the Crystal Structure of Hexamethylenediammonium Adipate

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The crystal structure of hexamethylenediammonium adipate has been further refined, with the use of three-dimensional X-ray diffraction data and electronic computing. The $R$ value over 1095 structure amplitudes is 6·1 %. The dimensions of the molecule are all within the usual range; the mean length of the three N–H–O hydrogen bonds is 2·778 Å, and the carboxyl group is twisted out of the plane of the carbon atoms in the adipate ion by 69·8°.

## Introduction

Hexamethylenediammonium adipate (nylon 6.6. salt) is one of the intermediates in nylon manufacture and its approximate crystal structure has already been published (Hirokawa, Ohashi & Nitta, 1954). The present determination was carried out independently from entirely different data, and completion of the refinement was delayed until electronic computing facilities became available; a short account of the results was presented at the Fifth Congress of the International Union of Crystallography at Cambridge (Brown, 1960). Subsequently the crystal structure of the homologue tetramethylenediammonium adipate has been completed (Hirokawa & Ashida, 1962).

## Experimental

Hexamethylenediammonium adipate crystallizes extremely readily from aqueous solutions, the main difficulty being to retard the rate of crystallization in order to avoid inclusions of mother liquor. At or below room temperatures, the stable crystalline modification is a dihydrate, while at slightly elevated temperatures it is anhydrous, the transition point between them, as determined by means of solubility measurements and a cooling curve, being 24·1° (Bantoft, 1952). The specimens used in this work were therefore prepared in warmish surroundings, and from the large crystals grown which were generally (010) plates, elongated along **c**, suitable fragments for taking X-ray photographs were cut with a razor blade.

The unit-cell dimensions of the anhydrous salt, obtained from the measurement of high-order reflexions, were

$$a=8{\cdot}489,\ b=15{\cdot}580,\ c=5{\cdot}598\ \text{Å};\ \beta=102{\cdot}9°.$$

The space group is unambiguously $P2_1/a$. The specific gravity, measured by flotation of the crystals in a mixture of carbon tetrachloride and ethanol, was 1·207, exactly that required for two molecules of $C_{12}H_{26}N_2O_4$ ($M=262{\cdot}2$) per unit cell.

The unit-cell dimensions of the dihydrate, which is triclinic, are approximately

$$a=11{\cdot}90,\ b=15{\cdot}59,\ c=4{\cdot}56\ \text{Å};$$
$$\alpha=94,\ \beta=98,\ \gamma=91°.$$

The specific gravity was found to be 1·224, which indicates two molecules of $C_{12}H_{30}N_2O_6$ per unit cell. No attempt was made to deduce the structure of the dihydrate, although the similarity of the length of $b$ to that of $b$ in the anhydrous salt indicates that the disposition of the molecules with respect to this axis might be the same in the two crystals.

The unit-cell dimensions of the nylon 6.10. salt (hexamethylenediammonium sebacate) have also been determined:

$$a=24{\cdot}25,\ b=8{\cdot}68,\ c=9{\cdot}56\ \text{Å};\ \beta=91{\cdot}6°.$$

The space group of this homologue is $C2/c$, and the specific gravity 1·159, but no further work on it was attempted.

Intensity data on the anhydrous salt were obtained from Weissenberg photographs about **b** (zero layer only) and **c** (zero and four layers), using multiple packs of films with different exposure times in the usual way. The reflexions were estimated visually by comparison with a time exposure calibrated film strip, and the intensities were corrected for the usual geometrical and polarization factors.

## Determination of the structure

The approximate structure was derived readily from space group considerations and 'trial-and-error' structure factor calculations. The mid-points of both cation and anion were required to lie on pairs of centres of symmetry in the unit cell, and since the $c$ axis was fairly short, there would be little chance of overlap of atoms in a **c** projection. Some trial $\{hk0\}$ structure factor calculations quickly gave reasonable agreement

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with the observed values, and two successive electron density projections confirmed the arrangement, and gave the electron density map shown in Fig. 1. Further work was then postponed until electronic computing facilities became available. Meanwhile the structure was published by Hirokawa, Ohashi & Nitta (1954), but as three-dimensional intensity data had already been measured by us, it was thought worth while to continue with the refinement in order to improve the accuracy of the results. Hirokawa, Ohashi & Nitta (1954) obtained the same structure as ours, but their choice of axial directions was different. Our $(x,y,z)$ correspond to their $(z,y,z-x)$.

Eight cycles of three-dimensional structure amplitudes and least-squares calculations were carried out on a Ferranti Pegasus computer with programs written by Cruickshank & Pilling (1961). Anisotropic thermal parameters were used throughout for the carbon, nitrogen and oxygen atoms, and calculated positional coordinates for the hydrogen atoms were included after the third cycle, although these were not refined. The hydrogen coordinates were recalculated after each cycle in order to keep C–H and N–H equal to 1·08 Å, and the isotropic thermal parameters were refined for each of the 13 hydrogen atoms to act as a check on their positions. The final values of the atomic parameters are given in Table 1, and the agreement between the observed and calculated structure amplitudes is shown in Table 2. The $R$ value over all the observed $\{hkl\}$ terms was finally 6·1%. In addition to the 1095 structure amplitudes used in the refinement, there were 247 planes within the range which had zero or unobserved intensities; structure amplitudes were calculated for all these and in every case were found to have negligible values.

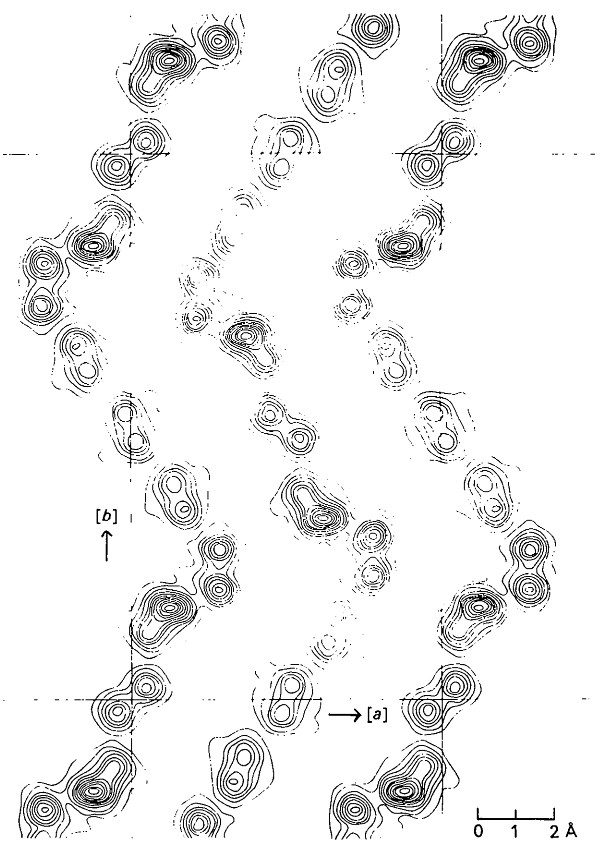

Fig. 1. Electron density projection on (001). The contours are at intervals of 1 e.Å$^{-2}$.

Table 1. *Atomic parameters*

The values of $B_{ij}$ are defined by the expression

$$\exp\left[-\tfrac{1}{4}(h^2a^{*2}B_{11}+2hka^*b^*B_{12}+\ldots)\right]$$

used in the structure amplitude calculations.

| | $x/a$ | $y/b$ | $z/c$ | $B_{11}$ | $B_{22}$ | $B_{33}$ | $B_{12}$ | $B_{23}$ | $B_{13}$ |
|---|---|---|---|---|---|---|---|---|---|
| C(1) | 0·0515 | 0·0244 | −0·0732 | 3·11 | 2·31 | 3·23 | −0·06 | 0·16 | 1·33 |
| C(2) | 0·0544 | 0·1208 | −0·0279 | 3·41 | 2·15 | 2·78 | −0·23 | −0·10 | 1·13 |
| C(3) | 0·1552 | 0·1677 | −0·1794 | 2·60 | 1·85 | 3·47 | 0·18 | 0·15 | 1·13 |
| C(4) | 0·0188 | 0·4734 | 0·3952 | 4·28 | 3·71 | 4·74 | 1·15 | −0·45 | 0·50 |
| C(5) | 0·1364 | 0·4005 | 0·4879 | 3·80 | 3·43 | 4·26 | 0·90 | −0·49 | 0·33 |
| C(6) | 0·1707 | 0·3477 | 0·2794 | 2·80 | 2·76 | 4·11 | 0·08 | 0·25 | 0·76 |
| N | 0·2872 | 0·2774 | 0·3672 | 2·59 | 2·58 | 2·93 | −0·04 | 0·04 | 0·86 |
| O(1) | 0·2867 | 0·1999 | −0·0751 | 3·06 | 4·35 | 3·15 | −1·03 | −0·42 | 0·75 |
| O(2) | 0·1005 | 0·1704 | −0·4093 | 3·21 | 3·77 | 2·12 | −0·28 | 0·46 | 0·51 |
| H(C1*a*) | 0·0044 | 0·0129 | −0·2663 | Mean isotropic $B=3\cdot39$ | | | | | |
| H(C1*b*) | 0·1738 | 0·0004 | −0·0238 | | | | | | |
| H(C2*a*) | −0·0677 | 0·1453 | −0·0731 | | | | | | |
| H(C2*b*) | 0·1033 | 0·1331 | 0·1642 | | | | | | |
| H(C4*a*) | −0·0915 | 0·4462 | 0·2877 | | | | | | |
| H(C4*b*) | 0·0693 | 0·5146 | 0·2765 | | | | | | |
| H(C5*a*) | 0·0868 | 0·3594 | 0·6080 | | | | | | |
| H(C5*b*) | 0·2481 | 0·4270 | 0·5932 | | | | | | |
| H(C6*a*) | 0·0596 | 0·3196 | 0·1773 | | | | | | |
| H(C6*b*) | 0·2201 | 0·3888 | 0·1592 | | | | | | |
| H(N*a*) | 0·2148 | 0·2358 | 0·4539 | | | | | | |
| H(N*b*) | 0·4083 | 0·2975 | 0·4532 | | | | | | |
| H(N*c*) | 0·2870 | 0·2470 | 0·1937 | | | | | | |

Table 2. *Observed and calculated structure amplitudes*

| H | K | L | F(OBS) | F(CAL) |
|---|---|---|---|---|
| 0 | 2 | 0 | 32.0 | -32.1 |
| 0 | 4 | 0 | 8.7 | -8.7 |
| 0 | 6 | 0 | 47.3 | +47.6 |
| 0 | 8 | 0 | 5.5 | +3.9 |
| 0 | 10 | 0 | 4.8 | +5.5 |
| 0 | 14 | 0 | 6.9 | -7.7 |
| 0 | 16 | 0 | 10.3 | -11.1 |
| 0 | 18 | 0 | 4.1 | +4.0 |
| 0 | 20 | 0 | 6.2 | -6.2 |
| 1 | 1 | 0 | 13.2 | +13.2 |
| 1 | 2 | 0 | 7.1 | -7.1 |
| 1 | 3 | 0 | 29.6 | -30.0 |
| 1 | 4 | 0 | 31.0 | +30.3 |
| 1 | 5 | 0 | 11.8 | +13.4 |
| 1 | 6 | 0 | 14.5 | -16.8 |
| 1 | 7 | 0 | 14.4 | +13.2 |
| 1 | 8 | 0 | 16.7 | -17.5 |
| 1 | 9 | 0 | 4.1 | -3.9 |
| 1 | 10 | 0 | 13.0 | +13.7 |
| 1 | 12 | 0 | 13.2 | -13.1 |
| 1 | 14 | 0 | 6.9 | +6.9 |
| 1 | 15 | 0 | 2.7 | -2.8 |
| 1 | 17 | 0 | 6.8 | +7.3 |
| 1 | 18 | 0 | 4.1 | -4.5 |
| 1 | 19 | 0 | 2.0 | -2.5 |
| 1 | 20 | 0 | 1.7 | -1.7 |
| 2 | 0 | 0 | 8.3 | -8.2 |
| 2 | 1 | 0 | 49.3 | -49.8 |
| 2 | 2 | 0 | 70.3 | +70.4 |
| 2 | 3 | 0 | 44.4 | -44.8 |
| 2 | 4 | 0 | 4.2 | -4.1 |
| 2 | 5 | 0 | 42.4 | +42.4 |
| 2 | 6 | 0 | 15.7 | +16.0 |
| 2 | 7 | 0 | 14.3 | -13.9 |
| 2 | 8 | 0 | 15.4 | +14.9 |
| 2 | 9 | 0 | 12.5 | -11.3 |
| 2 | 10 | 0 | 5.2 | -4.8 |
| 2 | 11 | 0 | 7.1 | +7.2 |
| 2 | 12 | 0 | 2.1 | -2.6 |
| 2 | 13 | 0 | 17.3 | -17.3 |
| 2 | 14 | 0 | 9.8 | -10.7 |
| 2 | 16 | 0 | 3.2 | +3.0 |
| 2 | 17 | 0 | 4.5 | +4.8 |
| 2 | 18 | 0 | 4.2 | -4.4 |
| 2 | 19 | 0 | 3.7 | -3.8 |
| 3 | 1 | 0 | 10.8 | +10.3 |
| 3 | 2 | 0 | 21.8 | -21.9 |
| 3 | 3 | 0 | 9.3 | -8.9 |
| 3 | 4 | 0 | 3.1 | +3.5 |
| 3 | 5 | 0 | 19.4 | -20.2 |
| 3 | 6 | 0 | 2.5 | +2.0 |
| 3 | 7 | 0 | 2.6 | +2.6 |
| 3 | 8 | 0 | 12.6 | -13.6 |
| 3 | 9 | 0 | 8.6 | +8.1 |
| 3 | 10 | 0 | 4.4 | -4.4 |
| 3 | 12 | 0 | 2.7 | +2.8 |
| 3 | 13 | 0 | 5.3 | -5.4 |
| 3 | 14 | 0 | 6.0 | -5.9 |
| 3 | 15 | 0 | 6.6 | +6.6 |
| 3 | 16 | 0 | 5.2 | +5.4 |
| 3 | 17 | 0 | 4.5 | -5.1 |
| 3 | 18 | 0 | 4.2 | -4.9 |
| 3 | 19 | 0 | 2.0 | +0.7 |
| 4 | 0 | 0 | 3.3 | -2.6 |
| 4 | 1 | 0 | 23.2 | -24.2 |
| 4 | 2 | 0 | 4.7 | +4.5 |
| 4 | 3 | 0 | 10.9 | +9.4 |
| 4 | 4 | 0 | 33.3 | +33.4 |
| 4 | 5 | 0 | 12.9 | -12.6 |
| 4 | 6 | 0 | 10.9 | -11.5 |
| 4 | 7 | 0 | 5.9 | -5.1 |
| 4 | 8 | 0 | 6.1 | +5.1 |
| 4 | 9 | 0 | 2.0 | -1.3 |
| 4 | 10 | 0 | 5.0 | +5.4 |
| 4 | 11 | 0 | 18.9 | -19.6 |
| 4 | 12 | 0 | 15.4 | -15.4 |
| 4 | 14 | 0 | 5.7 | +5.1 |
| 4 | 17 | 0 | 4.8 | -4.3 |
| 4 | 18 | 0 | 3.8 | -4.8 |
| 5 | 1 | 0 | 13.6 | -12.8 |
| 5 | 2 | 0 | 12.1 | -12.2 |
| 5 | 3 | 0 | 15.4 | +15.5 |
| 5 | 4 | 0 | 8.3 | -8.1 |
| 5 | 5 | 0 | 12.1 | -11.8 |
| 5 | 6 | 0 | 6.3 | +6.4 |
| 5 | 7 | 0 | 7.7 | -7.5 |
| 5 | 8 | 0 | 3.3 | +3.3 |
| 5 | 9 | 0 | 11.9 | +12.4 |
| 5 | 10 | 0 | 8.4 | -9.2 |
| 5 | 11 | 0 | 6.0 | -5.3 |
| 5 | 12 | 0 | 4.8 | -4.8 |
| 5 | 13 | 0 | 5.3 | +4.4 |
| 5 | 14 | 0 | 3.8 | +3.0 |
| 5 | 16 | 0 | 3.8 | -3.6 |
| 5 | 17 | 0 | 2.9 | +2.8 |
| 6 | 1 | 0 | 19.9 | +19.7 |
| 6 | 2 | 0 | 4.4 | +4.6 |
| 6 | 3 | 0 | 14.1 | -14.0 |
| 6 | 4 | 0 | 2.6 | -3.1 |
| 6 | 5 | 0 | 3.3 | -3.5 |
| 6 | 6 | 0 | 4.6 | +3.8 |
| 6 | 9 | 0 | 13.3 | -13.9 |
| 6 | 10 | 0 | 5.3 | -5.0 |
| 6 | 11 | 0 | 4.6 | -4.1 |
| 6 | 12 | 0 | 4.8 | +5.0 |
| 6 | 16 | 0 | 3.5 | -4.0 |
| 7 | 1 | 0 | 8.4 | -8.2 |
| 7 | 2 | 0 | 2.7 | +2.4 |
| 7 | 3 | 0 | 4.3 | -4.0 |
| 7 | 4 | 0 | 6.9 | -6.3 |
| 7 | 5 | 0 | 6.0 | +5.0 |
| 7 | 8 | 0 | 4.3 | +4.2 |
| 7 | 9 | 0 | 7.4 | -7.9 |
| 7 | 10 | 0 | 4.0 | -3.7 |
| 7 | 11 | 0 | 5.0 | +5.3 |
| 7 | 12 | 0 | 4.2 | -4.4 |
| 7 | 14 | 0 | 3.5 | +3.3 |
| 7 | 15 | 0 | 3.9 | +4.2 |
| 8 | 0 | 0 | 6.0 | -5.4 |
| 8 | 1 | 0 | 11.9 | -12.4 |
| 8 | 5 | 0 | 2.6 | -2.4 |
| 8 | 6 | 0 | 3.3 | -3.3 |
| 8 | 7 | 0 | 2.5 | -2.6 |
| 8 | 8 | 0 | 2.4 | -2.5 |
| 8 | 9 | 0 | 1.8 | -1.6 |
| 8 | 11 | 0 | 3.9 | -4.1 |
| 8 | 12 | 0 | 2.0 | +2.2 |
| 9 | 1 | 0 | 4.6 | -4.0 |
| 9 | 2 | 0 | 2.4 | +2.0 |
| 9 | 3 | 0 | 4.1 | -3.1 |
| 9 | 4 | 0 | 1.8 | +1.7 |
| 9 | 9 | 0 | 2.0 | -2.1 |
| 9 | 10 | 0 | 1.1 | -1.3 |
| 10 | 1 | 0 | 5.0 | +5.5 |
| 10 | 2 | 0 | 4.8 | -4.8 |
| 10 | 3 | 0 | 3.4 | -3.6 |
| 10 | 4 | 0 | 3.5 | +3.2 |
| 10 | 5 | 0 | 1.8 | -1.8 |
| 0 | 0 | 1 | 10.2 | -9.0 |
| 0 | 2 | 1 | 5.7 | +5.2 |
| 0 | 3 | 1 | 2.8 | +3.5 |
| 0 | 5 | 1 | 18.1 | -18.9 |
| 0 | 6 | 1 | 11.2 | +11.6 |
| 0 | 7 | 1 | 18.8 | +20.1 |
| 0 | 8 | 1 | 5.5 | +3.9 |
| 0 | 9 | 1 | 11.9 | -11.3 |
| 0 | 10 | 1 | 7.1 | +6.3 |
| 0 | 12 | 1 | 12.9 | -12.1 |
| 0 | 13 | 1 | 9.7 | +8.4 |
| 0 | 15 | 1 | 6.6 | -6.8 |
| 0 | 16 | 1 | 7.7 | +8.3 |
| 0 | 17 | 1 | 1.8 | +1.8 |
| 0 | 18 | 1 | 3.5 | -3.5 |
| 0 | 19 | 1 | 3.2 | +3.2 |
| 1 | 1 | 1 | 76.0 | +77.0 |
| 1 | 2 | 1 | 4.0 | -4.1 |
| 1 | 3 | 1 | 17.3 | -18.5 |
| 1 | 4 | 1 | 6.1 | +5.4 |
| 1 | 5 | 1 | 12.4 | +11.7 |
| 1 | 6 | 1 | 11.8 | -11.2 |
| 1 | 7 | 1 | 16.5 | +16.3 |
| 1 | 8 | 1 | 39.3 | +39.9 |
| 1 | 9 | 1 | 12.4 | +11.1 |
| 1 | 10 | 1 | 14.8 | -13.4 |
| 1 | 11 | 1 | 6.3 | -5.9 |
| 1 | 14 | 1 | 6.8 | +6.5 |
| 1 | 15 | 1 | 8.1 | -8.9 |
| 1 | 16 | 1 | 2.5 | -2.8 |
| 1 | 17 | 1 | 2.9 | -2.8 |
| 1 | 18 | 1 | 1.8 | +2.1 |
| 2 | 0 | 1 | 13.5 | +14.7 |
| 2 | 1 | 1 | 28.7 | +28.5 |
| 2 | 2 | 1 | 2.8 | -2.4 |
| 2 | 4 | 1 | 2.1 | -2.7 |
| 2 | 5 | 1 | 12.0 | -12.6 |
| 2 | 6 | 1 | 10.4 | -10.0 |
| 2 | 7 | 1 | 4.3 | -3.5 |
| 2 | 8 | 1 | 10.8 | +10.8 |
| 2 | 9 | 1 | 5.0 | -4.4 |
| 2 | 10 | 1 | 4.7 | -4.4 |
| 2 | 11 | 1 | 7.2 | -6.9 |
| 2 | 13 | 1 | 3.4 | +3.6 |
| 2 | 14 | 1 | 4.3 | -3.7 |
| 2 | 15 | 1 | 3.3 | +3.3 |
| 2 | 17 | 1 | 3.8 | -3.7 |
| 2 | 18 | 1 | 5.7 | +6.4 |
| 2 | 19 | 1 | 2.6 | -2.5 |
| 3 | 1 | 1 | 21.1 | +20.7 |
| 3 | 2 | 1 | 9.7 | -9.8 |
| 3 | 3 | 1 | 3.2 | +3.8 |
| 3 | 4 | 1 | 21.3 | -20.1 |
| 3 | 5 | 1 | 22.1 | +22.3 |
| 3 | 6 | 1 | 33.4 | +33.7 |
| 3 | 7 | 1 | 10.7 | +9.2 |
| 3 | 8 | 1 | 18.5 | -16.2 |
| 3 | 9 | 1 | 5.3 | -4.0 |
| 3 | 10 | 1 | 2.0 | +1.7 |
| 3 | 12 | 1 | 2.6 | -2.3 |
| 3 | 13 | 1 | 7.5 | -6.8 |
| 3 | 14 | 1 | 4.1 | -3.9 |
| 3 | 16 | 1 | 2.9 | +2.5 |
| 4 | 0 | 1 | 21.0 | +22.9 |
| 4 | 1 | 1 | 12.6 | -13.5 |
| 4 | 2 | 1 | 5.1 | -5.3 |
| 4 | 3 | 1 | 5.9 | -5.5 |
| 4 | 4 | 1 | 20.9 | -22.3 |
| 4 | 6 | 1 | 11.2 | +9.4 |
| 4 | 7 | 1 | 3.1 | -2.8 |
| 4 | 8 | 1 | 5.4 | -4.8 |
| 4 | 9 | 1 | 2.1 | -2.0 |
| 4 | 10 | 1 | 5.9 | +5.0 |
| 4 | 11 | 1 | 8.3 | -8.3 |
| 4 | 13 | 1 | 3.3 | -3.0 |
| 4 | 14 | 1 | 2.0 | -1.9 |
| 4 | 15 | 1 | 2.3 | +2.3 |
| 4 | 17 | 1 | 2.0 | -1.6 |
| 4 | 18 | 1 | 3.4 | -3.5 |
| 5 | 1 | 1 | 2.3 | +2.1 |
| 5 | 2 | 1 | 6.6 | -5.5 |
| 5 | 3 | 1 | 15.0 | +16.0 |
| 5 | 4 | 1 | 8.6 | +9.0 |
| 5 | 5 | 1 | 4.0 | +3.2 |
| 5 | 6 | 1 | 14.4 | -14.7 |
| 5 | 7 | 1 | 4.7 | +4.8 |
| 5 | 8 | 1 | 5.9 | +5.7 |
| 5 | 10 | 1 | 11.8 | -11.7 |
| 5 | 12 | 1 | 2.1 | -1.8 |
| 5 | 13 | 1 | 3.1 | -3.0 |
| 5 | 14 | 1 | 2.9 | +3.2 |
| 5 | 16 | 1 | 1.5 | -1.6 |
| 6 | 0 | 1 | 4.5 | -4.7 |
| 6 | 1 | 1 | 7.7 | -9.0 |
| 6 | 2 | 1 | 6.5 | -6.4 |
| 6 | 3 | 1 | 11.4 | -10.8 |
| 6 | 4 | 1 | 3.3 | +2.7 |
| 6 | 5 | 1 | 2.2 | +2.4 |
| 6 | 6 | 1 | 2.2 | -2.1 |
| 6 | 7 | 1 | 5.0 | +4.1 |
| 6 | 9 | 1 | 9.5 | -8.4 |
| 6 | 10 | 1 | 2.1 | -1.9 |
| 6 | 11 | 1 | 3.1 | -2.8 |
| 6 | 12 | 1 | 2.3 | +2.2 |
| 6 | 13 | 1 | 1.6 | -1.9 |
| 6 | 15 | 1 | 1.8 | -2.3 |
| 7 | 1 | 1 | 8.3 | +9.1 |
| 7 | 2 | 1 | 2.2 | -2.2 |
| 7 | 3 | 1 | 8.3 | -8.9 |
| 7 | 4 | 1 | 3.4 | -3.4 |
| 7 | 5 | 1 | 7.5 | +8.0 |
| 7 | 6 | 1 | 2.2 | +2.7 |
| 7 | 7 | 1 | 2.6 | -3.3 |
| 7 | 8 | 1 | 7.9 | -7.8 |
| 7 | 12 | 1 | 5.9 | -4.9 |
| 7 | 13 | 1 | 2.9 | +2.5 |
| 8 | 0 | 1 | 12.7 | -11.9 |
| 8 | 1 | 1 | 2.1 | -2.5 |
| 8 | 7 | 1 | 6.9 | -6.1 |
| 8 | 8 | 1 | 3.8 | -3.4 |
| 8 | 9 | 1 | 1.6 | +1.4 |
| 8 | 11 | 1 | 2.9 | -3.0 |
| 8 | 12 | 1 | 2.0 | +1.7 |
| 9 | 1 | 1 | 4.8 | -4.5 |
| 9 | 2 | 1 | 7.5 | -7.2 |
| 9 | 3 | 1 | 7.4 | +7.6 |
| 9 | 4 | 1 | 2.0 | +1.9 |
| 9 | 5 | 1 | 5.3 | -5.2 |
| 9 | 7 | 1 | 1.4 | +1.7 |
| 9 | 8 | 1 | 2.7 | -2.4 |
| 9 | 9 | 1 | 2.0 | -2.1 |
| 10 | 0 | 1 | 3.3 | -3.8 |
| 10 | 2 | 1 | 1.3 | -1.5 |
| 10 | 3 | 1 | 1.4 | +1.6 |
| 1 | 1 | -1 | 20.2 | +18.2 |
| 1 | 2 | -1 | 71.9 | -72.8 |
| 1 | 3 | -1 | 77.1 | +76.9 |
| 1 | 4 | -1 | 16.3 | +16.6 |
| 1 | 5 | -1 | 49.3 | -49.8 |
| 1 | 6 | -1 | 19.5 | -21.0 |
| 1 | 7 | -1 | 16.0 | +15.8 |
| 1 | 8 | -1 | 4.9 | -4.6 |
| 1 | 9 | -1 | 19.0 | +18.1 |
| 1 | 10 | -1 | 10.1 | -8.9 |
| 1 | 11 | -1 | 4.7 | -4.7 |
| 1 | 12 | -1 | 9.2 | -8.8 |
| 1 | 13 | -1 | 15.1 | -14.1 |
| 1 | 14 | -1 | 8.3 | -7.3 |
| 1 | 15 | -1 | 2.6 | +2.5 |
| 1 | 17 | -1 | 1.8 | -2.0 |
| 1 | 19 | -1 | 5.8 | -5.2 |
| 2 | 0 | -1 | 14.2 | -15.6 |
| 2 | 1 | -1 | 5.0 | +4.4 |
| 2 | 2 | -1 | 18.0 | +17.7 |
| 2 | 3 | -1 | 9.1 | +8.4 |
| 2 | 4 | -1 | 12.9 | -12.3 |
| 2 | 5 | -1 | 12.7 | -12.2 |
| 2 | 6 | -1 | 21.6 | -22.5 |
| 2 | 7 | -1 | 15.1 | +14.8 |
| 2 | 8 | -1 | 25.8 | +25.1 |
| 2 | 9 | -1 | 19.7 | -20.5 |
| 2 | 10 | -1 | 3.0 | -2.8 |
| 2 | 11 | -1 | 2.5 | -2.4 |
| 2 | 12 | -1 | 7.4 | -7.4 |
| 2 | 13 | -1 | 7.5 | +7.4 |
| 2 | 14 | -1 | 7.5 | +7.1 |
| 2 | 15 | -1 | 4.1 | -3.7 |
| 2 | 18 | -1 | 3.1 | -2.8 |
| 2 | 19 | -1 | 5.2 | -5.3 |
| 3 | 1 | -1 | 4.5 | +4.2 |
| 3 | 2 | -1 | 17.0 | +16.1 |
| 3 | 3 | -1 | 6.4 | -6.4 |
| 3 | 4 | -1 | 50.1 | -50.4 |
| 3 | 5 | -1 | 22.4 | +23.3 |
| 3 | 6 | -1 | 1.8 | +1.6 |
| 3 | 7 | -1 | 18.2 | -17.1 |
| 3 | 8 | -1 | 9.5 | +9.3 |
| 3 | 9 | -1 | 16.0 | +15.0 |
| 3 | 10 | -1 | 29.3 | -29.3 |
| 3 | 11 | -1 | 14.6 | -14.7 |
| 3 | 12 | -1 | 13.6 | -12.8 |
| 3 | 13 | -1 | 4.8 | -4.2 |
| 3 | 14 | -1 | 5.2 | +4.7 |
| 3 | 15 | -1 | 5.7 | +5.0 |
| 3 | 16 | -1 | 4.6 | -4.2 |
| 3 | 17 | -1 | 2.6 | -2.0 |
| 3 | 18 | -1 | 5.3 | -4.6 |
| 3 | 19 | -1 | 2.4 | -2.4 |
| 4 | 0 | -1 | 12.3 | +11.1 |
| 4 | 1 | -1 | 6.4 | +5.1 |
| 4 | 2 | -1 | 11.6 | -11.6 |
| 4 | 3 | -1 | 16.6 | -15.7 |
| 4 | 4 | -1 | 5.4 | -4.5 |
| 4 | 5 | -1 | 3.6 | -3.7 |
| 4 | 6 | -1 | 6.0 | -5.0 |
| 4 | 7 | -1 | 9.8 | +9.5 |
| 4 | 8 | -1 | 3.0 | -2.5 |
| 4 | 10 | -1 | 9.2 | +8.7 |
| 4 | 11 | -1 | 15.0 | -15.0 |
| 4 | 12 | -1 | 2.6 | +2.5 |
| 4 | 13 | -1 | 7.5 | +6.6 |
| 4 | 14 | -1 | 3.3 | -2.9 |
| 4 | 17 | -1 | 7.5 | -7.9 |
| 4 | 18 | -1 | 4.1 | +3.5 |
| 5 | 1 | -1 | 1.5 | +1.1 |
| 5 | 3 | -1 | 8.7 | -8.3 |
| 5 | 4 | -1 | 13.9 | -12.8 |
| 5 | 5 | -1 | 5.0 | -4.7 |
| 5 | 7 | -1 | 19.8 | +19.5 |
| 5 | 8 | -1 | 12.9 | -11.9 |
| 5 | 9 | -1 | 23.5 | -25.4 |
| 5 | 10 | -1 | 15.2 | -14.9 |
| 5 | 11 | -1 | 6.8 | +5.7 |
| 5 | 12 | -1 | 3.4 | -3.0 |
| 5 | 13 | -1 | 7.2 | +6.8 |
| 5 | 15 | -1 | 6.2 | -6.0 |
| 5 | 16 | -1 | 5.2 | -5.2 |
| 6 | 0 | -1 | 1.8 | +1.5 |
| 6 | 1 | -1 | 2.4 | -2.4 |
| 6 | 2 | -1 | 10.6 | -10.1 |
| 6 | 3 | -1 | 6.4 | -6.0 |
| 6 | 4 | -1 | 7.1 | -7.4 |
| 6 | 5 | -1 | 2.1 | -1.5 |
| 6 | 6 | -1 | 14.7 | -15.0 |
| 6 | 7 | -1 | 9.6 | +9.9 |
| 6 | 8 | -1 | 13.0 | -11.7 |
| 6 | 9 | -1 | 2.2 | -1.4 |
| 6 | 10 | -1 | 3.3 | -2.9 |
| 6 | 11 | -1 | 2.6 | -2.2 |
| 6 | 12 | -1 | 9.0 | +9.2 |
| 6 | 13 | -1 | 4.2 | -3.5 |
| 6 | 14 | -1 | 2.1 | -2.0 |
| 6 | 15 | -1 | 3.2 | +2.7 |
| 6 | 16 | -1 | 3.2 | +2.5 |
| 7 | 1 | -1 | 2.6 | -2.2 |
| 7 | 2 | -1 | 14.9 | -16.5 |
| 7 | 3 | -1 | 9.5 | -9.0 |
| 7 | 4 | -1 | 11.7 | +11.6 |
| 7 | 5 | -1 | 6.8 | +6.3 |
| 7 | 6 | -1 | 7.6 | -7.7 |
| 7 | 7 | -1 | 13.5 | -14.0 |
| 7 | 8 | -1 | 7.5 | -7.8 |
| 7 | 9 | -1 | 2.1 | +2.1 |
| 7 | 10 | -1 | 2.0 | -1.4 |
| 7 | 11 | -1 | 4.7 | +3.7 |
| 7 | 14 | -1 | 6.2 | -5.5 |
| 7 | 15 | -1 | 1.6 | -1.1 |
| 8 | 0 | -1 | 8.1 | -8.1 |
| 8 | 1 | -1 | 8.3 | -8.4 |
| 8 | 2 | -1 | 5.9 | -5.7 |
| 8 | 3 | -1 | 2.6 | +2.8 |
| 8 | 4 | -1 | 4.7 | +4.2 |
| 8 | 5 | -1 | 9.6 | +9.5 |
| 8 | 6 | -1 | 3.3 | -3.0 |
| 8 | 7 | -1 | 7.2 | -6.1 |
| 8 | 9 | -1 | 1.8 | +1.5 |
| 8 | 10 | -1 | 4.1 | -4.3 |
| 8 | 11 | -1 | 3.9 | +3.9 |
| 8 | 12 | -1 | 2.2 | +1.6 |
| 9 | 1 | -1 | 7.5 | -7.9 |
| 9 | 2 | -1 | 2.4 | +2.4 |
| 9 | 3 | -1 | 4.7 | +4.5 |
| 9 | 4 | -1 | 7.4 | -7.6 |
| 9 | 5 | -1 | 5.8 | -4.9 |
| 9 | 6 | -1 | 3.5 | +3.1 |
| 9 | 7 | -1 | 2.0 | -2.2 |
| 9 | 8 | -1 | 2.0 | -1.5 |
| 9 | 10 | -1 | 1.8 | +1.9 |
| 10 | 0 | -1 | 7.5 | -7.3 |
| 10 | 1 | -1 | 3.1 | +3.1 |
| 10 | 2 | -1 | 1.5 | +1.6 |
| 10 | 3 | -1 | 2.3 | +1.8 |
| 10 | 5 | -1 | 1.3 | +1.4 |
| 10 | 6 | -1 | 3.2 | -3.0 |
| 0 | 0 | 2 | 33.4 | +34.4 |
| 0 | 1 | 2 | 33.4 | +33.3 |
| 0 | 2 | 2 | 7.7 | +7.2 |
| 0 | 3 | 2 | 8.1 | -9.2 |
| 0 | 4 | 2 | 3.8 | -3.1 |
| 0 | 5 | 2 | 22.7 | +22.3 |
| 0 | 6 | 2 | 9.5 | -7.9 |
| 0 | 7 | 2 | 8.1 | +7.7 |
| 0 | 8 | 2 | 3.6 | +3.3 |
| 0 | 9 | 2 | 6.8 | +7.4 |
| 0 | 10 | 2 | 16.1 | +16.3 |
| 0 | 11 | 2 | 15.2 | +16.2 |
| 0 | 12 | 2 | 9.1 | -9.4 |
| 0 | 13 | 2 | 2.4 | +1.9 |
| 0 | 14 | 2 | 6.5 | -7.0 |
| 0 | 15 | 2 | 7.7 | +8.0 |
| 0 | 16 | 2 | 2.9 | +2.6 |
| 0 | 18 | 2 | 1.6 | -2.3 |
| 0 | 19 | 2 | 2.1 | +2.0 |
| 1 | 1 | 2 | 13.7 | +14.2 |
| 1 | 2 | 2 | 16.0 | -15.4 |
| 1 | 3 | 2 | 3.5 | -3.6 |
| 1 | 4 | 2 | 23.2 | +23.0 |
| 1 | 5 | 2 | 3.6 | +3.3 |
| 1 | 6 | 2 | 10.1 | -10.3 |
| 1 | 7 | 2 | 12.1 | +10.9 |
| 1 | 8 | 2 | 10.3 | -10.6 |
| 1 | 9 | 2 | 4.0 | -3.6 |
| 1 | 10 | 2 | 3.8 | +3.2 |
| 1 | 13 | 2 | 8.1 | -8.7 |
| 1 | 14 | 2 | 2.6 | +2.1 |
| 1 | 15 | 2 | 5.3 | +5.1 |
| 1 | 16 | 2 | 2.9 | -3.2 |
| 1 | 18 | 2 | 1.3 | +1.5 |
| 1 | 19 | 2 | 2.0 | -2.0 |
| 2 | 0 | 2 | 2.9 | +3.0 |
| 2 | 1 | 2 | 26.8 | -26.8 |
| 2 | 2 | 2 | 26.2 | +26.1 |
| 2 | 3 | 2 | 11.3 | +11.4 |
| 2 | 4 | 2 | 10.0 | -9.4 |
| 2 | 6 | 2 | 5.0 | +4.4 |
| 2 | 7 | 2 | 11.9 | +11.2 |
| 2 | 8 | 2 | 18.0 | +16.8 |
| 2 | 9 | 2 | 13.5 | +13.3 |
| 2 | 10 | 2 | 10.0 | -9.4 |
| 2 | 11 | 2 | 4.6 | -4.0 |
| 2 | 13 | 2 | 2.1 | +2.2 |
| 2 | 14 | 2 | 4.0 | -3.9 |
| 2 | 15 | 2 | 1.8 | +1.8 |
| 2 | 16 | 2 | 2.7 | -3.0 |
| 2 | 17 | 2 | 3.9 | +3.9 |
| 3 | 1 | 2 | 3.6 | +3.1 |
| 3 | 2 | 2 | 3.0 | +2.7 |
| 3 | 3 | 2 | 7.7 | +7.8 |
| 3 | 4 | 2 | 5.5 | -5.7 |
| 3 | 5 | 2 | 8.4 | -8.6 |
| 3 | 7 | 2 | 7.1 | -6.6 |
| 3 | 8 | 2 | 7.4 | -7.3 |
| 3 | 9 | 2 | 14.1 | +14.4 |
| 3 | 10 | 2 | 5.1 | -5.5 |
| 3 | 11 | 2 | 2.6 | -3.4 |
| 3 | 12 | 2 | 2.1 | +1.5 |
| 3 | 13 | 2 | 3.2 | -3.4 |
| 3 | 14 | 2 | 4.3 | -3.8 |
| 3 | 15 | 2 | 3.5 | -3.1 |
| 3 | 16 | 2 | 4.3 | +4.5 |
| 3 | 17 | 2 | 2.0 | +2.1 |
| 3 | 18 | 2 | 3.9 | -3.9 |
| 4 | 0 | 2 | 12.4 | +11.1 |
| 4 | 3 | 2 | 11.5 | -10.6 |
| 4 | 4 | 2 | 18.6 | +18.5 |
| 4 | 5 | 2 | 15.1 | +13.9 |
| 4 | 8 | 2 | 2.6 | +2.6 |
| 4 | 9 | 2 | 3.3 | -3.8 |
| 4 | 10 | 2 | 3.3 | +3.5 |
| 4 | 11 | 2 | 3.3 | +3.8 |
| 4 | 12 | 2 | 10.1 | -11.3 |
| 4 | 14 | 2 | 3.9 | +3.7 |
| 4 | 15 | 2 | 1.8 | +2.0 |
| 4 | 16 | 2 | 1.3 | -0.9 |
| 5 | 2 | 2 | 6.9 | -6.1 |
| 5 | 3 | 2 | 5.3 | +4.4 |
| 5 | 4 | 2 | 7.1 | -8.1 |
| 5 | 5 | 2 | 10.1 | -10.3 |
| 5 | 7 | 2 | 4.2 | +3.4 |
| 5 | 8 | 2 | 2.6 | +2.1 |
| 5 | 9 | 2 | 4.1 | +3.9 |
| 5 | 10 | 2 | 5.5 | -5.6 |
| 5 | 11 | 2 | 3.1 | -2.9 |
| 5 | 12 | 2 | 9.2 | -9.9 |
| 5 | 14 | 2 | 3.6 | +4.0 |
| 5 | 15 | 2 | 2.0 | +1.9 |
| 5 | 16 | 2 | 2.4 | -2.1 |
| 6 | 0 | 2 | 10.3 | -10.4 |
| 6 | 1 | 2 | 8.0 | +8.9 |
| 6 | 2 | 2 | 14.0 | +15.9 |
| 6 | 4 | 2 | 2.6 | +2.4 |
| 6 | 5 | 2 | 8.0 | -7.6 |
| 6 | 7 | 2 | 7.2 | +7.3 |
| 6 | 8 | 2 | 3.2 | +3.5 |
| 6 | 9 | 2 | 5.3 | -5.5 |
| 6 | 11 | 2 | 5.3 | -5.4 |
| 6 | 13 | 2 | 4.8 | +4.9 |
| 6 | 14 | 2 | 3.6 | -3.7 |
| 7 | 1 | 2 | 3.2 | -2.8 |
| 7 | 2 | 2 | 3.5 | -2.9 |
| 7 | 3 | 2 | 4.8 | -5.1 |
| 7 | 4 | 2 | 7.1 | -6.8 |
| 7 | 5 | 2 | 6.2 | +6.4 |
| 7 | 6 | 2 | 2.4 | +1.8 |
| 7 | 9 | 2 | 4.1 | -4.3 |
| 7 | 10 | 2 | 10.6 | -10.5 |
| 7 | 12 | 2 | 2.1 | +1.6 |
| 8 | 0 | 2 | 14.6 | +14.7 |
| 8 | 1 | 2 | 2.2 | -1.9 |
| 8 | 2 | 2 | 6.4 | -6.1 |
| 8 | 5 | 2 | 2.7 | -2.6 |
| 8 | 6 | 2 | 4.3 | +4.6 |
| 8 | 7 | 2 | 1.7 | +1.4 |
| 8 | 8 | 2 | 3.1 | -3.0 |
| 9 | 1 | 2 | 3.3 | -3.0 |
| 9 | 3 | 2 | 2.0 | -1.7 |
| 9 | 6 | 2 | 1.8 | -1.7 |
| 1 | 1 | -2 | 9.8 | +9.9 |
| 1 | 2 | -2 | 5.0 | -4.6 |
| 1 | 3 | -2 | 10.9 | -9.9 |
| 1 | 4 | -2 | 14.8 | +13.9 |
| 1 | 5 | -2 | 2.7 | +2.1 |
| 1 | 7 | -2 | 12.7 | +11.7 |
| 1 | 9 | -2 | 3.5 | +2.9 |
| 1 | 10 | -2 | 11.6 | -11.3 |
| 1 | 11 | -2 | 11.5 | -12.1 |
| 1 | 12 | -2 | 4.5 | +4.7 |
| 1 | 13 | -2 | 6.5 | +6.5 |
| 1 | 14 | -2 | 2.6 | +2.9 |
| 1 | 16 | -2 | 4.2 | -3.7 |
| 1 | 17 | -2 | 2.1 | +1.9 |
| 1 | 18 | -2 | 1.6 | -1.5 |
| 1 | 19 | -2 | 1.8 | -1.6 |
| 2 | 0 | -2 | 27.5 | +27.5 |
| 2 | 1 | -2 | 32.1 | +32.0 |
| 2 | 2 | -2 | 19.7 | -20.2 |
| 2 | 3 | -2 | 50.0 | -49.9 |
| 2 | 4 | -2 | 12.7 | -12.7 |
| 2 | 5 | -2 | 18.3 | -20.4 |
| 2 | 6 | -2 | 7.5 | +7.9 |
| 2 | 7 | -2 | 12.2 | +12.1 |
| 2 | 9 | -2 | 16.8 | -16.5 |
| 2 | 10 | -2 | 5.1 | -4.9 |
| 2 | 11 | -2 | 14.1 | -15.2 |
| 2 | 12 | -2 | 2.6 | -2.1 |
| 2 | 13 | -2 | 11.4 | -10.2 |
| 2 | 15 | -2 | 4.4 | +4.0 |
| 2 | 17 | -2 | 5.8 | -5.3 |
| 2 | 18 | -2 | 2.7 | +2.6 |
| 3 | 1 | -2 | 12.4 | +12.8 |
| 3 | 2 | -2 | 7.1 | -6.3 |
| 3 | 3 | -2 | 23.8 | -24.1 |
| 3 | 5 | -2 | 3.2 | +3.2 |
| 3 | 6 | -2 | 11.8 | +12.6 |
| 3 | 7 | -2 | 6.4 | -6.7 |
| 3 | 8 | -2 | 8.7 | -9.4 |
| 3 | 9 | -2 | 4.1 | -3.1 |
| 3 | 11 | -2 | 13.7 | +15.3 |
| 3 | 12 | -2 | 4.6 | -4.2 |
| 3 | 13 | -2 | 8.1 | -8.5 |
| 3 | 14 | -2 | 4.9 | +5.3 |
| 3 | 16 | -2 | 3.5 | -2.9 |
| 3 | 17 | -2 | 2.4 | +2.5 |
| 3 | 18 | -2 | 3.2 | -2.9 |
| 4 | 0 | -2 | 11.4 | -9.4 |
| 4 | 1 | -2 | 41.0 | -39.8 |
| 4 | 2 | -2 | 7.4 | -6.8 |
| 4 | 4 | -2 | 17.7 | -16.7 |
| 4 | 5 | -2 | 6.4 | +7.1 |
| 4 | 6 | -2 | 17.5 | +15.8 |
| 4 | 7 | -2 | 20.4 | -19.5 |
| 4 | 8 | -2 | 8.6 | -8.1 |
| 4 | 9 | -2 | 8.9 | -8.6 |
| 4 | 10 | -2 | 12.9 | -13.8 |
| 4 | 11 | -2 | 9.3 | -10.4 |
| 4 | 12 | -2 | 8.0 | +8.8 |
| 4 | 15 | -2 | 2.2 | -1.9 |
| 4 | 16 | -2 | 1.8 | +1.7 |
| 4 | 18 | -2 | 1.9 | +1.9 |
| 5 | 1 | -2 | 7.2 | -8.3 |
| 5 | 2 | -2 | 18.4 | -17.3 |
| 5 | 4 | -2 | 6.2 | +6.1 |
| 5 | 5 | -2 | 8.0 | -7.3 |
| 5 | 6 | -2 | 1.8 | +2.3 |
| 5 | 7 | -2 | 4.3 | +3.5 |
| 5 | 8 | -2 | 7.8 | +7.2 |
| 5 | 9 | -2 | 2.6 | -2.0 |
| 5 | 12 | -2 | 2.6 | -2.6 |
| 5 | 13 | -2 | 6.2 | +6.3 |
| 5 | 14 | -2 | 4.5 | -4.4 |
| 5 | 15 | -2 | 3.2 | -3.2 |
| 5 | 16 | -2 | 7.2 | +8.1 |
| 5 | 17 | -2 | 2.1 | +2.1 |
| 6 | 0 | -2 | 10.1 | -10.5 |
| 6 | 1 | -2 | 5.1 | -5.5 |
| 6 | 2 | -2 | 10.4 | -10.5 |
| 6 | 3 | -2 | 6.0 | -5.8 |
| 6 | 4 | -2 | 8.7 | +9.8 |
| 6 | 5 | -2 | 6.9 | +6.6 |
| 6 | 6 | -2 | 15.6 | -17.4 |
| 6 | 7 | -2 | 9.2 | -9.4 |
| 6 | 8 | -2 | 5.1 | -5.1 |
| 6 | 9 | -2 | 8.1 | -9.4 |
| 6 | 10 | -2 | 8.0 | +8.6 |
| 6 | 11 | -2 | 4.5 | +4.2 |
| 6 | 12 | -2 | 5.3 | -4.4 |
| 6 | 13 | -2 | 5.7 | -5.4 |
| 6 | 14 | -2 | 5.1 | +4.6 |

Table 2 (*cont.*)

| H | K | L | F(OBS) | F(CAL) |
|---|---|---|---|---|
| 6 | 15 | -2 | 1.3 | +1.3 |
| 7 | 1 | -2 | 11.1 | -13.0 |
| 7 | 2 | -2 | 5.5 | -6.0 |
| 7 | 3 | -2 | 5.5 | +6.0 |
| 7 | 4 | -2 | 5.1 | +5.6 |
| 7 | 5 | -2 | 2.6 | +1.0 |
| 7 | 6 | -2 | 5.1 | +5.6 |
| 7 | 7 | -2 | 5.8 | -6.3 |
| 7 | 8 | -2 | 3.3 | -2.9 |
| 7 | 9 | -2 | 4.0 | +3.0 |
| 7 | 10 | -2 | 4.7 | +5.3 |
| 7 | 11 | -2 | 5.1 | -5.3 |
| 7 | 12 | -2 | 2.7 | -2.2 |
| 7 | 13 | -2 | 3.4 | +3.2 |
| 7 | 14 | -2 | 4.5 | +3.9 |
| 7 | 15 | -2 | 5.3 | +6.4 |
| 8 | 0 | -2 | 20.8 | -21.7 |
| 8 | 1 | -2 | 5.8 | +6.8 |
| 8 | 2 | -2 | 5.8 | +5.9 |
| 8 | 4 | -2 | 4.6 | -4.4 |
| 8 | 5 | -2 | 7.2 | -7.2 |
| 8 | 6 | -2 | 7.7 | -8.2 |
| 8 | 7 | -2 | 5.3 | +5.1 |
| 8 | 8 | -2 | 2.4 | +1.5 |
| 8 | 9 | -2 | 3.4 | +3.0 |
| 8 | 10 | -2 | 2.7 | -2.2 |
| 8 | 11 | -2 | 5.3 | -5.9 |
| 8 | 12 | -2 | 2.0 | +2.3 |
| 8 | 13 | -2 | 1.8 | +2.1 |
| 9 | 1 | -2 | 5.3 | -5.4 |
| 9 | 2 | -2 | 9.5 | +10.2 |
| 9 | 3 | -2 | 5.1 | +5.2 |
| 9 | 4 | -2 | 4.6 | -3.7 |
| 9 | 6 | -2 | 2.7 | +1.9 |
| 9 | 7 | -2 | 2.6 | -1.9 |
| 9 | 8 | -2 | 5.3 | +5.7 |
| 9 | 9 | -2 | 3.2 | -2.6 |
| 9 | 10 | -2 | 3.7 | -3.7 |
| 9 | 11 | -2 | 3.8 | +3.9 |
| 10 | 0 | -2 | 4.3 | +4.2 |
| 10 | 2 | -2 | 9.1 | -8.8 |
| 10 | 3 | -2 | 2.4 | +1.8 |
| 10 | 5 | -2 | 1.6 | -1.9 |
| 10 | 6 | -2 | 3.7 | +3.8 |
| 10 | 7 | -2 | 2.9 | +3.0 |
| 10 | 8 | -2 | 4.6 | -4.8 |
| 0 | 0 | 3 | 12.5 | +12.1 |
| 0 | 1 | 3 | 26.6 | +26.6 |
| 0 | 2 | 3 | 2.6 | -1.8 |
| 0 | 3 | 3 | 2.4 | +2.4 |
| 0 | 4 | 3 | 10.2 | +9.5 |
| 0 | 5 | 3 | 20.4 | -20.1 |
| 0 | 6 | 3 | 2.3 | +1.9 |
| 0 | 7 | 3 | 15.8 | +15.6 |
| 0 | 8 | 3 | 6.5 | +6.7 |
| 0 | 12 | 3 | 8.8 | -9.6 |
| 0 | 13 | 3 | 2.4 | +1.9 |
| 0 | 14 | 3 | 4.6 | +4.7 |
| 0 | 16 | 3 | 1.8 | +1.3 |
| 0 | 17 | 3 | 2.0 | +2.4 |
| 0 | 18 | 3 | 1.6 | -1.2 |
| 1 | 1 | 3 | 13.8 | +13.4 |
| 1 | 2 | 3 | 14.4 | +12.6 |
| 1 | 3 | 3 | 14.9 | -13.3 |
| 1 | 4 | 3 | 7.2 | -6.9 |
| 1 | 5 | 3 | 9.5 | +7.9 |
| 1 | 6 | 3 | 10.3 | +10.2 |
| 1 | 7 | 3 | 14.5 | -13.6 |
| 1 | 8 | 3 | 15.4 | +15.6 |
| 1 | 9 | 3 | 16.0 | +15.2 |
| 1 | 10 | 3 | 5.3 | +5.0 |
| 1 | 13 | 3 | 7.4 | -8.3 |
| 1 | 14 | 3 | 8.1 | +8.6 |
| 1 | 16 | 3 | 1.7 | -1.8 |
| 1 | 17 | 3 | 1.7 | +2.1 |
| 2 | 0 | 3 | 19.0 | +18.5 |
| 2 | 1 | 3 | 6.2 | +5.4 |
| 2 | 2 | 3 | 6.2 | +6.2 |
| 2 | 3 | 3 | 1.7 | +1.6 |
| 2 | 4 | 3 | 10.7 | -11.2 |
| 2 | 6 | 3 | 12.0 | +13.0 |
| 2 | 7 | 3 | 5.5 | -5.7 |
| 2 | 8 | 3 | 4.5 | +4.5 |
| 2 | 10 | 3 | 5.3 | -5.2 |
| 2 | 11 | 3 | 2.5 | -2.7 |
| 2 | 12 | 3 | 3.1 | +2.9 |
| 2 | 13 | 3 | 6.0 | +6.0 |
| 2 | 14 | 3 | 4.7 | -5.6 |
| 2 | 17 | 3 | 4.5 | -5.0 |
| 3 | 1 | 3 | 8.8 | +10.2 |
| 3 | 2 | 3 | 2.5 | +2.3 |
| 3 | 3 | 3 | 5.8 | -6.5 |
| 3 | 4 | 3 | 3.8 | -4.0 |
| 3 | 6 | 3 | 7.5 | +8.3 |
| 3 | 7 | 3 | 18.0 | +18.2 |
| 3 | 8 | 3 | 9.3 | +9.7 |
| 3 | 9 | 3 | 13.6 | -14.1 |
| 3 | 10 | 3 | 2.0 | +1.5 |
| 3 | 11 | 3 | 3.9 | +3.3 |
| 3 | 14 | 3 | 2.1 | +1.9 |
| 3 | 15 | 3 | 3.7 | -3.6 |
| 3 | 16 | 3 | 4.6 | +4.1 |
| 4 | 0 | 3 | 20.5 | +20.9 |
| 4 | 1 | 3 | 11.6 | -12.9 |
| 4 | 2 | 3 | 2.9 | -2.4 |
| 4 | 5 | 3 | 3.1 | +2.5 |
| 4 | 6 | 3 | 3.9 | +3.7 |
| 4 | 7 | 3 | 3.9 | -3.8 |
| 4 | 8 | 3 | 3.9 | -3.1 |
| 4 | 9 | 3 | 4.3 | -3.6 |
| 4 | 10 | 3 | 6.8 | +7.0 |
| 4 | 13 | 3 | 4.8 | -3.9 |
| 4 | 14 | 3 | 6.9 | -5.9 |
| 5 | 1 | 3 | 5.3 | +4.8 |
| 5 | 2 | 3 | 15.2 | -14.6 |
| 5 | 4 | 3 | 17.1 | +17.8 |
| 5 | 5 | 3 | 7.7 | +7.5 |
| 5 | 8 | 3 | 6.0 | -5.3 |
| 5 | 9 | 3 | 4.1 | +4.2 |
| 5 | 10 | 3 | 4.2 | +4.2 |
| 5 | 11 | 3 | 5.1 | -5.3 |
| 5 | 12 | 3 | 1.7 | +1.3 |
| 5 | 13 | 3 | 1.4 | +1.6 |
| 6 | 0 | 3 | 5.3 | -5.1 |
| 6 | 1 | 3 | 4.3 | -3.9 |
| 6 | 2 | 3 | 5.3 | +5.5 |
| 6 | 3 | 3 | 8.6 | -8.3 |
| 6 | 4 | 3 | 2.4 | +2.3 |
| 6 | 5 | 3 | 2.3 | +2.2 |
| 6 | 6 | 3 | 4.1 | -3.7 |
| 6 | 7 | 3 | 2.2 | -2.1 |
| 6 | 8 | 3 | 6.0 | +6.1 |
| 6 | 10 | 3 | 3.1 | -2.8 |
| 6 | 11 | 3 | 3.4 | -3.3 |
| 6 | 12 | 3 | 2.3 | -2.4 |
| 7 | 2 | 3 | 2.2 | +2.0 |
| 7 | 3 | 3 | 9.2 | +10.1 |
| 7 | 4 | 3 | 3.7 | -3.8 |
| 7 | 5 | 3 | 4.4 | -3.9 |
| 7 | 6 | 3 | 5.3 | +5.2 |
| 7 | 7 | 3 | 1.6 | -1.8 |
| 7 | 8 | 3 | 2.6 | -2.9 |
| 7 | 9 | 3 | 4.7 | +4.2 |
| 8 | 0 | 3 | 4.8 | -4.9 |
| 8 | 1 | 3 | 3.1 | -2.3 |
| 8 | 2 | 3 | 1.6 | +0.5 |
| 8 | 4 | 3 | 2.6 | +2.3 |
| 8 | 5 | 3 | 2.5 | -1.9 |
| 1 | 1 | -3 | 5.7 | -5.4 |
| 1 | 2 | -3 | 12.5 | -11.3 |
| 1 | 3 | -3 | 11.1 | +9.2 |
| 1 | 4 | -3 | 20.7 | -19.7 |
| 1 | 5 | -3 | 28.0 | -28.3 |
| 1 | 6 | -3 | 2.3 | +2.4 |
| 1 | 7 | -3 | 9.8 | +9.6 |
| 1 | 8 | -3 | 8.3 | -7.9 |
| 1 | 9 | -3 | 2.9 | +3.4 |
| 1 | 10 | -3 | 6.0 | -4.7 |
| 1 | 11 | -3 | 3.1 | -2.9 |
| 1 | 12 | -3 | 15.4 | -17.5 |
| 1 | 14 | -3 | 1.8 | +1.7 |
| 1 | 17 | -3 | 1.7 | +1.6 |
| 1 | 18 | -3 | 3.2 | -3.2 |
| 2 | 0 | -3 | 16.0 | -16.4 |
| 2 | 1 | -3 | 10.1 | +10.0 |
| 2 | 2 | -3 | 15.6 | +15.2 |
| 2 | 3 | -3 | 4.1 | -3.5 |
| 2 | 4 | -3 | 2.6 | -2.3 |
| 2 | 5 | -3 | 2.1 | +1.8 |
| 2 | 6 | -3 | 10.2 | -10.1 |
| 2 | 7 | -3 | 5.5 | +5.8 |
| 2 | 8 | -3 | 2.1 | +1.6 |
| 2 | 9 | -3 | 11.3 | -11.6 |
| 2 | 10 | -3 | 6.8 | +6.5 |
| 2 | 12 | -3 | 3.1 | -2.8 |
| 2 | 13 | -3 | 3.1 | +2.4 |
| 2 | 14 | -3 | 4.6 | -4.3 |
| 2 | 15 | -3 | 1.7 | +1.4 |
| 2 | 16 | -3 | 4.7 | +4.9 |
| 2 | 17 | -3 | 3.9 | -3.5 |
| 2 | 18 | -3 | 2.0 | -2.1 |
| 3 | 1 | -3 | 12.3 | -12.5 |
| 3 | 2 | -3 | 24.7 | -24.9 |
| 3 | 3 | -3 | 15.4 | -15.1 |
| 3 | 4 | -3 | 5.8 | +6.4 |
| 3 | 5 | -3 | 4.3 | +4.5 |
| 3 | 6 | -3 | 8.5 | -10.1 |
| 3 | 7 | -3 | 3.3 | -3.3 |
| 3 | 8 | -3 | 2.0 | +1.6 |
| 3 | 9 | -3 | 2.8 | -2.9 |
| 3 | 10 | -3 | 15.9 | -15.9 |
| 3 | 11 | -3 | 3.1 | -3.2 |
| 3 | 12 | -3 | 2.5 | -1.7 |
| 3 | 13 | -3 | 2.0 | +1.7 |
| 3 | 14 | -3 | 1.8 | -1.6 |
| 3 | 15 | -3 | 5.8 | +5.3 |
| 4 | 1 | -3 | 9.5 | -9.5 |
| 4 | 2 | -3 | 1.6 | -1.0 |
| 4 | 4 | -3 | 2.5 | -1.9 |
| 4 | 5 | -3 | 4.4 | -4.0 |
| 4 | 6 | -3 | 8.4 | -8.7 |
| 4 | 7 | -3 | 8.0 | +7.4 |
| 4 | 8 | -3 | 5.8 | +5.8 |
| 4 | 9 | -3 | 6.8 | +5.8 |
| 4 | 10 | -3 | 2.4 | -2.9 |
| 4 | 11 | -3 | 8.8 | -9.7 |
| 4 | 12 | -3 | 2.4 | +2.4 |
| 4 | 14 | -3 | 3.9 | +3.5 |
| 4 | 15 | -3 | 2.5 | +2.2 |
| 4 | 16 | -3 | 5.1 | -5.7 |
| 5 | 1 | -3 | 17.8 | -16.4 |
| 5 | 2 | -3 | 7.2 | +7.2 |
| 5 | 3 | -3 | 5.3 | +4.5 |
| 5 | 4 | -3 | 14.9 | -13.2 |
| 5 | 5 | -3 | 6.9 | -6.7 |
| 5 | 6 | -3 | 10.1 | +9.9 |
| 5 | 7 | -3 | 9.5 | -9.3 |
| 5 | 8 | -3 | 10.7 | -10.6 |
| 5 | 9 | -3 | 2.4 | +1.8 |
| 5 | 10 | -3 | 3.1 | -2.5 |
| 5 | 13 | -3 | 4.2 | +4.3 |
| 5 | 14 | -3 | 2.5 | +2.6 |
| 5 | 15 | -3 | 3.1 | +2.4 |
| 5 | 16 | -3 | 1.3 | +1.0 |
| 6 | 0 | -3 | 5.1 | -5.3 |
| 6 | 1 | -3 | 5.8 | +5.4 |
| 6 | 2 | -3 | 5.1 | -4.7 |
| 6 | 3 | -3 | 9.3 | -9.4 |
| 6 | 4 | -3 | 2.4 | +1.9 |
| 6 | 5 | -3 | 6.9 | +6.8 |
| 6 | 6 | -3 | 5.3 | +4.7 |
| 6 | 7 | -3 | 7.7 | +7.8 |
| 6 | 8 | -3 | 5.3 | -6.1 |
| 6 | 9 | -3 | 2.4 | -2.7 |
| 6 | 14 | -3 | 2.9 | +2.6 |
| 6 | 15 | -3 | 1.1 | +1.1 |
| 7 | 1 | -3 | 5.3 | -5.3 |
| 7 | 2 | -3 | 3.1 | +2.4 |
| 7 | 3 | -3 | 6.2 | -5.7 |
| 7 | 5 | -3 | 5.3 | -4.7 |
| 7 | 6 | -3 | 6.2 | -5.8 |
| 7 | 8 | -3 | 10.4 | +10.5 |
| 7 | 9 | -3 | 6.4 | -6.0 |
| 7 | 10 | -3 | 4.6 | -4.5 |
| 7 | 11 | -3 | 5.1 | +5.1 |
| 7 | 13 | -3 | 3.4 | +2.6 |
| 7 | 14 | -3 | 4.1 | +4.9 |
| 8 | 0 | -3 | 6.2 | -6.7 |
| 8 | 1 | -3 | 2.0 | +1.3 |
| 8 | 3 | -3 | 6.2 | +6.4 |
| 8 | 4 | -3 | 4.3 | +4.1 |
| 8 | 5 | -3 | 3.0 | -3.4 |
| 8 | 6 | -3 | 1.8 | +1.3 |
| 8 | 7 | -3 | 5.5 | +5.7 |
| 8 | 8 | -3 | 4.1 | -4.0 |
| 8 | 10 | -3 | 2.3 | -2.5 |
| 8 | 12 | -3 | 4.8 | +4.8 |
| 9 | 3 | -3 | 8.4 | -9.2 |
| 9 | 5 | -3 | 6.2 | +6.1 |
| 9 | 6 | -3 | 5.3 | +5.4 |
| 9 | 7 | -3 | 3.2 | -2.9 |
| 9 | 8 | -3 | 1.5 | -1.3 |
| 9 | 9 | -3 | 3.1 | -2.5 |
| 9 | 10 | -3 | 1.7 | +1.9 |
| 10 | 0 | -3 | 6.0 | +6.9 |
| 10 | 2 | -3 | 2.9 | -3.7 |
| 10 | 3 | -3 | 2.1 | +1.9 |
| 10 | 5 | -3 | 4.3 | +4.1 |
| 10 | 6 | -3 | 3.5 | +3.3 |
| 10 | 7 | -3 | 4.5 | -4.1 |
| 0 | 0 | 4 | 10.9 | -9.8 |
| 0 | 1 | 4 | 1.0 | -1.1 |
| 0 | 2 | 4 | 9.1 | +8.9 |
| 0 | 3 | 4 | 2.7 | +2.8 |
| 0 | 4 | 4 | 14.5 | -15.3 |
| 0 | 5 | 4 | 14.1 | +13.8 |
| 0 | 6 | 4 | 6.8 | -6.4 |
| 0 | 7 | 4 | 4.3 | -4.1 |
| 0 | 8 | 4 | 7.5 | +7.3 |
| 0 | 11 | 4 | 18.6 | +16.7 |
| 0 | 13 | 4 | 3.6 | -2.7 |
| 0 | 16 | 4 | 2.6 | +2.7 |
| 1 | 1 | 4 | 1.3 | +1.0 |
| 1 | 2 | 4 | 5.1 | +3.4 |
| 1 | 3 | 4 | 8.3 | +7.2 |
| 1 | 4 | 4 | 5.5 | +5.9 |
| 1 | 5 | 4 | 1.8 | +1.8 |
| 1 | 6 | 4 | 7.1 | -6.8 |
| 1 | 7 | 4 | 2.7 | -2.5 |
| 1 | 8 | 4 | 4.7 | +4.5 |
| 1 | 9 | 4 | 9.7 | +10.6 |
| 1 | 10 | 4 | 2.2 | +1.7 |
| 1 | 11 | 4 | 6.7 | -7.5 |
| 1 | 12 | 4 | 3.7 | +3.7 |
| 1 | 13 | 4 | 5.5 | -5.9 |
| 1 | 15 | 4 | 6.4 | +7.2 |
| 1 | 16 | 4 | 2.7 | +3.1 |
| 2 | 0 | 4 | 5.1 | -5.9 |
| 2 | 1 | 4 | 4.7 | -4.7 |
| 2 | 2 | 4 | 3.1 | +2.9 |
| 2 | 3 | 4 | 4.3 | +4.2 |
| 2 | 4 | 4 | 6.2 | +5.8 |
| 2 | 6 | 4 | 17.0 | -16.4 |
| 2 | 7 | 4 | 8.7 | +9.3 |
| 2 | 8 | 4 | 7.8 | +8.8 |
| 2 | 9 | 4 | 7.8 | +8.3 |
| 2 | 10 | 4 | 6.8 | +6.4 |
| 2 | 11 | 4 | 2.1 | -1.8 |
| 2 | 12 | 4 | 8.7 | -9.7 |
| 2 | 13 | 4 | 5.1 | +5.0 |
| 2 | 14 | 4 | 3.1 | +2.7 |
| 3 | 1 | 4 | 6.5 | +7.0 |
| 3 | 2 | 4 | 8.4 | +9.5 |
| 3 | 3 | 4 | 4.7 | +5.1 |
| 3 | 4 | 4 | 8.1 | -9.2 |
| 3 | 5 | 4 | 6.2 | -6.5 |
| 3 | 6 | 4 | 6.2 | +6.4 |
| 3 | 7 | 4 | 5.5 | +5.1 |
| 3 | 9 | 4 | 4.8 | +5.4 |
| 3 | 10 | 4 | 3.7 | -3.7 |
| 3 | 11 | 4 | 3.9 | -4.1 |
| 3 | 12 | 4 | 2.5 | +2.7 |
| 3 | 14 | 4 | 4.3 | +4.3 |
| 4 | 0 | 4 | 3.9 | +4.0 |
| 4 | 1 | 4 | 12.3 | +14.4 |
| 4 | 3 | 4 | 3.9 | -4.1 |
| 4 | 4 | 4 | 3.5 | -2.9 |
| 4 | 5 | 4 | 2.8 | -2.4 |
| 4 | 6 | 4 | 7.7 | +8.2 |
| 4 | 7 | 4 | 9.3 | +11.0 |
| 4 | 8 | 4 | 1.6 | -1.6 |
| 4 | 10 | 4 | 1.8 | -1.6 |
| 4 | 11 | 4 | 1.6 | -1.4 |
| 4 | 13 | 4 | 3.1 | +3.1 |
| 5 | 1 | 4 | 6.8 | +7.2 |
| 5 | 2 | 4 | 5.3 | -5.4 |
| 5 | 3 | 4 | 1.6 | -1.5 |
| 5 | 5 | 4 | 2.6 | +2.6 |
| 5 | 6 | 4 | 2.0 | -1.2 |
| 5 | 7 | 4 | 1.5 | +1.6 |
| 5 | 8 | 4 | 3.9 | +3.9 |
| 5 | 9 | 4 | 1.6 | -1.4 |
| 5 | 10 | 4 | 3.7 | -4.2 |
| 5 | 11 | 4 | 3.7 | +4.2 |
| 6 | 0 | 4 | 5.3 | +5.9 |
| 6 | 1 | 4 | 6.0 | -6.4 |
| 6 | 3 | 4 | 4.0 | +3.7 |
| 6 | 4 | 4 | 1.8 | -1.9 |
| 6 | 5 | 4 | 3.4 | +2.8 |
| 6 | 6 | 4 | 3.6 | +3.7 |
| 6 | 8 | 4 | 1.6 | +1.1 |
| 6 | 9 | 4 | 1.1 | -0.9 |
| 7 | 1 | 4 | 1.6 | +1.8 |
| 7 | 2 | 4 | 2.6 | -2.1 |
| 7 | 4 | 4 | 3.0 | -2.7 |
| 7 | 5 | 4 | 3.5 | +4.2 |
| 7 | 6 | 4 | 1.7 | +1.7 |
| 1 | 1 | -4 | 2.6 | -2.3 |
| 1 | 2 | -4 | 9.7 | +10.5 |
| 1 | 3 | -4 | 8.8 | +7.5 |
| 1 | 4 | -4 | 13.7 | -12.6 |
| 1 | 5 | -4 | 7.8 | -7.5 |
| 1 | 7 | -4 | 6.0 | +6.4 |
| 1 | 8 | -4 | 7.4 | +6.9 |
| 1 | 9 | -4 | 2.7 | -3.1 |
| 1 | 10 | -4 | 17.7 | -17.4 |
| 1 | 12 | -4 | 3.9 | +3.7 |
| 1 | 14 | -4 | 3.8 | +3.4 |
| 1 | 15 | -4 | 1.6 | -1.5 |
| 1 | 16 | -4 | 3.0 | -3.0 |
| 2 | 0 | -4 | 14.2 | +14.6 |
| 2 | 1 | -4 | 2.4 | -2.5 |
| 2 | 2 | -4 | 24.6 | -24.9 |
| 2 | 3 | -4 | 13.9 | -12.4 |
| 2 | 4 | -4 | 6.5 | -5.9 |
| 2 | 5 | -4 | 3.2 | -2.6 |
| 2 | 6 | -4 | 6.4 | +5.5 |
| 2 | 7 | -4 | 8.7 | +10.2 |
| 2 | 8 | -4 | 7.4 | -7.4 |
| 2 | 9 | -4 | 9.5 | -10.6 |
| 2 | 11 | -4 | 4.8 | -4.9 |
| 2 | 12 | -4 | 2.2 | +2.3 |
| 2 | 14 | -4 | 2.4 | +2.3 |
| 2 | 15 | -4 | 3.0 | -2.8 |
| 3 | 1 | -4 | 13.0 | +11.5 |
| 3 | 2 | -4 | 7.1 | -6.3 |
| 3 | 3 | -4 | 21.3 | -22.4 |
| 3 | 4 | -4 | 2.1 | +2.1 |
| 3 | 5 | -4 | 8.9 | +9.5 |
| 3 | 6 | -4 | 1.5 | +0.9 |
| 3 | 7 | -4 | 4.0 | -3.8 |
| 3 | 8 | -4 | 2.1 | -1.6 |
| 3 | 9 | -4 | 3.9 | -3.6 |
| 3 | 11 | -4 | 2.8 | +2.2 |
| 3 | 12 | -4 | 6.0 | -5.5 |
| 3 | 13 | -4 | 5.1 | +4.4 |
| 3 | 14 | -4 | 3.9 | +3.6 |
| 3 | 15 | -4 | 4.4 | -4.9 |
| 4 | 0 | -4 | 23.2 | -23.3 |
| 4 | 1 | -4 | 2.9 | -2.8 |
| 4 | 2 | -4 | 3.0 | -3.0 |
| 4 | 3 | -4 | 5.1 | -4.9 |
| 4 | 4 | -4 | 5.1 | -4.4 |
| 4 | 5 | -4 | 8.6 | +9.1 |
| 4 | 6 | -4 | 2.0 | -1.2 |
| 4 | 7 | -4 | 1.8 | -4.5 |
| 4 | 8 | -4 | 3.5 | -3.8 |
| 4 | 9 | -4 | 2.2 | -1.4 |
| 4 | 10 | -4 | 4.3 | -3.6 |
| 4 | 12 | -4 | 5.3 | +5.9 |
| 4 | 13 | -4 | 1.8 | -1.6 |
| 4 | 15 | -4 | 4.8 | +5.5 |
| 4 | 16 | -4 | 4.5 | +4.5 |
| 5 | 1 | -4 | 4.8 | -4.4 |
| 5 | 2 | -4 | 9.3 | -10.7 |
| 5 | 3 | -4 | 4.3 | +3.7 |
| 5 | 5 | -4 | 3.4 | -2.9 |
| 5 | 6 | -4 | 9.5 | +10.6 |
| 5 | 7 | -4 | 2.8 | +2.1 |
| 5 | 8 | -4 | 3.9 | -3.8 |
| 5 | 9 | -4 | 5.5 | -5.4 |
| 5 | 11 | -4 | 4.5 | +4.1 |
| 5 | 12 | -4 | 1.8 | +2.0 |
| 5 | 13 | -4 | 1.6 | -1.8 |
| 5 | 14 | -4 | 4.5 | -4.2 |
| 5 | 15 | -4 | 2.2 | +2.3 |
| 6 | 0 | -4 | 1.4 | -1.7 |
| 6 | 1 | -4 | 4.6 | -5.4 |
| 6 | 2 | -4 | 7.7 | -9.0 |
| 6 | 3 | -4 | 6.2 | +6.8 |
| 6 | 6 | -4 | 1.7 | -2.1 |
| 6 | 7 | -4 | 2.2 | +1.5 |
| 6 | 8 | -4 | 5.5 | -5.9 |
| 6 | 9 | -4 | 3.4 | +3.0 |
| 6 | 10 | -4 | 2.6 | +2.8 |
| 6 | 12 | -4 | 2.2 | +1.8 |
| 6 | 13 | -4 | 4.9 | +4.4 |
| 6 | 14 | -4 | 3.5 | +4.3 |
| 7 | 1 | -4 | 6.4 | -6.6 |
| 7 | 3 | -4 | 5.5 | +5.8 |
| 7 | 4 | -4 | 5.5 | +5.6 |
| 7 | 5 | -4 | 5.5 | +5.2 |
| 7 | 6 | -4 | 2.7 | -2.8 |
| 7 | 7 | -4 | 6.0 | -6.4 |
| 7 | 8 | -4 | 5.8 | +6.2 |
| 7 | 11 | -4 | 2.8 | -2.5 |
| 7 | 13 | -4 | 3.0 | +3.0 |
| 8 | 0 | -4 | 5.3 | -5.6 |
| 8 | 1 | -4 | 2.7 | +2.9 |
| 8 | 3 | -4 | 3.4 | -3.3 |
| 8 | 5 | -4 | 5.8 | +5.7 |
| 8 | 6 | -4 | 2.5 | -2.5 |
| 8 | 7 | -4 | 1.8 | +2.0 |
| 8 | 9 | -4 | 1.5 | +1.8 |
| 8 | 11 | -4 | 5.5 | +5.7 |
| 9 | 2 | -4 | 5.1 | +5.2 |
| 9 | 5 | -4 | 2.0 | +1.8 |
| 9 | 6 | -4 | 1.8 | +1.9 |
| 9 | 7 | -4 | 1.3 | +1.3 |
| 9 | 8 | -4 | 2.2 | -2.1 |
| 10 | 0 | -4 | 5.1 | -5.9 |
| 10 | 1 | -4 | 2.2 | -2.0 |
| 10 | 2 | -4 | 2.6 | +2.3 |
| 10 | 3 | -4 | 4.3 | +4.4 |
| 10 | 4 | -4 | 2.6 | +2.6 |
| 0 | 0 | 5 | 5.8 | +5.7 |
| 2 | 0 | 5 | 2.9 | +3.3 |
| 4 | 0 | 5 | 8.7 | +8.7 |
| 6 | 0 | 5 | 2.4 | -2.6 |
| 2 | 0 | -5 | 8.2 | +8.1 |
| 4 | 0 | -5 | 18.5 | -19.4 |
| 6 | 0 | -5 | 1.7 | -1.7 |
| 0 | 0 | 6 | 6.0 | -5.9 |
| 2 | 0 | 6 | 5.3 | +4.2 |
| 2 | 0 | -6 | 9.8 | -9.7 |
| 4 | 0 | -6 | 1.8 | -1.3 |
| 6 | 0 | -6 | 1.1 | +1.3 |
| 0 | 0 | 7 | 2.1 | -2.2 |
| 2 | 0 | -7 | 3.1 | +3.0 |
| 4 | 0 | -7 | 4.2 | -4.1 |

## Description of the structure

Nylon 6.6. salt is a true organic salt, *i.e.* it is formed by the neutralization of an organic acid with an organic base. The neutral ions exist as such in the crystal, and, by virtue of the acid being dibasic and the base diacidic with the functional groups at both ends, continuous hydrogen-bonded chains of $[NH_3 . C_6H_{12} . NH_3]^{2+} . [OOC . C_4H_8 . COO]^{2-}$ are formed, and these extend along **b**. Each ion is centrosymmetrical and makes use of this by crystallizing with the mid-points of the ions on centres of symmetry in the crystal lattice. There are also hydrogen bonds linking nitrogen and oxygen atoms of neighbouring chains so as to form a continuous hydrogen-bond system along **a**. The packing of the molecules in the unit cell is shown in Fig. 2.

The bond lengths and inter-bond angles are listed in Table 3; the mean C–C bond in the chains is 1·522 Å, and the mean tetrahedral angle subtended at a carbon atom by bonds to neighbouring non-hydrogen atoms is 112·1°. The mean of the three N–H–O hydrogen bonds is 2·778 Å. It is interesting to compare the results of this work with those obtained by Hirokawa & Ashida (1962) for tetramethylenediammonium adipate (Fig. 3). The similarity between the dimensions of the adipate ions in the two structures is striking, and these compare favourably also with the dimensions found in adipic acid (Morrison & Robertson, 1949) although this was refined by means of two-dimensional projections only. The dimensions of the hexamethylenediammonium ion may be compared with those of hexamethylenediamine (Binnie & Robertson, 1950), hexamethylenediamine dihydrochloride (Binnie & Robertson, 1949*a*), and hexamethylenediamine dihydrobromide (Binnie & Robertson, 1949*b*).

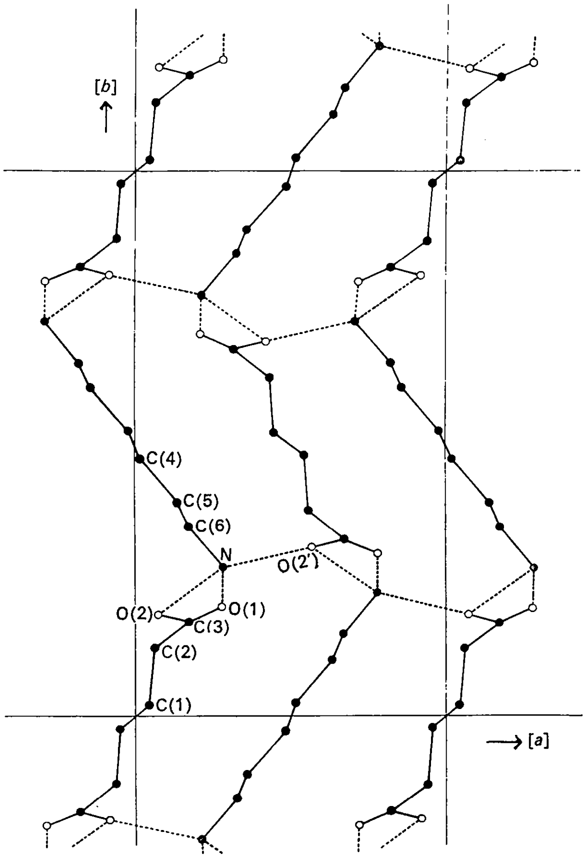

Fig. 2. Diagram of contents of unit cell, showing hydrogen bond system by broken lines.

Table 3. *Bond lengths and inter-bond angles*

| | | | |
|---|---|---|---|
| C(1′)-C(1) | 1·528 Å | C(1′)-C(1)-C(2) | 113·0° |
| C(1) -C(2) | 1·522 | C(1) -C(2)-C(3) | 111·8 |
| C(2) -C(3) | 1·520 | C(2) -C(3)-O(1) | 119·4 |
| C(3) -O(1) | 1·244 | C(2) -C(3)-O(2) | 117·3 |
| C(3) -O(2) | 1·268 | O(1) -C(3)-O(2) | 123·4 |
| C(4′)-C(4) | 1·528 | C(4′)-C(4)-C(5) | 112·1 |
| C(4) -C(5) | 1·524 | C(4) -C(5)-C(6) | 111·6 |
| C(5) -C(6) | 1·509 | C(5) -C(6)-N | 112·1 |
| C(6) -N | 1·483 | | |

Mean standard deviation of a bond length, 0·0033 Å.
Mean standard deviation of an inter-bond angle, 0·2°.

| | |
|---|---|
| N-H-O(1) | 2·757 Å |
| N-H-O(2) | 2·782 |
| N-H-O(2′) | 2·795 |

The planarity of the ions was investigated by considering the equation of the mean plane through atoms C(1), C(2), C(3) and the origin (0,0,0), which, referred to standard orthogonal axes, is:

$$0{\cdot}6556X-0{\cdot}1093Y+0{\cdot}7472Z=0\,.$$

Atom C(1) is +0·006 Å out of this plane, C(2) is +0·006 Å and C(3) −0·006 Å.

The mean plane through atoms C(2), C(3), O(1) and O(2) is

$$0{\cdot}5060X-0{\cdot}8559Y-0{\cdot}1069Z=-1{\cdot}3451$$

from which the out-of-plane distances are C(2) +0·002; C(3) −0·006; O(1) +0·002 and O(2) +0·002 Å. The angle between the normals to these two planes is 69·8°, which is the angle through which the carboxyl group is twisted with respect to the plane of the carbon atoms in the chain. Hirokawa, Ohashi & Nitta (1954) quote 19° for this angle, but this appears to be a miscalculation. This compares with 67·5° found in tetramethylenediammonium adipate, but only 6° found in adipic acid. This big difference is presumably due to the geometrical strains imposed by the difference in hydrogen-bonding conditions.

The plane through atoms C(4), C(5), C(6) and the point $(0,\frac{1}{2},\frac{1}{2})$ is:

$$-0{\cdot}7975X-0{\cdot}6020Y+0{\cdot}0397Z=-4{\cdot}0827$$

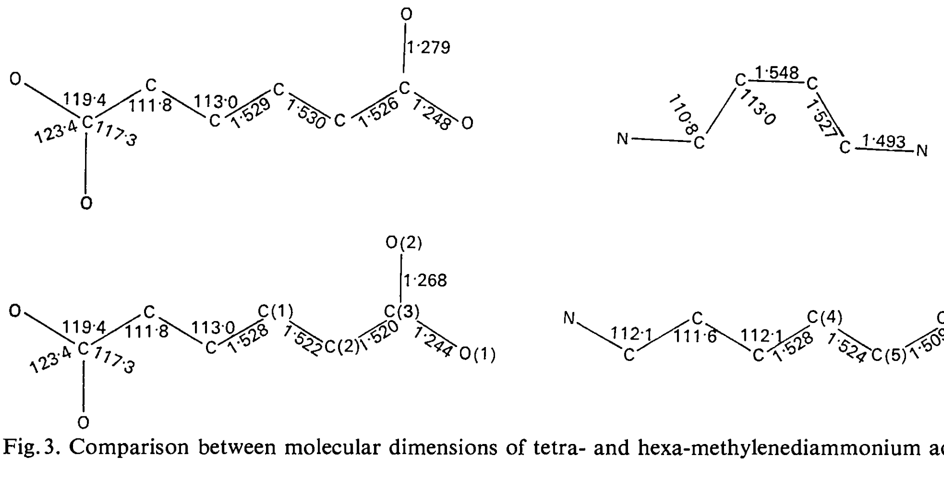

Fig. 3. Comparison between molecular dimensions of tetra- and hexa-methylenediammonium adipate.

and from this plane C(4) is displaced by $-0{\cdot}005$, C(5) by $-0{\cdot}005$, and C(6) by $+0{\cdot}005$ Å. The plane through C(5), C(6) and N is

$$-0{\cdot}7906X-0{\cdot}6096Y+0{\cdot}0573Z=-4{\cdot}0845\,,$$

which is inclined by 1·2° to the plane through C(4), C(5) and C(6), so that the hexamethylenediammonium ion may be regarded as planar within reasonable limits of error.

Consideration of the thermal parameters in Table 1 indicates that there are no particularly anisotropic vibrations; a fuller analysis of these has not been carried out.

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# The Etching of Matched Fracture Faces of Quartz

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Small synthetic quartz crystals from the Bell Telephone Laboratory, U.S.A., grown in molar sodium hydroxide solution at 360–400°C and 20,000 lb.in$^{-2}$ were cleaved along planes parallel to the rhombohedral faces. The cleavage faces were etched in different concentrations of hydrofluoric acid. When they were etched in a solution containing 40 parts of the acid in 60 parts of water, etch spirals together with the usual etch pits were produced. By etching matched cleavage pairs, one-to-one correspondence of the spirals produced was established. It is conjectured that the spirals are associated with the etching of screw dislocations in the crystal.

Efforts were made to cleave these crystals parallel to the prism faces along which it has no natural cleavage. Cleavage (fractured) faces more or less parallel to the prism faces were obtained. Matched pairs of the fractured faces were etched in hydrofluoric acid and one-to-one correspondence in the distribution of etch pits was established. It has thus been shown for the first time that the etch patterns on fractured faces which may not be truly crystallographic planes completely match. By etching the prism fractured faces successively for different periods, and by comparing the etch patterns on the two sides of a thin plate, it was shown that the etch pits reveal the sites of dislocations.

## Introduction

A number of investigators have applied etch methods for gaining information regarding the history of growth of crystals and their dislocation content. Chief amongst them are Gilman & Johnston (1956), Patel & Tolansky (1957), Patel & Goswami (1962) and Patel & Ramanathan (1962). For such studies the cleavage faces are usually used. Tsinzerling & Mironova (1963) and Patel, Bahl & Vagh (1965) have studied fractured rhombohedral $(10\bar{1}1)$ faces of quartz. Patel & Bahl (1965) have reported that spiral pits can be produced at the sites of screw dislocations in graphite by selecting a suitable etchant and the right etching conditions. Since a number of investigators have reported the existence of spiral growths on the rhombohedral faces of quartz crystals, it was thought that some screw dislocations might be threading through these faces. When the crystals are cleaved parallel to these faces, the dislocations may be cut into two and will have their terminations on both the cleavage faces. Patel, Bahl & Vagh have etched such cleavages, but no spiral pits were reported. It was thought that by selecting a proper etchant and conditions of etching, the screw dislocations might be revealed by spiral etch pits developing on the crystal faces as reported by Patel & Bahl (1965). The present paper describes experiments carried out with rhombohedral and prism fractured faces of quartz.

## Experimental

The investigations were carried out on small synthetic quartz crystals obtained from Bell Telephone Laboratories (U.S.A.). They were grown in molar sodium hydroxide solution at a temperature of 360 – 400 °C, and a pressure of 20,000 lb.in$^{-2}$. Selected crystals were cleaved parallel to the rhombohedral faces and the cleavage faces were etched in a number of solutions of