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# Refinement of the Structure of Potassium Heptafluoroniobate, $K_2NbF_7$, from Neutron-Diffraction Data*

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The crystal structure of $K_2NbF_7$, determined originally from two-dimensional X-ray data, has been refined by the method of least-squares with three-dimensional data. The refinement included adjustment of the scattering factors of niobium and potassium relative to the fluorine scattering factor. For potassium the new value $0\cdot370 \pm 0\cdot004$ $cm^{-12}$ was found. Effects of extinction were minimized by omitting from the final refinement a number of the observations, as suggested by systematic analysis of the quality of agreement between calculated and observed $F^2$ values as a function of observed intensity. Shifts of atomic positions by as much as 0·15 Å occurred in the refinement, but the general description of the structure, including the description of the $NbF_7^{2-}$ ion, remains unchanged. The standard errors of the coordinates are about 0·001 Å for the niobium atom and 0·002 to 0·003 Å for the fluorine atoms and the potassium ions. Analysis of the anisotropic thermal parameters shows that the $NbF_7^{2-}$ ion is a nearly ideal case for use of the 'riding' model in calculating bond-length corrections, and the Nb–F bond lengths have been corrected accordingly. Exceptionally large vibrational displacements of two of the fluorine atoms seem plausible in view of the packing in the crystal.

## Introduction

Our interest in the crystal structure of potassium heptafluoroniobate, $K_2NbF_7$, arose in connection with recent work (Keller, 1963) in this laboratory on the Raman and infrared spectroscopy of complex ions of niobium(V) in hydrofluoric acid solutions. The original determination of the $K_2NbF_7$ structure (Hoard, 1939), from two zones of X-ray reflections showed that the $NbF_7^{2-}$ ion has approximately the symmetry $C_{2v}$. The polyhedron of fluorine atoms was visualized as derived from an $NbF_6$ group in the form of a trigonal prism by the addition of a seventh fluorine atom through the center of one square face, followed by some distortion. Although there was no particular reason to question the essential correctness of this structure, we thought it desirable to confirm the coordination about niobium and to refine the structure from three-dimensional neutron data. For $K_2NbF_7$ it is advantageous to employ neutron analysis, rather than X-ray analysis, for two reasons. First, the narrow range of values of the neutron scattering factors of K, Nb, and F nuclei ensures that the structural parameters of all atoms will be determined with nearly the same degree of precision. Second, the low absorption coefficient of $K_2NbF_7$ for thermal neutrons makes it possible to obtain intensity data virtually free from absorption errors.

## Experimental data

### Unit-cell parameters

The unit-cell parameters (Table 1) were redetermined from measurements of angles of Bragg X-ray reflections from a small crystal specimen. The measurements were made on a Picker diffractometer fitted with a General Electric single-crystal orienter. The translations *a*, *b*, *c* were determined from high-order reflections *h*00, 0*k*0, and 00*l* for which the Cu $K\alpha_1$–$\alpha_2$ doublet was resolved. The monoclinic angle $\beta$ was determined directly by measurement of the angles between the reciprocal axes **a*** and **c***. The new cell parameters are close to those of Hoard, but more precisely determined.

Table 1. *Unit-cell parameters of* $K_2NbF_7$

| | Hoard* | This research† |
|---|---|---|
| *a* | 5·85 Å | $5\cdot846 \pm 0\cdot003$ Å |
| *b* | 12·67 | $12\cdot693 \pm 0\cdot006$ |
| *c* | 8·50 | $8\cdot515 \pm 0\cdot004$ |
| $\beta$ | 90° | $90\cdot0° \pm 0\cdot1°$ |
| *Z* | 4 | |
| Space group | $P2_1/c$ | |

* 'Estimated limits of error' in translations are 0·02 to 0·03 Å.

† The wavelengths assumed for Cu $K\alpha_1$ and Cu $K\alpha_2$ radiation were 1·54050 and 1·54434 Å. The indicated error for each parameter is the root-mean-square deviation from the mean of several observations.

### Intensity data

A crystal specimen weighing 18·5 mg was selected from a bottle of reagent-grade material purchased from the Kawecki Chemical Company. Its over-all dimensions were about 1·3, 1·9, and 3·6 mm. Preliminary X-ray precession photography established that it was a single crystal. The crystal was mounted with the *a* axis (3·6 mm dimension) parallel to the spindle axis, then sealed inside a thin-walled tube of quartz, for protection from moisture.

Intensity data were recorded by the $\theta$–$2\theta$ step-scan method with the Oak Ridge automatic neutron dif-

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fractometer (Busing, Smith, Peterson & Levy, 1964). Counts were recorded at 0·1° intervals over a $2\theta$ range of 5° centered around the expected peak position of each reflection. For each point in the scan counts were registered during the period ($\sim$8 seconds) required for a counter monitoring the primary beam to register a preset fixed count. The neutron wavelength was 1·078 Å. For the [100] and [001] zones, data were recorded out to 111° in $2\theta$, the limit of the accessible range. Subsequently the general reflections *hkl* were recorded in successive concentric shells in reciprocal space to the $2\theta$ limit of $\sim$85°, beyond which it appeared that the number of data with intensities above background would be small. Two reflections of high intensity, 400 and 060, were recorded at regular intervals, after every 50 observations, as a check on the performance of the instrument and the stability of the crystal. A number of other reflections were also observed more than once. In all, 1752 observations were made, corresponding to 1355 independent reflections.

### *Preliminary data processing*

In a series of calculations on the CDC 1604 and IBM 7090 computers the raw data on punched paper tape were converted into a set of least-squares data on cards. After a preliminary step* in which the net count $N$ and the background count $B$ under each peak were computed, the calculations* implied by the following equations were performed:

$$F_o^2 = A_c SLN \tag{1}$$

$$\sigma^2_{F2} = (A_c SL)^2[N+(1+r)B] \tag{2}$$

where $F_o^2$ is the observed value of the square of the structure factor; $\sigma^2_{F2}$ is the variance of $F_o^2$ from the counting statistics; $S$ is a scale factor derived from the weight of the crystal specimen and measurements of the intensity of reflection 400 from a standard crystal of sodium chloride; $A_c$ is the correction factor for absorption; $L$ is the Lorentz factor, sin $2\theta$; and $r$ is the ratio of the number of points under the peak to the number of neighboring exterior points used in establishing the background count.

In the calculation of the absorption factors $A_c$ (Wehe, Busing & Levy, 1962) the crystal specimen was described by specifying the equations of 13 surface planes. The value 0·243 cm$^{-1}$, calculated from tabulated neutron cross sections (S.W. Peterson, unpublished) was used for the linear absorption coefficient. The largest factor $A_c$ was 1·044.

For those reflections for which two or more observations were made the average $F_o^2$ values and their appropriate variances were computed, using weights equal to the reciprocals of the variances of the individual observations. Negative values of $F_o^2$ were replaced by zero values. Finally, an empirical correction term $(0{\cdot}03\ F_o^2)^2$ was added to the variance $\sigma^2_{F2}$ of the average $F_o^2$ for each independent reflection. The extra term makes allowance for possible instability in the instrument, errors in corrections for absorption, deficiencies of the structure model, and so forth (see Peterson & Levy, 1957).

The entire procedure followed typically in neutron-diffraction analyses in this laboratory is presented in more detail in an earlier paper on problems and procedures in data collection (Brown & Levy, 1964). The same paper presents a statistical analysis of consistency among the many replicate measurements of the two standard reflections, 400 and 060, of $K_2NbF_7$.

## Refinement

Refinement was carried out by the method of least squares, using various versions of the full-matrix least-squares program of Busing, Martin & Levy (1962). The function minimized was $\Sigma w_{F2}(F_o^2 - F_c^2)^2$, where $F_o^2$ and $F_c^2$ are the observed and calculated structure-factor squares, and $w_{F2}$ is the weight of the observation $F_o^2$, given by $w_{F2} = 1/\sigma^2_{F2}$.

When the 288 data of the [100] and [001] zones had been recorded and processed, a least-squares calculation was made to establish preliminary values of the isotropic thermal parameters $B$ of the various atoms, since the work of Hoard did not provide any thermal parameters. In three cycles of least-squares adjustment, in which Hoard's atomic coordinates were held fixed, the values of $B$ spread from the common assumed input value 1·0 Å$^2$ through the range from 1·03 Å$^2$ (for niobium) to 4·23 Å$^2$. The value of the discrepancy index

$$R_{F2} = \Sigma|F_o^2 - F_c^2|/\Sigma F_o^2$$

was reduced from 0·57 to 0·21 (omitting 28 reflections of uncertain sign).

Fourier syntheses along [100] and [001] were computed with the use of the observed structure factors $F_o$ with the signs of the structure factors $F_c$ calculated after adjustment of the thermal parameters. The maps of nuclear scattering density were very similar to the electron-density maps of Hoard, except for the expected changes in relative peak heights. It was clear at this stage that the Hoard structure is basically correct.

The subsequent least-squares refinement on the three-dimensional data was generally straightforward. Special care was exercised, however, to evaluate the extent of extinction errors in the data and to minimize the effects of these errors. While refining the structural parameters, we were also concerned with the possibility of obtaining more reliable relative values of the scattering factors of potassium, niobium, and fluorine. Accordingly, we carried out six different refinements to convergence, as described in Table 2 and explained below. The convergence in every case was such that the final shift for each parameter was less than 0·02 times the corresponding standard error.

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* The indicated calculations were performed with computer programs of W. R. Busing.

Soon after the three-dimensional refinement was started, comparison of the values $F_o^2$ and $F_c^2$ showed that a number of the values $F_o^2$ for reflections of high intensity were in error because of extinction. To estimate the extent of the errors, we examined plots of the functions $\langle F_o^2\rangle/\langle F_c^2\rangle$ and $\langle|F_o^2-F_c^2|/\sigma_{F_o}\rangle$ against $\langle I_o\rangle$, where $I_o=F_o^2/\sin 2\theta$ and the brackets $\langle\ \rangle$ denote an average value over a small range of $I_o$. In Fig. 1, curves (*a*) and (*b*) show the plots of these functions after refinement to convergence of the atomic coordinates and anisotropic thermal parameters* and the scale factor, when 1354 $F_o^2$ data** were included (refinement *A* in Table 2). The plots suggest that the $F_o^2$ values corresponding to $I_o$ greater than about 40 (in units of $10^{-24}$ cm$^2$) are nearly all low by a few per cent because of extinction. When refinement was carried to convergence omitting the data corresponding to $I_o>40$, the scale factor on $F_o^2$ decreased, and examination of the new fit of the data showed that even more reflections are affected by extinction. Thus, estimation of the extent and magnitude of extinction errors is necessarily an iterative process. In our final refinement (*E*) we have excluded 112 observations, all those with $I_o>19{\cdot}80$. The curves (*c*) and (*d*) of Fig. 1 show the general agreement of $F_o^2$ and $F_c^2$ values in this case. The choice of the final cut-off level of $I_o$ is somewhat arbitrary; but, fortunately, it appears not to be critical. Thus, comparisons of entries *B* and *D* and of entries *C* and *E* of Table 2 show that the goodness of fit as measured by the values of $\sigma_1$, the standard deviation of an observation of unit weight,† is insensitive to a change in the cut-off level of $I_o$ from 26·61 to 19·80. The scale factor is similarly insensitive to this change. Comparison of entries *A*, *B*, and *D* shows that the fit is distinctly poorer when all of the data are used in the refinement. It is of interest that the discrepancy factor, whether calculated on $|F|$ or on $|F|^2$, is a very unreliable index of quality of fit in these refinements.‡

The entries under *F* show approximately the quality of fit which might have been expected if there had been no perturbation from extinction. For this refinement the data corresponding to $I_o>19{\cdot}80$ were corrected by multiplication of each $F_o^2$ value by the reciprocal of the ordinate read from the curve of Fig. 1(*c*) at the appropriate $I_o$.

The shapes of the curves (*a*) and (*c*) of Fig. 1 are not precisely what one might expect. For one thing, it seems that our intensity measurements for the very weak reflections are on the average slightly high be-

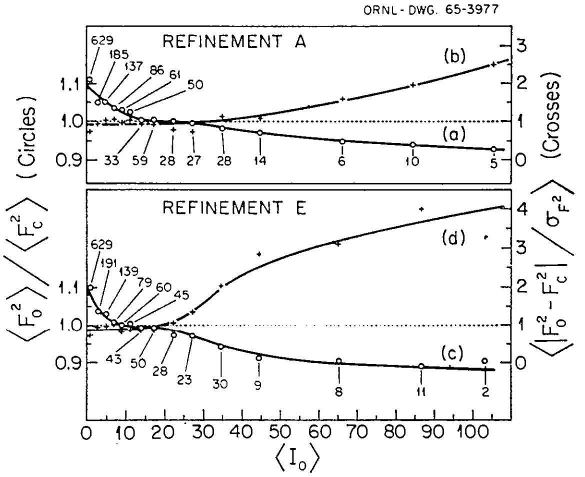

Fig. 1. Curves showing the agreement between the values $F_c^2$ and $F_o^2$ as a function of intensity (see text). The numbers attached to the data points are the numbers of reflections included in the ranges of intensity over which the averages were taken.

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* The thermal parameters $\beta_{ij}$ are the coefficients in the exponent of the temperature factor $\exp-[\beta_{11}h^2+\beta_{22}k^2+\beta_{33}l^2+2\beta_{12}hk+2\beta_{13}hl+2\beta_{23}kl]$.

** Reflection $34\bar{2}$ was omitted from all refinements after it was noticed that the shape of its peak on the recorder chart was anomalous, probably because of an instrumental aberration.

† The quantity $\sigma_1$ is given by the expression $[\Sigma w_{F^2}(F_o{}^2-F_c{}^2)/(n-p)]^{\frac{1}{2}}$, where $n$ is the number of observations and $p$ is the number of parameters adjusted. Unit value is expected for $\sigma_1$ at convergence when the observational errors are normally distributed and correctly estimated and the model is correct.

‡ The fact that the discrepancy factor computed on $|F|$ can equal or exceed the factor computed on $|F|^2$ has been discussed by Brown (1966).

Table 2. *Description of the six different refinements to convergence*

In all six cases the scale factor on $|F_c|$ and the three coordinates and six anisotropic thermal parameters of each atom were adjusted. The scattering factor of fluorine was fixed at 0·55 × $10^{-12}$ cm throughout. The scattering factors of potassium and niobium were held fixed in refinements *A*, *B*, and *D* at 0·35 × $10^{-12}$ cm and 0·691 × $10^{-12}$ cm but were adjusted in *C*, *E*, and *F*. The values of the discrepancy indices in parentheses were computed from all 1355 data

| Refinement | Number of data included | Scattering factors derived (units of $10^{-12}$ cm) | Scale factor derived | $(\Sigma\|\|F_o\|^x-\|F_c\|^x\|)/\Sigma\|F_o\|^x$ $x=2$ | $x=1$ | $\sigma_1$ |
|---|---|---|---|---|---|---|
| *A* | 1355 | not adjusted | 0·984 ± 0·003 | 0·081 | 0·090 | 1·20 |
| *B* | 1278<br>($I_o\le 26{\cdot}61$) | not adjusted | 1·006 ± 0·003 | 0·095<br>(0·090) | 0·103<br>(0·092) | 1·14 |
| *C* | 1278<br>($I_o\le 26{\cdot}61$) | $f_{\rm Nb}=0{\cdot}695\pm0{\cdot}005$<br>$f_{\rm K}=0{\cdot}370\pm0{\cdot}004$ | 1·001 ± 0·004 | 0·094<br>(0·090) | 0·103<br>(0·092) | 1·13 |
| *D* | 1243<br>($I_o\le 19{\cdot}80$) | not adjusted | 1·014 ± 0·004 | 0·104<br>(0·094) | 0·110<br>(0·093) | 1·14 |
| *E* | 1243<br>($I_o\le 19{\cdot}80$) | $f_{\rm Nb}=0{\cdot}692\pm0{\cdot}005$<br>$f_{\rm K}=0{\cdot}370\pm0{\cdot}004$ | 1·009 ± 0·004 | 0·103<br>(0·094) | 0·109<br>(0·093) | 1·13 |
| *F* | 1355<br>(corr. data) | $f_{\rm Nb}=0{\cdot}695\pm0{\cdot}003$<br>$f_{\rm K}=0{\cdot}366\pm0{\cdot}003$ | 1·005 ± 0·003 | 0·070 | — | 1·11 |

cause of some systematic errors of unknown origin, possibly effects of multiple reflection. Although curves (*a*) and (*c*) give a rather exaggerated impression of this effect, curves (*b*) and (*d*) show that the systematic error is not large in relation to the standard error of measurement. At high intensities the ratio $\langle F_o^2\rangle/\langle F_c\rangle$ does not fall so abruptly from unit value as one would expect for an extinction effect. The detailed shape of the curve will be affected, however, by anisotropy in the extinction and, possibly, by some errors from other sources. In any case, we make use of the curve mainly to avoid the inclusion of questionable data in the refinement, not for any quantitative analysis of extinction effects.

The sets of atomic coordinates from the various refinements differ only slightly from each other. From refinements *B*, *C*, *D*, and *E*, the average ratio over all 30 coordinates of the range of coordinate variation to the corresponding coordinate standard error is about 0·3 and the extreme ratio is 0·7. Even when refinement *A* is included, the average and extreme ratios are only 0·6 and 1·2. Between *D* and *E*, which differ only as regards adjustment of $f_{\rm Nb}$ and $f_{\rm K}$, the average ratio is 0·2 and the extreme is 0·4.

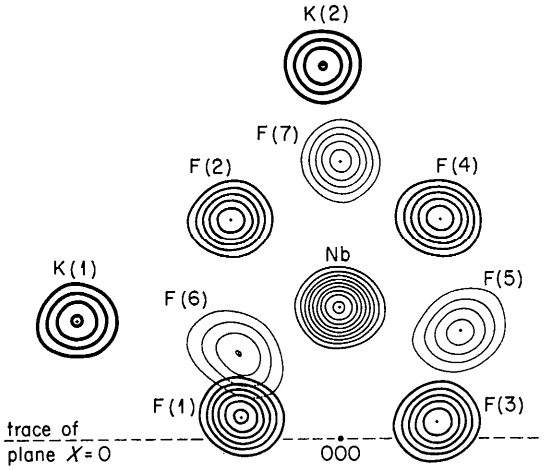

Fig. 2. Final composite three-dimensional Fourier map of the asymmetric unit of the $K_2NbF_7$ structure. The direction of view has direction cosines 0·0, −0·669, −0·743 with respect to the crystal axes **a**, **b**, **c**. The contour interval is 0·5 in units of $10^{-12}$ cmÅ$^{-3}$. The lowest contour is at the 0·5 level.

In general, the thermal parameters are different only by similar small amounts from one refinement to another. The largest shifts noted are the increases of about two standard deviations in the $\beta_{tt}$ parameters of the potassium ions from *B* to *C* and from *D* to *E*, which accompany the increase in the value of $f_{\rm K}$. The inclusion in refinement *A* of the data affected by extinction is compensated for largely by a change in the scale factor, but partly by slight decreases in most of the $\beta_{tt}$.

For no single parameter is there a significant change in the standard error from one refinement to another. It is interesting, however, that the computed errors of the $\beta_{tt}$ parameters of niobium and potassium are all 10 to 15% larger when $f_{\rm Nb}$ and $f_{\rm K}$ are included in the least-squares adjustment, reflecting the strong correlation between the scattering factor and the parameters $\beta_{tt}$ of a given atom.

It would have been unnecessary to adjust the scattering factors $f_{\rm Nb}$ and $f_{\rm K}$, as was done in refinements *C*, *E*, and *F*, if these parameters had already been precisely determined. However, the tabulated values (Bacon, 1963) of $f_{\rm Nb}$, $f_{\rm K}$, and $f_{\rm F}$ are certainly subject to revision. They were determined from powder data, along with the scattering factors of many other elements, by Shull & Wollan (1951), who claimed in general only an accuracy of 3 to 4%. Hughes & Harvey's (1955) estimates of the standard errors of the coherent scattering cross-sections correspond to standard errors of about 1·7, 3·5, and 4·0% in the values $f_{\rm Nb}$, $f_{\rm K}$, and $f_{\rm F}$, respectively. Since scale factors are normally obtained in least-squares refinements of crystal structures with considerably smaller standard errors than these, it seemed to us desirable to attempt refinement of the relative values of the scattering factors. From our results (Table 2) the true value of $f_{\rm K}$ relative to the value 0·55 cm$^{-12}$ for fluorine* appears to be 0·370 ± 0·004 cm$^{-12}$, significantly different from the tabulated value 0·35 cm$^{-12}$. The value of $f_{\rm Nb}$ is not changed significantly. Although these

Table 3. *Final atomic coordinates and thermal parameters of the* $K_2NbF_7$ *crystal structure (from refinement E)*

| | Fractional coordinates × 10⁴ (standard errors × 10⁵) | | | Thermal parameters × 10⁵ (standard errors × 10⁵) | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| Atom | *x* | *y* | *z* | $\beta_{11}$ | $\beta_{22}$ | $\beta_{33}$ | $\beta_{12}$ | $\beta_{13}$ | $\beta_{23}$ |
| Nb | 2718(20) | 1288(9) | 2229(14) | 799(36) | 157(7) | 376(15) | 3(12) | 34(17) | 20(8) |
| K(1) | 2397(46) | 4404(19) | 1833(31) | 1699(82) | 232(15) | 698(37) | −42(27) | −37(41) | −40(18) |
| K(2) | 7626(45) | 2845(22) | 4446(30) | 1343(81) | 424(18) | 590(36) | −8(30) | −1(42) | −98(19) |
| F(1) | 463(28) | 2431(13) | 1994(20) | 1188(46) | 275(10) | 759(22) | −152(18) | 38(27) | 10(13) |
| F(2) | 4573(31) | 2567(14) | 2114(24) | 1198(47) | 240(9) | 1159(28) | 116(18) | 239(29) | 99(14) |
| F(3) | 448(30) | 1072(14) | 3880(21) | 1326(50) | 355(11) | 673(22) | 59(18) | −288(27) | 48(13) |
| F(4) | 4599(31) | 1107(16) | 4101(20) | 1263(50) | 542(13) | 530(21) | 81(20) | 223(27) | 137(14) |
| F(5) | 2166(41) | −207(15) | 2159(30) | 3041(81) | 228(10) | 1682(40) | 296(22) | −1043(47) | −198(16) |
| F(6) | 1875(46) | 1216(21) | 63(25) | 3675(90) | 834(20) | 633(24) | −949(35) | 628(39) | −278(19) |
| F(7) | 5572(31) | 792(15) | 1317(24) | 1336(52) | 407(12) | 927(26) | −142(20) | −337(29) | 15(14) |

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* The choice of the scattering factor of fluorine as reference is arbitrary.

Table 4. *Observed and calculated structure-factor squares*

Reflections are identified by the indices *h* and *l* in the sub-headings and the index *k* under the main heading *K*. For each reflection $|F_c(hkl)|^2$, $|F_o(hkl)|^2$, and the standard error $\sigma_{F^2}$ (*SIG*) are given (all in units of $10^{-26}$ cm$^2$), along with the sign of $F_c(hkl)$. A letter to the left of an index *k* indicates omission of the reflection in the final refinement, because of extinction error (*E*), an aberrant measurement (*A*), or a missing observation (*M*).

| | K | CALC | OBS | SIG | |
|---|---|---|---|---|---|
| ••• | | 0 | K | 0 | ••• |
| | 0 | 44630 | | | + |
| | 2 | 22 | 13 | 4 | - |
| | 4 | 486 | 491 | 19 | - |
| E | 6 | 5590 | 4872 | 146 | - |
| E | 8 | 6231 | 5691 | 174 | + |
| | 10 | 42 | 25 | 20 | - |
| | 12 | 39 | 25 | 24 | - |
| | 14 | 377 | 343 | 26 | - |
| | 16 | 1046 | 1078 | 45 | + |
| | 18 | 189 | 158 | 30 | - |
| ••• | | 0 | K | 1 | ••• |
| | 1 | 46 | 51 | 6 | + |
| E | 2 | 2433 | 2235 | 70 | - |
| | 3 | 44 | 44 | 12 | - |
| | 4 | 4 | 0 | 10 | - |
| | 5 | 388 | 376 | 19 | + |
| E | 6 | 4801 | 4389 | 137 | + |
| | 7 | 38 | 47 | 17 | - |
| | 8 | 1111 | 1062 | 41 | + |
| | 9 | 3 | 5 | 20 | + |
| | 10 | 59 | 81 | 93 | - |
| | 11 | 110 | 99 | 46 | - |
| | 12 | 0 | 0 | 34 | - |
| M | 13 | 321 | 0 | 29 | + |
| | 14 | 465 | 421 | 43 | + |
| | 15 | 145 | 170 | 40 | + |
| | 16 | 20 | 32 | 40 | + |
| | 17 | 109 | 83 | 43 | - |
| | 18 | 23 | 28 | 40 | - |
| | 19 | 192 | 87 | 42 | - |
| ••• | | 0 | K | 2 | ••• |
| E | 0 | 2155 | 1989 | 62 | - |
| | 1 | 71 | 91 | 10 | - |
| | 2 | 14 | 42 | 10 | + |
| | 3 | 224 | 206 | 15 | + |
| E | 4 | 2225 | 2077 | 68 | - |
| E | 5 | 1340 | 1297 | 46 | - |
| | 6 | 175 | 190 | 19 | + |
| | 7 | 1 | 3 | 23 | - |
| | 8 | 995 | 1008 | 41 | - |
| | 9 | 54 | 89 | 22 | - |
| E | 10 | 1973 | 1911 | 80 | + |
| E | 11 | 3442 | 3252 | 118 | + |
| | 12 | 241 | 188 | 37 | - |
| | 13 | 106 | 61 | 35 | + |
| | 14 | 104 | 95 | 36 | + |
| | 15 | 65 | 51 | 40 | + |
| | 16 | 899 | 893 | 59 | - |
| | 17 | 710 | 857 | 56 | - |
| | 18 | 686 | 741 | 54 | + |
| | 19 | 70 | 67 | 41 | + |
| ••• | | 0 | K | 3 | ••• |
| | 1 | 19 | 5 | 10 | + |
| E | 2 | 834 | 817 | 31 | - |
| | 3 | 301 | 282 | 19 | - |
| E | 4 | 2295 | 2140 | 71 | - |
| | 5 | 197 | 215 | 21 | - |
| | 6 | 1 | 20 | 16 | + |
| | 7 | 131 | 101 | 20 | - |
| | 8 | 188 | 182 | 24 | + |
| | 9 | 3 | 1 | 35 | + |
| | 10 | 69 | 82 | 38 | + |
| | 11 | 47 | 48 | 35 | - |
| | 12 | 1074 | 1039 | 55 | - |
| | 13 | 147 | 157 | 37 | - |
| | 14 | 642 | 707 | 50 | - |
| | 15 | 30 | 48 | 41 | - |
| | 16 | 132 | 180 | 43 | + |
| | 17 | 91 | 92 | 41 | + |
| | 18 | 15 | 0 | 39 | + |
| ••• | | 0 | K | 4 | ••• |
| | 0 | 313 | 334 | 20 | + |
| E | 1 | 5679 | 4843 | 150 | + |
| | 2 | 663 | 689 | 30 | - |
| | 3 | 43 | 46 | 17 | - |
| | 4 | 502 | 498 | 28 | + |
| | 5 | 603 | 628 | 49 | + |
| | 6 | 664 | 670 | 34 | + |
| E | 7 | 3098 | 2877 | 95 | - |
| E | 8 | 1729 | 1657 | 71 | + |
| | 9 | 429 | 478 | 45 | + |
| | 10 | 1101 | 1104 | 57 | - |
| | 11 | 335 | 301 | 38 | - |
| | 12 | 381 | 319 | 39 | - |
| | 13 | 171 | 183 | 37 | - |
| | 14 | 1 | 0 | 39 | + |
| | 15 | 1055 | 1064 | 62 | - |
| | 16 | 300 | 227 | 46 | + |
| | 17 | 108 | 101 | 41 | + |
| | 18 | 311 | 334 | 45 | - |
| ••• | | 0 | K | 5 | ••• |
| E | 1 | 2265 | 2135 | 71 | + |
| | 2 | 10 | 28 | 16 | - |
| | 3 | 1157 | 1159 | 45 | - |
| | 4 | 3 | 21 | 18 | + |
| | 5 | 1209 | 1169 | 48 | - |
| | 6 | 1152 | 1138 | 55 | + |
| E | 7 | 2308 | 2224 | 87 | + |
| | 8 | 361 | 426 | 44 | - |
| E | 9 | 3268 | 3197 | 115 | + |
| | 10 | 56 | 54 | 34 | - |
| | 11 | 70 | 72 | 35 | - |
| | 12 | 19 | 0 | 35 | + |
| | 13 | 877 | 939 | 57 | - |
| | 14 | 897 | 891 | 58 | + |
| | 15 | 35 | 5 | 42 | - |
| | 16 | 5 | 20 | 39 | + |
| | 17 | 43 | 68 | 39 | + |
| ••• | | 0 | K | 6 | ••• |
| | 0 | 182 | 187 | 23 | - |
| | 1 | 1864 | 1811 | 64 | - |
| | 2 | 13 | 2 | 20 | + |
| | 3 | 1 | 0 | 20 | - |
| | 4 | 287 | 271 | 36 | + |
| | 5 | 21 | 30 | 36 | + |
| | 6 | 19 | 26 | 38 | - |
| E | 7 | 3275 | 3311 | 118 | + |
| | 8 | 397 | 321 | 40 | - |
| | 9 | 1230 | 1229 | 60 | - |
| | 10 | 347 | 308 | 39 | - |
| | 11 | 72 | 86 | 38 | + |
| | 12 | 354 | 366 | 44 | + |
| | 13 | 5 | 0 | 42 | - |
| | 14 | 55 | 156 | 41 | + |
| E | 15 | 2006 | 2000 | 88 | + |
| | 16 | 31 | 1 | 40 | + |
| | 17 | 99 | 76 | 39 | - |
| ••• | | 0 | K | 7 | ••• |
| | 1 | 14 | 65 | 36 | + |
| E | 2 | 1957 | 1854 | 77 | + |
| | 3 | 1 | 36 | 38 | - |
| | 4 | 360 | 371 | 42 | + |
| | 5 | 719 | 761 | 49 | - |
| | 6 | 193 | 215 | 37 | - |
| | 7 | 255 | 169 | 36 | - |
| | 8 | 289 | 265 | 38 | - |
| | 9 | 108 | 117 | 34 | - |
| | 10 | 20 | 0 | 36 | + |
| | 11 | 250 | 321 | 41 | + |
| | 12 | 4 | 0 | 42 | + |
| | 13 | 57 | 62 | 42 | + |
| | 14 | 155 | 93 | 43 | - |
| | 15 | 58 | 14 | 40 | - |
| | 16 | 0 | 44 | 37 | - |
| ••• | | 0 | K | 8 | ••• |
| | 0 | 12 | 0 | 34 | + |
| | 1 | 215 | 204 | 36 | + |
| | 2 | 583 | 534 | 43 | + |
| | 3 | 1535 | 1582 | 69 | - |
| E | 4 | 2444 | 2372 | 92 | - |
| | 5 | 0 | 34 | 32 | + |
| | 6 | 10 | 10 | 33 | + |
| | 7 | 83 | 104 | 36 | - |
| | 8 | 19 | 0 | 36 | + |
| | 9 | 787 | 736 | 53 | + |
| | 10 | 369 | 399 | 46 | + |
| | 11 | 449 | 459 | 49 | - |
| | 12 | 41 | 5 | 42 | - |
| | 13 | 299 | 282 | 46 | - |
| | 14 | 1 | 40 | 38 | - |
| | 15 | 85 | 198 | 40 | - |
| ••• | | 0 | K | 9 | ••• |
| | 1 | 395 | 402 | 41 | + |
| | 2 | 24 | 29 | 34 | - |
| | 3 | 8 | 0 | 33 | - |
| | 4 | 230 | 259 | 38 | - |
| | 5 | 148 | 134 | 40 | - |
| | 6 | 107 | 90 | 40 | - |
| | 7 | 750 | 726 | 54 | + |
| | 8 | 1 | 0 | 41 | - |
| | 9 | 91 | 138 | 43 | - |
| | 10 | 250 | 292 | 47 | + |
| | 11 | 364 | 419 | 47 | - |
| | 12 | 14 | 0 | 39 | + |
| | 13 | 33 | 70 | 39 | - |
| | 14 | 1 | 15 | 39 | + |
| ••• | | 0 | K | 10 | ••• |
| | 0 | 1758 | 1743 | 77 | + |
| | 1 | 593 | 554 | 48 | - |
| | 2 | 2 | 66 | 39 | - |
| | 3 | 107 | 130 | 40 | - |
| | 4 | 209 | 156 | 42 | - |
| | 5 | 270 | 321 | 45 | + |
| | 6 | 345 | 309 | 47 | - |
| | 7 | 585 | 528 | 53 | + |
| | 8 | 1046 | 1133 | 66 | + |
| | 9 | 104 | 53 | 41 | - |
| | 10 | 19 | 50 | 41 | - |
| | 11 | 57 | 71 | 39 | - |
| | 12 | 10 | 0 | 39 | + |
| ••• | | 0 | K | 11 | ••• |
| | 1 | 858 | 818 | 57 | - |
| | 2 | 451 | 491 | 50 | - |
| | 3 | 88 | 92 | 43 | + |
| | 4 | 12 | 14 | 40 | - |
| | 5 | 313 | 346 | 48 | + |
| | 6 | 81 | 61 | 41 | + |
| | 7 | 5 | 0 | 40 | + |
| | 8 | 285 | 307 | 44 | + |
| | 9 | 64 | 93 | 39 | - |
| | 10 | 67 | 45 | 39 | - |
| ••• | | 0 | K | 12 | ••• |
| | 0 | 172 | 159 | 42 | - |
| | 1 | 31 | 0 | 39 | + |
| | 2 | 95 | 122 | 39 | + |
| | 3 | 439 | 568 | 50 | + |
| | 4 | 229 | 205 | 44 | - |
| | 5 | 475 | 477 | 50 | - |
| | 6 | 139 | 203 | 41 | + |
| | 7 | 215 | 198 | 42 | - |
| ••• | | 1 | K | 0 | ••• |
| | 0 | 95 | 67 | 7 | - |
| E | 1 | 1378 | 1320 | 42 | + |
| | 2 | 22 | 27 | 9 | - |
| E | 3 | 1429 | 1311 | 43 | + |
| | 4 | 81 | 98 | 12 | + |
| E | 5 | 1025 | 932 | 34 | - |
| | 6 | 222 | 214 | 18 | + |
| E | 7 | 2438 | 2268 | 75 | - |
| | 8 | 145 | 129 | 20 | + |
| | 9 | 619 | 609 | 32 | + |
| | 10 | 34 | 68 | 27 | + |
| | 11 | 158 | 172 | 29 | - |
| | 12 | 76 | 62 | 24 | - |
| | 13 | 903 | 921 | 55 | - |
| | 14 | 114 | 127 | 26 | - |
| E | 15 | 1970 | 2068 | 76 | - |
| | 16 | 22 | 86 | 29 | - |
| | 17 | 159 | 146 | 42 | + |
| | 18 | 94 | 91 | 41 | + |
| | 19 | 1 | 0 | 40 | + |
| ••• | | 1 | K | 1 | ••• |
| E | 1 | 587 | 569 | 21 | + |
| | 2 | 127 | 145 | 12 | + |
| | 3 | 429 | 404 | 18 | - |
| | 4 | 201 | 202 | 15 | + |
| | 5 | 144 | 157 | 16 | + |
| E | 6 | 1323 | 1236 | 45 | - |
| E | 7 | 2896 | 2697 | 88 | + |
| | 8 | 534 | 537 | 28 | + |
| E | 9 | 2416 | 2221 | 85 | + |
| | 10 | 152 | 110 | 39 | - |
| | 11 | 649 | 590 | 47 | - |
| | 12 | 98 | 87 | 40 | + |
| | 13 | 537 | 547 | 45 | - |
| | 14 | 358 | 319 | 42 | - |
| | 15 | 1 | 0 | 40 | + |
| ••• | | 1 | K | -1 | ••• |
| E | 1 | 980 | 938 | 31 | - |
| | 2 | 134 | 152 | 12 | - |
| | 3 | 37 | 92 | 13 | + |
| | 4 | 571 | 531 | 23 | - |
| | 5 | 13 | 46 | 14 | + |
| | 6 | 625 | 605 | 28 | - |
| | 7 | 933 | 1004 | 39 | - |
| | 8 | 21 | 52 | 19 | + |
| | 9 | 704 | 736 | 44 | - |
| | 10 | 455 | 565 | 46 | + |
| | 11 | 7 | 0 | 37 | + |
| | 12 | 64 | 50 | 35 | - |
| | 13 | 192 | 136 | 39 | + |
| | 14 | 5 | 0 | 37 | + |
| | 15 | 69 | 67 | 41 | - |
| ••• | | 1 | K | 2 | ••• |
| E | 0 | 1002 | 922 | 32 | + |
| | 1 | 242 | 275 | 15 | - |
| | 2 | 93 | 81 | 13 | - |
| E | 3 | 1569 | 1434 | 48 | + |
| | 4 | 192 | 215 | 16 | + |
| | 5 | 38 | 53 | 15 | + |
| | 6 | 2 | 0 | 16 | - |
| | 7 | 185 | 187 | 27 | + |
| | 8 | 81 | 100 | 21 | + |
| | 9 | 295 | 279 | 27 | - |
| | 10 | 230 | 252 | 41 | - |
| | 11 | 245 | 331 | 40 | + |
| | 12 | 160 | 238 | 36 | - |
| | 13 | 1486 | 1404 | 66 | + |
| | 14 | 48 | 85 | 36 | + |
| | 15 | 640 | 664 | 53 | + |
| ••• | | 1 | K | -2 | ••• |
| | 0 | 417 | 445 | 19 | - |
| E | 1 | 1021 | 904 | 32 | - |
| | 2 | 0 | 3 | 10 | - |
| | 3 | 31 | 72 | 12 | - |
| | 4 | 780 | 716 | 28 | - |
| | 5 | 172 | 157 | 19 | - |
| | 6 | 90 | 75 | 19 | - |
| | 7 | 534 | 470 | 34 | + |
| | 8 | 6 | 50 | 19 | - |
| | 9 | 167 | 149 | 25 | + |
| | 10 | 123 | 187 | 40 | + |
| | 11 | 451 | 441 | 44 | + |
| | 12 | 77 | 159 | 36 | + |
| | 13 | 762 | 749 | 52 | + |
| | 14 | 59 | 58 | 38 | - |
| | 15 | 84 | 164 | 42 | + |
| ••• | | 1 | K | 3 | ••• |
| | 1 | 21 | 55 | 13 | + |
| E | 2 | 958 | 875 | 33 | + |
| | 3 | 200 | 221 | 17 | - |
| | 4 | 1086 | 1022 | 38 | + |
| | 5 | 774 | 843 | 34 | - |
| | 6 | 237 | 221 | 20 | + |
| E | 7 | 1532 | 1451 | 53 | - |
| | 8 | 85 | 70 | 23 | - |
| | 9 | 4 | 22 | 36 | - |
| | 10 | 74 | 38 | 53 | - |
| | 11 | 543 | 529 | 44 | + |
| | 12 | 80 | 77 | 35 | - |
| | 13 | 4 | 0 | 35 | - |
| | 14 | 299 | 239 | 42 | + |
| ••• | | 1 | K | -3 | ••• |
| E | 1 | 1593 | 1463 | 49 | + |
| | 2 | 127 | 163 | 15 | + |
| | 3 | 0 | 41 | 14 | - |
| | 4 | 736 | 753 | 32 | + |
| E | 5 | 1292 | 1297 | 47 | - |
| | 6 | 3 | 0 | 17 | - |
| | 7 | 939 | 918 | 38 | + |
| | 8 | 437 | 399 | 28 | + |
| | 9 | 3 | 43 | 37 | - |
| | 10 | 42 | 27 | 46 | - |
| | 11 | 18 | 52 | 34 | + |
| | 12 | 421 | 461 | 44 | + |
| | 13 | 230 | 225 | 41 | - |
| | 14 | 20 | 40 | 38 | - |
| ••• | | 1 | K | 4 | ••• |
| | 0 | 671 | 732 | 30 | - |
| | 1 | 362 | 387 | 22 | + |
| | 2 | 67 | 88 | 18 | - |
| | 3 | 1 | 28 | 16 | - |
| | 4 | 3 | 4 | 16 | - |
| | 5 | 368 | 351 | 23 | - |
| | 6 | 49 | 42 | 21 | - |
| | 7 | 468 | 538 | 40 | - |
| | 8 | 1178 | 1145 | 58 | - |
| | 9 | 107 | 91 | 47 | + |
| | 10 | 487 | 503 | 43 | + |
| | 11 | 174 | 132 | 36 | - |
| | 12 | 107 | 54 | 36 | + |
| | 13 | 532 | 556 | 48 | - |
| | 14 | 242 | 324 | 44 | + |
| ••• | | 1 | K | -4 | ••• |
| E | 0 | 2746 | 2402 | 78 | + |
| | 1 | 954 | 903 | 35 | + |
| E | 2 | 1316 | 1212 | 44 | + |
| | 3 | 1093 | 1117 | 41 | + |
| | 4 | 215 | 204 | 20 | - |
| | 5 | 979 | 942 | 38 | - |
| | 6 | 104 | 84 | 20 | - |
| | 7 | 1107 | 1134 | 54 | - |
| | 8 | 75 | 73 | 40 | + |
| | 9 | 181 | 142 | 44 | + |
| | 10 | 213 | 260 | 38 | - |
| | 11 | 13 | 0 | 34 | + |
| | 12 | 17 | 42 | 33 | + |
| | 13 | 356 | 328 | 43 | - |
| | 14 | 32 | 70 | 41 | + |
| ••• | | 1 | K | 5 | ••• |
| E | 1 | 2262 | 2058 | 76 | + |
| E | 2 | 1378 | 1410 | 58 | + |
| | 3 | 150 | 145 | 28 | - |
| | 4 | 436 | 409 | 35 | - |
| | 5 | 27 | 69 | 29 | - |
| E | 6 | 3782 | 3472 | 113 | - |
| | 7 | 258 | 237 | 42 | - |
| | 8 | 115 | 118 | 44 | - |
| | 9 | 630 | 601 | 44 | + |
| | 10 | 1148 | 1175 | 59 | + |
| | 11 | 389 | 352 | 41 | - |
| | 12 | 85 | 128 | 37 | + |
| | 13 | 55 | 73 | 40 | + |
| ••• | | 1 | K | -5 | ••• |
| | 1 | 377 | 443 | 32 | - |
| E | 2 | 1345 | 1283 | 54 | + |
| | 3 | 116 | 139 | 20 | + |
| | 4 | 16 | 42 | 27 | - |
| | 5 | 15 | 30 | 29 | + |
| | 6 | 1341 | 1330 | 52 | - |
| | 7 | 130 | 106 | 41 | - |
| | 8 | 435 | 474 | 46 | - |
| | 9 | 0 | 0 | 34 | + |
| | 10 | 57 | 88 | 36 | + |
| | 11 | 246 | 149 | 39 | + |
| | 12 | 95 | 107 | 37 | - |
| | 13 | 381 | 422 | 48 | + |
| ••• | | 1 | K | 6 | ••• |
| E | 0 | 8130 | 7078 | 219 | + |
| | 1 | 41 | 76 | 31 | - |
| | 2 | 9 | 0 | 21 | - |
| | 3 | 493 | 526 | 31 | - |
| | 4 | 129 | 88 | 38 | - |
| | 5 | 93 | 109 | 39 | + |
| | 6 | 412 | 432 | 44 | - |
| | 7 | 747 | 775 | 50 | + |
| | 8 | 1701 | 1665 | 72 | + |
| | 9 | 171 | 173 | 37 | + |
| | 10 | 3 | 0 | 34 | - |
| | 11 | 1 | 0 | 36 | - |
| | 12 | 279 | 279 | 45 | - |
| ••• | | 1 | K | -6 | ••• |
| E | 0 | 2312 | 2437 | 82 | - |
| | 1 | 382 | 384 | 35 | + |
| | 2 | 49 | 63 | 21 | - |
| | 3 | 453 | 441 | 29 | + |
| | 4 | 5 | 46 | 35 | - |
| | 5 | 504 | 466 | 44 | + |
| | 6 | 45 | 76 | 37 | + |
| | 7 | 8 | 3 | 37 | - |
| | 8 | 60 | 130 | 47 | - |
| | 9 | 282 | 311 | 40 | - |
| | 10 | 0 | 59 | 33 | + |
| | 11 | 40 | 3 | 38 | - |
| | 12 | 95 | 99 | 41 | + |
| ••• | | 1 | K | 7 | ••• |
| | 1 | 1129 | 1154 | 60 | - |
| | 2 | 44 | 100 | 38 | - |
| | 3 | 2 | 0 | 36 | + |
| | 4 | 20 | 0 | 37 | - |
| | 5 | 501 | 592 | 44 | + |
| | 6 | 401 | 350 | 39 | + |
| | 7 | 92 | 122 | 36 | + |
| | 8 | 7 | 0 | 34 | + |
| | 9 | 4 | 15 | 35 | - |
| | 10 | 43 | 80 | 37 | + |
| | 11 | 44 | 51 | 41 | + |
| ••• | | 1 | K | -7 | ••• |
| | 1 | 5 | 44 | 37 | - |
| | 2 | 27 | 80 | 37 | + |
| | 3 | 10 | 25 | 36 | + |
| | 4 | 5 | 37 | 36 | + |
| | 5 | 55 | 117 | 34 | - |
| | 6 | 467 | 486 | 41 | + |
| | 7 | 497 | 481 | 43 | + |
| | 8 | 39 | 60 | 34 | - |
| | 9 | 461 | 406 | 43 | - |
| | 10 | 4 | 36 | 37 | + |
| | 11 | 87 | 42 | 44 | - |
| ••• | | 1 | K | 8 | ••• |
| | 0 | 464 | 452 | 42 | - |
| | 1 | 166 | 121 | 36 | + |
| | 2 | 1 | 0 | 33 | + |
| | 3 | 8 | 29 | 33 | + |
| | 4 | 223 | 175 | 38 | - |
| | 5 | 448 | 478 | 42 | - |
| | 6 | 7 | 0 | 34 | - |
| | 7 | 118 | 141 | 35 | - |
| | 8 | 90 | 161 | 37 | - |
| | 9 | 1 | 0 | 39 | - |
| | 10 | 443 | 450 | 51 | + |
| ••• | | 1 | K | -8 | ••• |
| | 0 | 284 | 232 | 37 | + |
| | 1 | 167 | 198 | 36 | + |
| | 2 | 18 | 33 | 32 | + |
| | 3 | 65 | 109 | 39 | - |
| | 4 | 602 | 582 | 43 | - |
| | 5 | 439 | 465 | 41 | - |
| | 6 | 282 | 356 | 38 | + |
| | 7 | 72 | 45 | 36 | - |
| | 8 | 964 | 992 | 58 | + |
| | 9 | 161 | 138 | 42 | + |
| | 10 | 1 | 11 | 42 | - |
| ••• | | 1 | K | 9 | ••• |
| | 1 | 22 | 30 | 35 | - |
| | 2 | 540 | 594 | 46 | + |
| | 3 | 16 | 33 | 35 | + |
| | 4 | 100 | 185 | 36 | - |
| | 5 | 49 | 4 | 37 | + |
| | 6 | 11 | 60 | 38 | - |
| | 7 | 25 | 0 | 40 | - |
| ••• | | 1 | K | -9 | ••• |
| | 1 | 64 | 89 | 35 | - |
| | 2 | 1553 | 1514 | 69 | + |
| | 3 | 1 | 0 | 34 | + |
| | 4 | 59 | 90 | 36 | - |
| | 5 | 150 | 137 | 39 | + |
| | 6 | 1066 | 1129 | 61 | - |
| | 7 | 11 | 69 | 40 | + |
| ••• | | 1 | K | 10 | ••• |
| | 0 | 632 | 543 | 51 | + |
| | 1 | 1834 | 1797 | 80 | + |
| | 2 | 10 | 36 | 41 | - |
| | 3 | 478 | 540 | 49 | + |
| | 4 | 252 | 336 | 47 | - |
| ••• | | 1 | K | -10 | ••• |
| | 0 | 46 | 91 | 41 | - |
| | 1 | 59 | 110 | 41 | + |
| | 2 | 0 | 49 | 39 | - |
| | 3 | 59 | 29 | 41 | - |
| | 4 | 458 | 442 | 48 | + |
| ••• | | 2 | K | 0 | ••• |
| | 0 | 7 | 0 | 15 | + |
| | 1 | 83 | 67 | 12 | + |
| E | 2 | 1048 | 953 | 34 | - |
| | 3 | 418 | 413 | 20 | + |
| | 4 | 103 | 127 | 16 | + |
| | 5 | 38 | 36 | 22 | + |
| E | 6 | 4470 | 4083 | 129 | - |
| | 7 | 251 | 259 | 22 | - |
| | 8 | 173 | 143 | 22 | + |
| | 9 | 397 | 381 | 31 | - |
| | 10 | 14 | 60 | 27 | + |
| | 11 | 114 | 144 | 29 | - |
| E | 12 | 3255 | 3047 | 102 | + |
| | 13 | 19 | 17 | 25 | + |
| | 14 | 1620 | 1665 | 65 | - |
| | 15 | 8 | 33 | 29 | - |
| | 16 | 4 | 0 | 41 | - |
| | 17 | 42 | 75 | 42 | - |
| | 18 | 2 | 29 | 39 | - |
| ••• | | 2 | K | 1 | ••• |
| | 1 | 161 | 186 | 14 | - |
| | 2 | 3 | 44 | 12 | - |
| E | 3 | 1701 | 1552 | 52 | + |
| | 4 | 803 | 810 | 32 | - |
| | 5 | 345 | 341 | 22 | + |
| E | 6 | 2136 | 2009 | 74 | - |
| | 7 | 969 | 999 | 48 | - |
| | 8 | 6 | 0 | 29 | + |
| | 9 | 1079 | 1122 | 58 | - |
| | 10 | 101 | 114 | 48 | + |
| | 11 | 238 | 205 | 40 | + |
| | 12 | 339 | 359 | 40 | - |
| | 13 | 1181 | 1222 | 63 | + |
| | 14 | 549 | 542 | 49 | - |
| | 15 | 57 | 136 | 43 | - |
| ••• | | 2 | K | -1 | ••• |
| E | 1 | 1508 | 1362 | 45 | - |
| E | 2 | 1393 | 1260 | 43 | + |
| | 3 | 588 | 597 | 25 | + |
| E | 4 | 2055 | 1889 | 63 | + |
| | 5 | 56 | 65 | 18 | + |
| | 6 | 656 | 657 | 30 | + |
| | 7 | 16 | 53 | 26 | - |
| | 8 | 431 | 412 | 39 | - |
| E | 9 | 2658 | 2482 | 94 | - |
| | 10 | 140 | 79 | 40 | - |
| | 11 | 1 | 4 | 34 | + |
| | 12 | 203 | 250 | 38 | + |
| | 13 | 33 | 36 | 36 | + |
| | 14 | 598 | 541 | 50 | + |
| | 15 | 360 | 464 | 51 | + |
| ••• | | 2 | K | 2 | ••• |
| | 0 | 623 | 606 | 26 | + |
| | 1 | 40 | 54 | 14 | + |
| E | 2 | 4561 | 3995 | 125 | + |
| E | 3 | 4066 | 3487 | 110 | + |
| E | 4 | 1328 | 1310 | 53 | - |
| E | 5 | 2135 | 1943 | 65 | - |
| | 6 | 471 | 493 | 34 | + |
| | 7 | 363 | 374 | 34 | - |
| | 8 | 91 | 76 | 34 | - |
| | 9 | 65 | 130 | 41 | + |
| | 10 | 220 | 226 | 41 | + |
| | 11 | 1547 | 1527 | 69 | + |
| | 12 | 479 | 518 | 45 | - |
| | 13 | 208 | 183 | 39 | - |
| | 14 | 281 | 353 | 43 | + |
| ••• | | 2 | K | -2 | ••• |
| | 0 | 216 | 268 | 17 | - |
| | 1 | 187 | 180 | 16 | + |
| E | 2 | 2608 | 2454 | 79 | + |
| E | 3 | 1414 | 1470 | 50 | - |
| E | 4 | 1954 | 1786 | 60 | - |
| | 5 | 844 | 875 | 36 | + |
| E | 6 | 1703 | 1637 | 64 | + |
| | 7 | 32 | 18 | 27 | + |
| | 8 | 1 | 0 | 30 | + |
| | 9 | 295 | 282 | 44 | + |
| | 10 | 152 | 84 | 41 | + |
| | 11 | 196 | 226 | 39 | - |
| | 12 | 1697 | 1606 | 72 | - |
| | 13 | 261 | 308 | 39 | + |
| | 14 | 21 | 10 | 38 | + |
| ••• | | 2 | K | 3 | ••• |
| | 1 | 6 | 3 | 15 | + |
| E | 2 | 2094 | 2040 | 68 | - |
| | 3 | 35 | 34 | 16 | - |
| | 4 | 17 | 12 | 17 | + |
| | 5 | 240 | 244 | 28 | - |
| | 6 | 668 | 746 | 33 | + |
| | 7 | 786 | 806 | 46 | + |
| E | 8 | 2108 | 2044 | 82 | + |
| | 9 | 289 | 411 | 42 | + |
| | 10 | 44 | 0 | 37 | - |
| | 11 | 124 | 128 | 36 | + |
| | 12 | 267 | 252 | 40 | + |
| | 13 | 433 | 444 | 44 | - |
| | 14 | 83 | 86 | 42 | + |
| ••• | | 2 | K | -3 | ••• |
| | 1 | 199 | 176 | 18 | - |
| E | 2 | 4789 | 4310 | 135 | + |
| | 3 | 81 | 53 | 17 | + |
| | 4 | 0 | 0 | 16 | - |
| | 5 | 310 | 281 | 30 | + |
| E | 6 | 3405 | 3093 | 100 | - |
| | 7 | 1239 | 1148 | 56 | + |
| | 8 | 773 | 878 | 53 | - |
| | 9 | 413 | 389 | 45 | + |
| | 10 | 1236 | 1209 | 60 | + |
| | 11 | 7 | 0 | 34 | + |
| | 12 | 112 | 85 | 40 | + |
| | 13 | 146 | 158 | 40 | - |
| | 14 | 825 | 842 | 58 | - |
| ••• | | 2 | K | 4 | ••• |
| E | 0 | 4721 | 4195 | 132 | - |
| | 1 | 39 | 31 | 17 | + |
| E | 2 | 1374 | 1300 | 47 | - |
| | 3 | 608 | 566 | 28 | - |
| E | 4 | 1876 | 1931 | 73 | + |
| | 5 | 111 | 72 | 30 | + |
| | 6 | 15 | 0 | 32 | + |
| | 7 | 662 | 691 | 46 | - |
| | 8 | 418 | 487 | 46 | - |
| | 9 | 656 | 613 | 47 | + |
| | 10 | 439 | 436 | 42 | - |
| | 11 | 0 | 0 | 33 | - |
| | 12 | 201 | 257 | 41 | + |
| E | 13 | 2130 | 2095 | 88 | + |
| ••• | | 2 | K | -4 | ••• |
| E | 0 | 4722 | 4350 | 137 | - |
| | 1 | 415 | 446 | 24 | - |
| | 2 | 560 | 595 | 28 | - |
| | 3 | 105 | 89 | 19 | + |
| E | 4 | 5622 | 5109 | 166 | + |
| | 5 | 1030 | 1002 | 41 | - |
| | 6 | 35 | 52 | 30 | + |
| | 7 | 944 | 866 | 53 | + |
| | 8 | 822 | 840 | 54 | - |
| | 9 | 283 | 269 | 42 | - |
| | 10 | 625 | 566 | 47 | - |
| | 11 | 95 | 92 | 35 | + |
| | 12 | 1248 | 1236 | 63 | + |
| E | 13 | 2786 | 2752 | 107 | - |
| ••• | | 2 | K | 5 | ••• |
| | 1 | 238 | 272 | 23 | - |
| | 2 | 614 | 602 | 41 | + |
| | 3 | 13 | 24 | 20 | + |
| | 4 | 126 | 134 | 32 | + |
| | 5 | 12 | 28 | 34 | + |
| | 6 | 757 | 750 | 50 | - |
| | 7 | 833 | 804 | 53 | - |
| | 8 | 233 | 154 | 37 | - |
| | 9 | 1 | 17 | 32 | - |
| | 10 | 1 | 80 | 32 | - |
| | 11 | 59 | 103 | 35 | + |
| | 12 | 172 | 145 | 42 | - |
| ••• | | 2 | K | -5 | ••• |
| | 1 | 52 | 71 | 19 | + |
| | 2 | 477 | 525 | 36 | - |
| | 3 | 6 | 0 | 20 | - |
| | 4 | 656 | 668 | 42 | - |
| | 5 | 608 | 535 | 42 | - |
| | 6 | 325 | 311 | 42 | + |
| | 7 | 824 | 760 | 53 | - |
| | 8 | 44 | 28 | 35 | - |
| | 9 | 0 | 72 | 33 | + |
| | 10 | 130 | 152 | 37 | + |
| | 11 | 261 | 242 | 41 | + |
| | 12 | 66 | 84 | 39 | + |
| ••• | | 2 | K | 6 | ••• |
| | 0 | 91 | 113 | 27 | - |
| | 1 | 22 | 73 | 25 | - |
| | 2 | 194 | 172 | 40 | - |
| E | 3 | 2948 | 2722 | 102 | + |
| | 4 | 949 | 932 | 55 | + |
| | 5 | 1126 | 1225 | 62 | - |
| | 6 | 214 | 229 | 38 | + |
| | 7 | 3 | 15 | 34 | + |
| | 8 | 60 | 89 | 35 | + |
| | 9 | 101 | 42 | 35 | - |
| | 10 | 1 | 0 | 34 | - |
| | 11 | 4 | 53 | 37 | - |
| ••• | | 2 | K | -6 | ••• |
| | 0 | 298 | 322 | 28 | - |
| E | 1 | 1759 | 1734 | 72 | + |
| | 2 | 192 | 240 | 37 | - |
| | 3 | 1009 | 1073 | 56 | - |
| | 4 | 276 | 422 | 42 | + |
| | 5 | 920 | 992 | 56 | + |
| | 6 | 55 | 0 | 36 | + |
| | 7 | 999 | 871 | 52 | - |
| | 8 | 52 | 78 | 35 | - |
| | 9 | 405 | 432 | 42 | + |
| | 10 | 152 | 154 | 38 | - |
| | 11 | 38 | 0 | 39 | + |
| | 12 | 1 | 0 | 43 | + |
| ••• | | 2 | K | 7 | ••• |
| | 1 | 741 | 741 | 50 | + |
| | 2 | 673 | 663 | 48 | + |
| | 3 | 116 | 103 | 35 | - |
| | 4 | 518 | 457 | 42 | + |
| | 5 | 0 | 43 | 34 | + |
| | 6 | 732 | 668 | 47 | - |
| | 7 | 113 | 65 | 36 | + |
| | 8 | 77 | 16 | 36 | - |
| | 9 | 515 | 527 | 46 | + |
| | 10 | 7 | 34 | 38 | - |
| ••• | | 2 | K | -7 | ••• |
| | 1 | 660 | 624 | 48 | + |
| | 2 | 24 | 38 | 35 | + |
| | 3 | 48 | 0 | 35 | - |
| | 4 | 28 | 54 | 33 | - |
| | 5 | 56 | 47 | 33 | - |
| | 6 | 231 | 240 | 37 | + |
| | 7 | 0 | 55 | 34 | - |
| | 8 | 166 | 164 | 37 | + |
| | 9 | 151 | 108 | 38 | + |
| | 10 | 20 | 16 | 41 | + |
| ••• | | 2 | K | 8 | ••• |
| | 0 | 364 | 334 | 41 | + |
| | 1 | 31 | 34 | 34 | - |
| | 2 | 283 | 346 | 39 | + |
| | 3 | 230 | 286 | 40 | - |
| | 4 | 147 | 86 | 36 | - |
| | 5 | 923 | 909 | 53 | + |
| | 6 | 289 | 260 | 38 | - |
| | 7 | 146 | 154 | 40 | + |
| | 8 | 659 | 725 | 52 | - |
| | 9 | 6 | 0 | 41 | - |
| ••• | | 2 | K | -8 | ••• |
| | 0 | 15 | 34 | 34 | - |
| | 1 | 583 | 586 | 44 | + |
| | 2 | 386 | 363 | 40 | + |
| E | 3 | 3376 | 2989 | 110 | + |
| | 4 | 59 | 32 | 35 | + |
| | 5 | 1641 | 1584 | 72 | - |
| | 6 | 6 | 0 | 36 | - |
| | 7 | 783 | 804 | 53 | - |
| | 8 | 1112 | 1096 | 63 | - |
| | 9 | 89 | 115 | 43 | - |
| ••• | | 2 | K | 9 | ••• |
| | 1 | 544 | 589 | 47 | - |
| | 2 | 112 | 129 | 38 | + |
| | 3 | 99 | 126 | 39 | + |
| | 4 | 1 | 0 | 37 | + |
| | 5 | 186 | 242 | 42 | + |
| | 6 | 2 | 27 | 42 | + |
| ••• | | 2 | K | -9 | ••• |
| E | 1 | 2159 | 2173 | 89 | - |
| | 2 | 144 | 159 | 38 | - |
| | 3 | 442 | 388 | 45 | + |
| | 4 | 8 | 0 | 38 | - |
| E | 5 | 2214 | 2075 | 87 | + |
| | 6 | 84 | 48 | 42 | + |
| ••• | | 2 | K | 10 | ••• |
| | 0 | 141 | 146 | 46 | + |
| | 1 | 194 | 94 | 50 | + |
| ••• | | 2 | K | -10 | ••• |
| | 0 | 650 | 589 | 55 | + |
| | 1 | 0 | 70 | 41 | - |
| ••• | | 3 | K | 0 | ••• |
| | 0 | 974 | 1039 | 39 | - |
| | 1 | 710 | 729 | 31 | + |
| | 2 | 206 | 229 | 20 | - |
| E | 3 | 2952 | 2673 | 93 | - |
| | 4 | 23 | 26 | 24 | - |
| E | 5 | 1381 | 1403 | 51 | + |
| | 6 | 60 | 21 | 28 | + |
| | 7 | 583 | 605 | 34 | - |
| | 8 | 0 | 30 | 26 | + |
| | 9 | 362 | 425 | 33 | + |
| | 10 | 41 | 25 | 24 | - |
| | 11 | 271 | 289 | 29 | + |
| | 12 | 70 | 99 | 25 | - |
| E | 13 | 2745 | 2630 | 103 | + |

Table 4 (*cont.*)

| | K | CALC | OBS | SIG | |
|---|---|---|---|---|---|
| ••• 3 K 0 ••• | | | | | |
| | 14 | 3 | 17 | 28 | - |
| | 15 | 13 | 66 | 30 | - |
| | 16 | 1 | 0 | 43 | + |
| | 17 | 211 | 190 | 43 | - |
| | 18 | 5 | 0 | 40 | + |
| ••• 3 K 1 ••• | | | | | |
| | 1 | 1127 | 1047 | 40 | - |
| E | 2 | 1855 | 1771 | 60 | - |
| | 3 | 77 | 77 | 18 | + |
| | 4 | 151 | 168 | 21 | - |
| | 5 | 1 | 27 | 26 | + |
| | 6 | 1303 | 1225 | 56 | + |
| | 7 | 308 | 312 | 38 | - |
| | 8 | 34 | 111 | 38 | + |
| | 9 | 436 | 412 | 45 | - |
| | 10 | 809 | 795 | 51 | - |
| | 11 | 64 | 108 | 34 | + |
| | 12 | 73 | 111 | 36 | - |
| | 13 | 0 | 0 | 38 | - |
| | 14 | 14 | 104 | 41 | - |
| ••• 3 K -1 ••• | | | | | |
| | 1 | 5 | 65 | 24 | + |
| | 2 | 452 | 480 | 24 | - |
| | 3 | 4 | 0 | 16 | - |
| | 4 | 534 | 486 | 27 | - |
| | 5 | 284 | 294 | 31 | + |
| E | 6 | 1698 | 1655 | 68 | + |
| | 7 | 328 | 337 | 38 | + |
| | 8 | 108 | 162 | 38 | + |
| | 9 | 57 | 90 | 37 | + |
| | 10 | 678 | 689 | 48 | + |
| | 11 | 318 | 343 | 41 | - |
| | 12 | 206 | 241 | 39 | - |
| | 13 | 25 | 0 | 37 | + |
| | 14 | 76 | 102 | 43 | + |
| ••• 3 K 2 ••• | | | | | |
| E | 0 | 5633 | 4798 | 150 | - |
| | 1 | 16 | 45 | 16 | + |
| | 2 | 560 | 504 | 27 | - |
| E | 3 | 2334 | 2275 | 76 | + |
| E | 4 | 5330 | 4700 | 148 | + |
| | 5 | 618 | 622 | 31 | - |
| | 6 | 544 | 467 | 40 | + |
| | 7 | 62 | 65 | 35 | - |
| | 8 | 53 | 135 | 43 | - |
| | 9 | 1228 | 1202 | 61 | - |
| | 10 | 788 | 718 | 50 | - |
| | 11 | 54 | 38 | 35 | + |
| | 12 | 1133 | 1034 | 60 | + |
| | 13 | 1636 | 1702 | 78 | - |
| | 14 | 117 | 186 | 46 | - |
| ••• 3 K -2 ••• | | | | | |
| E | 0 | 1329 | 1451 | 51 | + |
| | 1 | 20 | 8 | 16 | + |
| E | 2 | 1561 | 1488 | 52 | + |
| | 3 | 803 | 892 | 36 | + |
| A | 4 | 68 | 297 | 23 | - |
| | 5 | 1358 | 1320 | 49 | - |
| | 6 | 96 | 93 | 32 | - |
| | 7 | 0 | 0 | 35 | - |
| | 8 | 467 | 536 | 49 | - |
| | 9 | 71 | 108 | 38 | - |
| | 10 | 467 | 467 | 42 | + |
| | 11 | 797 | 886 | 53 | + |
| | 12 | 3 | 103 | 36 | + |
| | 13 | 1778 | 1744 | 80 | - |
| ••• 3 K 3 ••• | | | | | |
| | 1 | 882 | 903 | 37 | + |
| E | 2 | 1386 | 1356 | 50 | + |
| | 3 | 25 | 101 | 27 | + |
| | 4 | 39 | 14 | 28 | + |
| | 5 | 375 | 366 | 37 | - |
| | 6 | 451 | 469 | 31 | - |
| | 7 | 1453 | 1521 | 69 | + |
| | 8 | 696 | 696 | 51 | - |
| | 9 | 348 | 301 | 41 | + |
| | 10 | 236 | 172 | 36 | + |
| | 11 | 61 | 74 | 37 | + |
| | 12 | 0 | 13 | 36 | + |
| | 13 | 513 | 523 | 48 | - |
| ••• 3 K -3 ••• | | | | | |
| | 1 | 966 | 975 | 38 | - |
| | 2 | 19 | 99 | 18 | - |
| | 3 | 1078 | 987 | 48 | + |
| | 4 | 90 | 78 | 32 | - |
| | 5 | 1000 | 888 | 49 | + |
| | 6 | 121 | 77 | 26 | - |
| | 7 | 1567 | 1546 | 70 | - |
| | 8 | 487 | 552 | 47 | - |
| | 9 | 1383 | 1388 | 66 | - |
| | 10 | 115 | 75 | 39 | - |
| | 11 | 839 | 889 | 54 | + |
| | 12 | 76 | 16 | 37 | + |
| | 13 | 608 | 639 | 52 | + |
| ••• 3 K 4 ••• | | | | | |
| | 0 | 337 | 301 | 34 | - |
| | 1 | 54 | 49 | 28 | - |
| E | 2 | 2299 | 2160 | 74 | + |
| | 3 | 488 | 567 | 39 | - |
| | 4 | 637 | 587 | 44 | - |
| | 5 | 2117 | 2038 | 82 | + |
| | 6 | 584 | 516 | 48 | + |
| | 7 | 503 | 422 | 46 | + |
| | 8 | 1056 | 1009 | 57 | - |
| | 9 | 1 | 0 | 33 | + |
| | 10 | 73 | 10 | 35 | - |
| | 11 | 303 | 330 | 43 | - |
| | 12 | 182 | 210 | 42 | - |
| | 13 | 554 | 645 | 54 | + |
| ••• 3 K -4 ••• | | | | | |
| | 0 | 47 | 0 | 27 | + |
| | 1 | 58 | 58 | 22 | - |
| E | 2 | 1820 | 1778 | 62 | - |
| | 3 | 738 | 732 | 35 | - |
| | 4 | 510 | 463 | 41 | + |
| E | 5 | 2544 | 2385 | 91 | + |
| | 6 | 730 | 716 | 50 | - |
| | 7 | 269 | 367 | 42 | + |
| | 8 | 71 | 19 | 47 | + |
| | 9 | 427 | 426 | 41 | - |
| | 10 | 6 | 28 | 34 | - |
| | 11 | 1031 | 894 | 57 | - |
| | 12 | 277 | 341 | 44 | + |
| | 13 | 313 | 381 | 49 | + |
| ••• 3 K 5 ••• | | | | | |
| | 1 | 307 | 252 | 41 | - |
| | 2 | 173 | 114 | 38 | + |
| | 3 | 13 | 43 | 38 | + |
| | 4 | 1168 | 1098 | 59 | + |
| | 5 | 192 | 183 | 42 | - |
| | 6 | 54 | 69 | 35 | + |
| | 7 | 468 | 509 | 45 | - |
| | 8 | 255 | 292 | 39 | - |
| | 9 | 293 | 350 | 40 | - |
| | 10 | 191 | 233 | 40 | - |
| | 11 | 199 | 196 | 42 | + |
| | 12 | 576 | 579 | 51 | + |
| ••• 3 K -5 ••• | | | | | |
| | 1 | 35 | 67 | 34 | + |
| | 2 | 175 | 250 | 37 | + |
| | 3 | 211 | 231 | 39 | - |
| | 4 | 810 | 843 | 53 | + |
| | 5 | 211 | 248 | 43 | + |
| | 6 | 448 | 387 | 43 | + |
| | 7 | 91 | 131 | 35 | - |
| | 8 | 148 | 108 | 37 | - |
| | 9 | 119 | 114 | 40 | + |
| | 10 | 129 | 89 | 39 | - |
| | 11 | 1181 | 1134 | 64 | - |
| | 12 | 38 | 71 | 42 | + |
| ••• 3 K 6 ••• | | | | | |
| | 0 | 1 | 27 | 36 | - |
| | 1 | 793 | 849 | 54 | - |
| | 2 | 342 | 315 | 44 | - |
| | 3 | 1162 | 1141 | 60 | - |
| | 4 | 399 | 367 | 41 | + |
| | 5 | 122 | 118 | 37 | - |
| | 6 | 1655 | 1648 | 72 | - |
| | 7 | 23 | 0 | 34 | + |
| | 8 | 6 | 34 | 34 | - |
| | 9 | 16 | 15 | 35 | + |
| | 10 | 0 | 0 | 38 | - |
| ••• 3 K -6 ••• | | | | | |
| | 0 | 152 | 143 | 38 | - |
| | 1 | 9 | 48 | 37 | - |
| | 2 | 43 | 0 | 37 | + |
| | 3 | 71 | 112 | 39 | - |
| | 4 | 2 | 3 | 35 | + |
| | 5 | 95 | 149 | 35 | - |
| | 6 | 1017 | 1002 | 55 | + |
| | 7 | 7 | 0 | 33 | - |
| | 8 | 47 | 0 | 35 | - |
| | 9 | 188 | 189 | 40 | + |
| | 10 | 7 | 41 | 38 | + |
| ••• 3 K 7 ••• | | | | | |
| | 1 | 208 | 219 | 39 | - |
| | 2 | 41 | 24 | 33 | + |
| | 3 | 969 | 998 | 55 | + |
| | 4 | 108 | 138 | 37 | - |
| | 5 | 841 | 819 | 52 | + |
| | 6 | 1786 | 1657 | 72 | - |
| | 7 | 6 | 0 | 37 | - |
| | 8 | 41 | 73 | 38 | + |
| | 9 | 1258 | 1295 | 67 | - |
| ••• 3 K -7 ••• | | | | | |
| | 1 | 695 | 730 | 47 | - |
| | 2 | 18 | 0 | 33 | + |
| | 3 | 150 | 145 | 35 | + |
| | 4 | 21 | 55 | 34 | - |
| | 5 | 523 | 500 | 44 | + |
| | 6 | 344 | 416 | 41 | - |
| | 7 | 1 | 0 | 34 | - |
| | 8 | 9 | 0 | 37 | + |
| | 9 | 6 | 8 | 41 | + |
| ••• 3 K 8 ••• | | | | | |
| | 0 | 361 | 412 | 43 | + |
| | 1 | 144 | 266 | 39 | - |
| | 2 | 468 | 483 | 42 | + |
| | 3 | 980 | 969 | 56 | + |
| | 4 | 1438 | 1359 | 66 | - |
| | 5 | 76 | 104 | 38 | - |
| | 6 | 398 | 312 | 44 | + |
| | 7 | 50 | 106 | 41 | + |
| ••• 3 K -8 ••• | | | | | |
| | 0 | 497 | 530 | 45 | - |
| | 1 | 1160 | 1015 | 59 | - |
| | 2 | 239 | 253 | 38 | - |
| | 3 | 18 | 62 | 36 | - |
| | 4 | 1772 | 1714 | 76 | + |
| | 5 | 46 | 47 | 37 | + |
| | 6 | 64 | 51 | 40 | - |
| | 7 | 519 | 536 | 49 | + |
| ••• 3 K 9 ••• | | | | | |
| | 1 | 717 | 657 | 52 | - |
| | 2 | 1586 | 1692 | 77 | - |
| | 3 | 1 | 20 | 40 | - |
| | 4 | 4 | 0 | 42 | + |
| | 5 | 476 | 450 | 50 | + |
| ••• 3 K -9 ••• | | | | | |
| | 1 | 118 | 92 | 41 | + |
| | 2 | 1527 | 1611 | 77 | - |
| | 3 | 34 | 19 | 41 | - |
| | 4 | 2 | 0 | 42 | - |
| | 5 | 41 | 34 | 43 | - |
| ••• 4 K 0 ••• | | | | | |
| E | 0 | 7669 | 6897 | 207 | + |
| | 1 | 189 | 205 | 23 | + |
| | 2 | 51 | 43 | 20 | - |
| | 3 | 224 | 301 | 25 | + |
| | 4 | 32 | 14 | 23 | - |
| | 5 | 166 | 137 | 41 | + |
| | 6 | 438 | 422 | 35 | - |
| | 7 | 3 | 15 | 26 | - |
| | 8 | 936 | 851 | 43 | + |
| | 9 | 7 | 25 | 23 | - |
| | 10 | 199 | 187 | 28 | - |
| | 11 | 20 | 11 | 25 | + |
| | 12 | 353 | 424 | 35 | - |
| | 13 | 51 | 48 | 29 | + |
| | 14 | 0 | 6 | 43 | - |
| | 15 | 191 | 101 | 45 | + |
| | 16 | 332 | 303 | 47 | + |
| | 17 | 159 | 207 | 42 | + |
| ••• 4 K 1 ••• | | | | | |
| | 1 | 62 | 94 | 22 | + |
| | 2 | 263 | 253 | 26 | - |
| | 3 | 620 | 591 | 33 | - |
| | 4 | 248 | 247 | 39 | + |
| | 5 | 1 | 78 | 37 | - |
| E | 6 | 3172 | 2901 | 109 | + |
| | 7 | 368 | 386 | 45 | - |
| | 8 | 16 | 11 | 34 | + |
| | 9 | 65 | 77 | 33 | + |
| | 10 | 446 | 401 | 44 | - |
| | 11 | 684 | 759 | 53 | - |
| | 12 | 51 | 25 | 39 | - |
| ••• 4 K -1 ••• | | | | | |
| | 1 | 523 | 546 | 29 | - |
| | 2 | 606 | 614 | 32 | + |
| | 3 | 202 | 261 | 26 | + |
| | 4 | 11 | 27 | 32 | - |
| E | 5 | 1941 | 1847 | 78 | + |
| | 6 | 632 | 732 | 54 | - |
| | 7 | 236 | 231 | 42 | + |
| | 8 | 256 | 380 | 42 | - |
| | 9 | 59 | 77 | 40 | - |
| | 10 | 115 | 104 | 38 | + |
| | 11 | 9 | 0 | 36 | + |
| | 12 | 96 | 100 | 40 | - |
| ••• 4 K 2 ••• | | | | | |
| | 0 | 528 | 532 | 32 | - |
| | 1 | 249 | 258 | 26 | - |
| | 2 | 59 | 44 | 28 | - |
| | 3 | 370 | 418 | 30 | - |
| | 4 | 123 | 158 | 36 | - |
| | 5 | 14 | 64 | 38 | + |
| | 6 | 57 | 110 | 46 | - |
| | 7 | 39 | 0 | 36 | + |
| | 8 | 681 | 578 | 48 | - |
| | 9 | 885 | 870 | 55 | - |
| | 10 | 163 | 176 | 38 | + |
| | 11 | 38 | 107 | 38 | + |
| | 12 | 125 | 187 | 43 | + |
| ••• 4 K -2 ••• | | | | | |
| E | 0 | 2081 | 2004 | 69 | - |
| | 1 | 319 | 334 | 26 | - |
| | 2 | 67 | 22 | 23 | - |
| | 3 | 850 | 908 | 40 | - |
| | 4 | 150 | 130 | 39 | + |
| | 5 | 733 | 716 | 52 | + |
| | 6 | 516 | 504 | 47 | + |
| | 7 | 277 | 281 | 44 | + |
| | 8 | 306 | 269 | 40 | - |
| | 9 | 83 | 84 | 35 | - |
| | 10 | 558 | 507 | 46 | + |
| | 11 | 1681 | 1708 | 77 | - |
| | 12 | 4 | 0 | 41 | - |
| ••• 4 K 3 ••• | | | | | |
| | 1 | 50 | 54 | 86 | + |
| | 2 | 568 | 668 | 47 | - |
| | 3 | 477 | 534 | 47 | + |
| | 4 | 1258 | 1216 | 63 | - |
| | 5 | 0 | 0 | 38 | - |
| | 6 | 52 | 0 | 37 | - |
| | 7 | 411 | 333 | 41 | - |
| | 8 | 0 | 0 | 33 | + |
| | 9 | 411 | 398 | 43 | - |
| | 10 | 3 | 0 | 36 | + |
| | 11 | 650 | 641 | 51 | + |
| | 12 | 52 | 88 | 42 | - |
| ••• 4 K -3 ••• | | | | | |
| | 1 | 0 | 14 | 34 | + |
| | 2 | 865 | 911 | 51 | - |
| | 3 | 366 | 429 | 42 | - |
| | 4 | 119 | 252 | 41 | + |
| | 5 | 3 | 23 | 38 | + |
| | 6 | 63 | 27 | 36 | + |
| | 7 | 82 | 38 | 48 | - |
| | 8 | 50 | 32 | 34 | + |
| | 9 | 20 | 70 | 34 | + |
| | 10 | 519 | 489 | 46 | - |
| | 11 | 541 | 517 | 50 | - |
| | 12 | 638 | 750 | 57 | + |
| ••• 4 K 4 ••• | | | | | |
| | 0 | 99 | 181 | 41 | + |
| | 1 | 665 | 756 | 53 | + |
| | 2 | 49 | 81 | 41 | + |
| | 3 | 236 | 325 | 44 | - |
| | 4 | 202 | 233 | 42 | + |
| | 5 | 0 | 0 | 34 | + |
| | 6 | 463 | 419 | 44 | + |
| | 7 | 38 | 93 | 36 | - |
| | 8 | 193 | 201 | 39 | + |
| | 9 | 727 | 743 | 51 | + |
| | 10 | 156 | 168 | 41 | - |
| | 11 | 17 | 0 | 41 | + |
| ••• 4 K -4 ••• | | | | | |
| | 0 | 248 | 311 | 40 | + |
| E | 1 | 2842 | 2662 | 100 | - |
| | 2 | 519 | 519 | 46 | - |
| | 3 | 321 | 374 | 42 | - |
| | 4 | 2 | 51 | 36 | + |
| | 5 | 350 | 350 | 40 | - |
| | 6 | 25 | 71 | 33 | + |
| | 7 | 1520 | 1432 | 67 | + |
| | 8 | 406 | 372 | 43 | + |
| | 9 | 19 | 67 | 36 | + |
| | 10 | 409 | 444 | 46 | - |
| | 11 | 96 | 29 | 43 | + |
| ••• 4 K 5 ••• | | | | | |
| | 1 | 4 | 11 | 36 | - |
| | 2 | 51 | 73 | 36 | - |
| | 3 | 387 | 339 | 46 | - |
| | 4 | 84 | 96 | 38 | - |
| | 5 | 0 | 28 | 33 | + |
| | 6 | 820 | 746 | 51 | + |
| | 7 | 1554 | 1367 | 67 | + |
| | 8 | 32 | 61 | 36 | - |
| | 9 | 488 | 496 | 48 | + |
| | 10 | 374 | 383 | 47 | + |
| ••• 4 K -5 ••• | | | | | |
| | 1 | 993 | 931 | 54 | + |
| | 2 | 6 | 12 | 34 | + |
| | 3 | 271 | 234 | 39 | - |
| | 4 | 0 | 0 | 33 | - |
| | 5 | 578 | 566 | 43 | - |
| | 6 | 80 | 98 | 36 | - |
| | 7 | 104 | 168 | 44 | + |
| | 8 | 165 | 171 | 41 | + |
| | 9 | 645 | 593 | 49 | + |
| | 10 | 256 | 277 | 44 | + |
| ••• 4 K 6 ••• | | | | | |
| | 0 | 603 | 573 | 47 | - |
| | 1 | 403 | 435 | 44 | - |
| | 2 | 148 | 202 | 38 | - |
| | 3 | 450 | 402 | 44 | - |
| | 4 | 5 | 0 | 35 | - |
| | 5 | 360 | 288 | 42 | + |
| | 6 | 6 | 0 | 35 | + |
| | 7 | 536 | 622 | 47 | + |
| | 8 | 0 | 0 | 40 | + |
| | 9 | 402 | 565 | 51 | - |
| ••• 4 K -6 ••• | | | | | |
| | 0 | 184 | 228 | 36 | - |
| | 1 | 300 | 358 | 40 | + |
| | 2 | 138 | 79 | 36 | + |
| | 3 | 7 | 3 | 33 | + |
| | 4 | 861 | 931 | 53 | + |
| | 5 | 45 | 33 | 35 | + |
| | 6 | 21 | 15 | 35 | - |
| | 7 | 693 | 704 | 51 | - |
| | 8 | 1094 | 1160 | 64 | - |
| | 9 | 165 | 200 | 46 | + |
| ••• 4 K 7 ••• | | | | | |
| | 1 | 696 | 744 | 50 | - |
| | 2 | 17 | 30 | 37 | + |
| | 3 | 226 | 195 | 42 | + |
| | 4 | 286 | 282 | 43 | + |
| | 5 | 77 | 117 | 36 | + |
| | 6 | 57 | 128 | 40 | + |
| | 7 | 73 | 121 | 41 | - |
| ••• 4 K -7 ••• | | | | | |
| | 1 | 4 | 0 | 36 | - |
| E | 2 | 2786 | 2673 | 102 | - |
| | 3 | 37 | 81 | 37 | + |
| | 4 | 3 | 24 | 36 | - |
| | 5 | 70 | 21 | 38 | - |
| | 6 | 610 | 540 | 50 | + |
| | 7 | 20 | 103 | 41 | - |
| ••• 4 K 8 ••• | | | | | |
| | 0 | 212 | 243 | 45 | - |
| | 1 | 233 | 264 | 45 | + |
| | 2 | 218 | 213 | 44 | + |
| | 3 | 321 | 274 | 45 | + |
| | 4 | 1 | 58 | 41 | - |
| | 5 | 55 | 48 | 42 | - |
| ••• 4 K -8 ••• | | | | | |
| | 0 | 4 | 0 | 38 | + |
| | 1 | 327 | 365 | 45 | - |
| | 2 | 45 | 92 | 39 | + |
| | 3 | 678 | 689 | 52 | + |
| | 4 | 1301 | 1278 | 67 | - |
| | 5 | 28 | 0 | 43 | + |
| ••• 5 K 0 ••• | | | | | |
| | 0 | 85 | 92 | 30 | + |
| E | 1 | 2307 | 2231 | 80 | + |
| | 2 | 105 | 91 | 35 | + |
| | 3 | 0 | 45 | 26 | + |
| | 4 | 201 | 142 | 31 | + |
| | 5 | 0 | 0 | 24 | - |
| | 6 | 9 | 0 | 24 | + |
| E | 7 | 2796 | 2540 | 100 | - |
| | 8 | 70 | 53 | 26 | - |
| | 9 | 1888 | 1775 | 68 | + |
| | 10 | 76 | 38 | 28 | + |
| | 11 | 60 | 48 | 30 | - |
| | 12 | 23 | 0 | 30 | + |
| | 13 | 27 | 27 | 42 | + |
| | 14 | 13 | 0 | 42 | + |
| | 15 | 1679 | 1679 | 80 | - |
| | 16 | 128 | 132 | 42 | - |
| ••• 5 K 1 ••• | | | | | |
| | 1 | 531 | 497 | 50 | + |
| | 2 | 526 | 738 | 53 | - |
| | 3 | 792 | 775 | 57 | - |
| | 4 | 232 | 233 | 45 | - |
| | 5 | 118 | 121 | 49 | - |
| | 6 | 1 | 0 | 35 | - |
| | 7 | 817 | 801 | 53 | + |
| | 8 | 1624 | 1636 | 75 | + |
| | 9 | 1638 | 1592 | 74 | + |
| | 10 | 163 | 216 | 45 | + |
| | 11 | 158 | 204 | 44 | - |
| ••• 5 K -1 ••• | | | | | |
| | 1 | 17 | 19 | 39 | - |
| E | 2 | 3010 | 2878 | 108 | - |
| | 3 | 274 | 321 | 45 | + |
| | 4 | 488 | 517 | 47 | - |
| | 5 | 68 | 65 | 43 | + |
| | 6 | 137 | 141 | 37 | + |
| | 7 | 767 | 765 | 50 | - |
| | 8 | 333 | 388 | 45 | + |
| | 9 | 446 | 432 | 46 | - |
| | 10 | 414 | 537 | 50 | - |
| | 11 | 123 | 113 | 43 | + |
| ••• 5 K 2 ••• | | | | | |
| | 0 | 14 | 0 | 38 | + |
| | 1 | 1425 | 1512 | 73 | - |
| E | 2 | 2014 | 1881 | 82 | - |
| | 3 | 281 | 261 | 44 | + |
| | 4 | 557 | 676 | 51 | + |
| | 5 | 446 | 401 | 46 | - |
| | 6 | 40 | 122 | 35 | + |
| | 7 | 1000 | 1035 | 58 | + |
| | 8 | 450 | 461 | 46 | + |
| | 9 | 130 | 131 | 40 | - |
| | 10 | 1651 | 1655 | 77 | - |
| ••• 5 K -2 ••• | | | | | |
| | 0 | 503 | 582 | 46 | + |
| E | 1 | 2062 | 2102 | 84 | - |
| | 2 | 253 | 302 | 43 | + |
| | 3 | 809 | 695 | 50 | + |
| E | 4 | 4435 | 4034 | 140 | - |
| | 5 | 305 | 331 | 42 | - |
| | 6 | 102 | 0 | 37 | - |
| | 7 | 825 | 938 | 56 | + |
| | 8 | 162 | 138 | 41 | + |
| | 9 | 1191 | 1201 | 64 | - |
| | 10 | 715 | 652 | 57 | + |
| ••• 5 K 3 ••• | | | | | |
| | 1 | 30 | 17 | 35 | + |
| | 2 | 0 | 9 | 35 | - |
| | 3 | 647 | 580 | 50 | - |
| | 4 | 397 | 432 | 44 | + |
| | 5 | 250 | 200 | 43 | - |
| | 6 | 340 | 315 | 42 | + |
| | 7 | 55 | 79 | 35 | - |
| | 8 | 175 | 218 | 40 | - |
| | 9 | 354 | 394 | 47 | + |
| | 10 | 925 | 918 | 64 | - |
| ••• 5 K -3 ••• | | | | | |
| | 1 | 15 | 0 | 36 | + |
| | 2 | 425 | 356 | 40 | + |
| | 3 | 37 | 75 | 38 | - |
| | 4 | 781 | 774 | 50 | + |
| | 5 | 68 | 50 | 36 | - |
| | 6 | 4 | 61 | 34 | - |
| | 7 | 438 | 423 | 43 | + |
| | 8 | 65 | 74 | 38 | + |
| | 9 | 203 | 221 | 44 | - |
| | 10 | 75 | 60 | 47 | - |
| ••• 5 K 4 ••• | | | | | |
| E | 0 | 2753 | 2552 | 101 | - |
| | 1 | 18 | 37 | 33 | + |
| | 2 | 99 | 106 | 37 | + |
| | 3 | 476 | 417 | 47 | - |
| | 4 | 168 | 43 | 42 | + |
| | 5 | 216 | 171 | 40 | + |
| | 6 | 1009 | 1062 | 61 | + |
| | 7 | 11 | 6 | 37 | + |
| | 8 | 1230 | 1178 | 66 | - |
| | 9 | 46 | 29 | 42 | - |
| ••• 5 K -4 ••• | | | | | |
| E | 0 | 2638 | 2394 | 92 | + |
| | 1 | 236 | 183 | 35 | + |
| | 2 | 32 | 30 | 36 | - |
| | 3 | 45 | 24 | 35 | - |
| | 4 | 3 | 39 | 35 | + |
| | 5 | 0 | 0 | 35 | + |
| | 6 | 633 | 693 | 50 | - |
| | 7 | 70 | 45 | 39 | - |
| | 8 | 1103 | 1086 | 61 | + |
| | 9 | 706 | 712 | 56 | + |
| ••• 5 K 5 ••• | | | | | |
| | 1 | 585 | 583 | 50 | + |
| E | 2 | 2432 | 2395 | 95 | + |
| | 3 | 16 | 51 | 35 | + |
| | 4 | 6 | 0 | 35 | + |
| | 5 | 63 | 110 | 37 | + |
| | 6 | 1345 | 1322 | 69 | - |
| | 7 | 3 | 3 | 41 | - |
| | 8 | 433 | 494 | 55 | - |
| ••• 5 K -5 ••• | | | | | |
| | 1 | 526 | 532 | 44 | - |
| | 2 | 437 | 505 | 43 | + |
| | 3 | 177 | 83 | 44 | + |
| | 4 | 8 | 0 | 35 | + |
| | 5 | 221 | 229 | 40 | + |
| | 6 | 585 | 562 | 48 | - |
| | 7 | 33 | 0 | 40 | + |
| | 8 | 157 | 192 | 44 | - |
| ••• 5 K 6 ••• | | | | | |
| E | 0 | 8063 | 7339 | 241 | + |
| | 1 | 2 | 34 | 37 | + |
| | 2 | 73 | 35 | 38 | - |
| | 3 | 338 | 426 | 46 | - |
| | 4 | 291 | 329 | 46 | - |
| | 5 | 385 | 384 | 48 | + |
| | 6 | 1685 | 1716 | 82 | - |
| ••• 5 K -6 ••• | | | | | |
| E | 0 | 2463 | 2624 | 102 | - |
| | 1 | 552 | 607 | 48 | - |
| | 2 | 419 | 431 | 45 | + |
| | 3 | 917 | 929 | 56 | - |
| | 4 | 447 | 533 | 46 | - |
| | 5 | 536 | 505 | 48 | + |
| | 6 | 1156 | 1126 | 63 | + |
| ••• 5 K 7 ••• | | | | | |
| | 1 | 338 | 363 | 50 | - |
| | 2 | 300 | 247 | 50 | - |
| | 3 | 74 | 198 | 42 | - |
| | 4 | 69 | 155 | 42 | - |
| ••• 5 K -7 ••• | | | | | |
| | 1 | 980 | 992 | 60 | + |
| | 2 | 32 | 68 | 41 | - |
| | 3 | 372 | 377 | 47 | - |
| | 4 | 78 | 85 | 43 | - |
| ••• 6 K 0 ••• | | | | | |
| | 0 | 1526 | 1615 | 61 | - |
| | 1 | 47 | 54 | 34 | - |
| | 2 | 1 | 12 | 24 | + |
| | 3 | 52 | 9 | 25 | + |
| | 4 | 18 | 0 | 25 | + |
| | 5 | 151 | 174 | 42 | + |
| | 6 | 111 | 135 | 47 | + |
| | 7 | 0 | 0 | 27 | + |
| | 8 | 5 | 17 | 29 | - |
| | 9 | 521 | 570 | 42 | - |
| | 10 | 77 | 92 | 49 | + |
| | 11 | 353 | 260 | 53 | - |
| | 12 | 92 | 46 | 42 | + |
| | 13 | 16 | 6 | 41 | + |
| | 14 | 79 | 119 | 41 | - |
| ••• 6 K 1 ••• | | | | | |
| | 1 | 12 | 51 | 34 | + |
| | 2 | 317 | 287 | 46 | + |
| | 3 | 325 | 283 | 47 | + |
| | 4 | 10 | 2 | 35 | + |
| | 5 | 77 | 109 | 37 | + |
| | 6 | 830 | 833 | 58 | - |
| | 7 | 213 | 279 | 46 | - |
| | 8 | 9 | 0 | 43 | - |
| ••• 6 K -1 ••• | | | | | |
| | 1 | 12 | 0 | 34 | - |
| | 2 | 13 | 73 | 35 | - |
| | 3 | 52 | 65 | 56 | - |
| | 4 | 43 | 43 | 37 | + |
| | 5 | 452 | 499 | 48 | - |
| | 6 | 372 | 329 | 50 | + |
| | 7 | 690 | 740 | 58 | + |
| | 8 | 3 | 0 | 44 | - |
| ••• 6 K 2 ••• | | | | | |
| | 0 | 1301 | 1263 | 67 | + |
| | 1 | 162 | 224 | 42 | + |
| | 2 | 257 | 239 | 43 | + |
| | 3 | 670 | 699 | 54 | + |
| | 4 | 47 | 0 | 36 | - |
| | 5 | 66 | 126 | 39 | - |
| | 6 | 170 | 169 | 45 | - |
| | 7 | 49 | 38 | 41 | - |
| | 8 | 1 | 61 | 43 | + |
| ••• 6 K -2 ••• | | | | | |
| | 0 | 44 | 98 | 37 | + |
| | 1 | 21 | 0 | 36 | + |
| | 2 | 12 | 22 | 36 | + |
| | 3 | 45 | 96 | 37 | - |
| | 4 | 516 | 507 | 48 | - |
| | 5 | 271 | 292 | 44 | - |
| | 6 | 21 | 62 | 38 | + |
| | 7 | 1 | 0 | 41 | - |
| | 9 | 491 | 526 | 56 | + |
| ••• 6 K 3 ••• | | | | | |
| | 1 | 241 | 273 | 48 | + |
| | 2 | 3 | 0 | 36 | + |
| | 3 | 356 | 393 | 48 | - |
| | 4 | 209 | 181 | 47 | + |
| | 5 | 1080 | 1060 | 66 | - |
| | 6 | 40 | 75 | 42 | - |
| | 7 | 110 | 110 | 44 | + |
| ••• 6 K -3 ••• | | | | | |
| | 1 | 7 | 66 | 35 | - |
| | 2 | 375 | 349 | 42 | + |
| | 3 | 10 | 46 | 36 | + |
| | 4 | 89 | 61 | 38 | - |
| | 5 | 1 | 0 | 41 | - |
| | 6 | 819 | 838 | 57 | - |
| | 7 | 65 | 61 | 42 | + |
| ••• 6 K 4 ••• | | | | | |
| | 0 | 1 | 0 | 37 | + |
| | 1 | 1360 | 1379 | 71 | - |
| | 2 | 12 | 8 | 38 | - |
| | 3 | 635 | 684 | 57 | - |
| | 4 | 346 | 314 | 50 | - |
| | 5 | 105 | 88 | 43 | - |
| | 6 | 104 | 174 | 44 | - |
| ••• 6 K -4 ••• | | | | | |
| | 0 | 234 | 272 | 41 | - |
| | 1 | 27 | 91 | 39 | + |
| | 2 | 173 | 168 | 41 | + |
| | 3 | 41 | 97 | 39 | - |
| | 4 | 279 | 245 | 44 | + |
| | 5 | 4 | 54 | 40 | - |
| | 6 | 67 | 168 | 44 | - |
| ••• 6 K 5 ••• | | | | | |
| | 1 | 162 | 167 | 42 | - |
| | 2 | 121 | 139 | 43 | - |
| | 3 | 258 | 279 | 50 | + |
| | 4 | 160 | 222 | 48 | - |
| ••• 6 K -5 ••• | | | | | |
| | 1 | 320 | 315 | 45 | + |
| | 2 | 224 | 216 | 44 | - |
| | 3 | 8 | 0 | 43 | - |
| | 4 | 211 | 197 | 46 | - |
| ••• 7 K 0 ••• | | | | | |
| | 0 | 55 | 23 | 30 | + |
| | 1 | 1054 | 995 | 66 | + |
| | 2 | 6 | 2 | 31 | + |
| | 3 | 523 | 555 | 44 | - |
| | 4 | 601 | 659 | 46 | - |
| | 5 | 260 | 242 | 38 | + |
| | 6 | 18 | 45 | 32 | - |
| | 7 | 1251 | 1310 | 75 | - |
| | 8 | 34 | 43 | 44 | + |
| | 9 | 167 | 107 | 45 | + |
| | 10 | 0 | 117 | 42 | + |
| | 11 | 47 | 36 | 42 | + |
| ••• 7 K 1 ••• | | | | | |
| | 1 | 94 | 128 | 46 | + |
| | 2 | 1402 | 1237 | 77 | - |
| | 3 | 33 | 34 | 45 | + |
| ••• 7 K -1 ••• | | | | | |
| | 1 | 15 | 21 | 44 | + |
| | 2 | 53 | 161 | 45 | + |
| | 3 | 4 | 0 | 44 | - |
| ••• 7 K 2 ••• | | | | | |
| | 0 | 1495 | 1401 | 80 | - |
| | 1 | 75 | 74 | 45 | - |
| | 2 | 97 | 121 | 46 | - |
| ••• 7 K -2 ••• | | | | | |
| | 0 | 156 | 194 | 47 | + |
| | 1 | 130 | 84 | 46 | + |
| | 2 | 655 | 576 | 57 | + |
| ••• 8 K 0 ••• | | | | | |
| | 0 | 298 | 243 | 50 | - |
| | 1 | 155 | 184 | 48 | + |
| | 2 | 13 | 0 | 46 | + |
| | 3 | 77 | 102 | 47 | + |
| | 4 | 4 | 0 | 47 | - |
| | 5 | 56 | 64 | 48 | - |
| | 6 | 434 | 331 | 61 | + |
| | 7 | 26 | 0 | 46 | - |
| | 8 | 142 | 66 | 48 | - |

results should not be taken as final, we believe that collection of a large body of scattering-factor data from least-squares refinements of many different crystal structures may lead, on careful evaluation, to a more reliable set of nuclear scattering factors of the elements.

The atomic coordinates and thermal parameters from refinement *E* should in principle be the most reliable. In any case, as already discussed, these parameters differ only slightly from those of the other sets. We therefore report them and their standard errors in

Table 3. When converted to Å, the standard errors of the coordinates amount to about 0·001 Å for the niobium atom and 0·002 to 0·003 Å for the fluorine and potassium atoms.

Table 4 presents the values $F_o^2$, $\sigma_{F^2}$, and $F_c^2$, with the sign of $F_c$, for all of the observations.

A composite three-dimensional Fourier map is shown in Fig. 2. The $F_o$ coefficients used in the synthesis were obtained from the set of $F_o^2$ data corrected for extinction that was used in refinement *F*. The signs were from the $F_c$ values computed with the use of the parameters in Table 3. A final three-dimensional difference map showed only random background ripples. The greatest excursion of density on each side of zero was in magnitude only about 2% of the peak density of the niobium atom.

## Structure and thermal motion

### *Structure*

An understanding of the sense in which our results confirm those of Hoard may be reached quickly by viewing the shifts of atomic positions between the two structures (Table 5). The shifts are small in the sense that the basic geometric description of the crystal structure remains unchanged, including the description of the distorted $NbF_7^{2-}$ anion. On the other hand, the shifts are all large relative to the standard errors. The direction of the shifts of the various atoms vary widely. The two largest shifts, those of F(3) and F(6), are at right angles.

Table 5. *Shifts of atomic positions in the refinement of* $K_2NbF_7$

| Atom | Shift | Atom | Shift |
|---|---|---|---|
| Nb | 0·036 Å | F(3) | 0·106 Å |
| K(1) | 0·046 | F(4) | 0·030 |
| K(2) | 0·066 | F(5) | 0·074 |
| F(1) | 0·026 | F(6) | 0·154 |
| F(2) | 0·060 | F(7) | 0·088 |

General features of the crystal structure are shown in the stereoscopic view of Fig. 3. This view, showing a ball-and-stick model for the complex ion $NbF_7^{2-}$, complements the packing drawing shown by Hoard in his Fig. 3(*b*).[*]

Details of the structure of the complex ion $NbF_7^{2-}$ are shown in Fig. 4, where Nb–F bond lengths and F–F neighbor distances are given, and in Table 6, where F–Nb–F angles are given.[†] The first value in Fig. 4 for each Nb–F bond length is the value of the internuclear separation computed directly from the coordinates of Table 3. The second value, in parentheses, has been corrected for the effects of thermal motion by the method of Busing & Levy (1964), assuming the so-called 'riding' model (see the discussion of thermal motion below). The range of Nb–F bond lengths after correction is 1·940–1·978 Å, only half as wide as the range 1·911–1·971 Å before correction.

The widely varying intraionic F–F neighbor distances (2·357–2·912 Å) and F–Nb–F valence angles

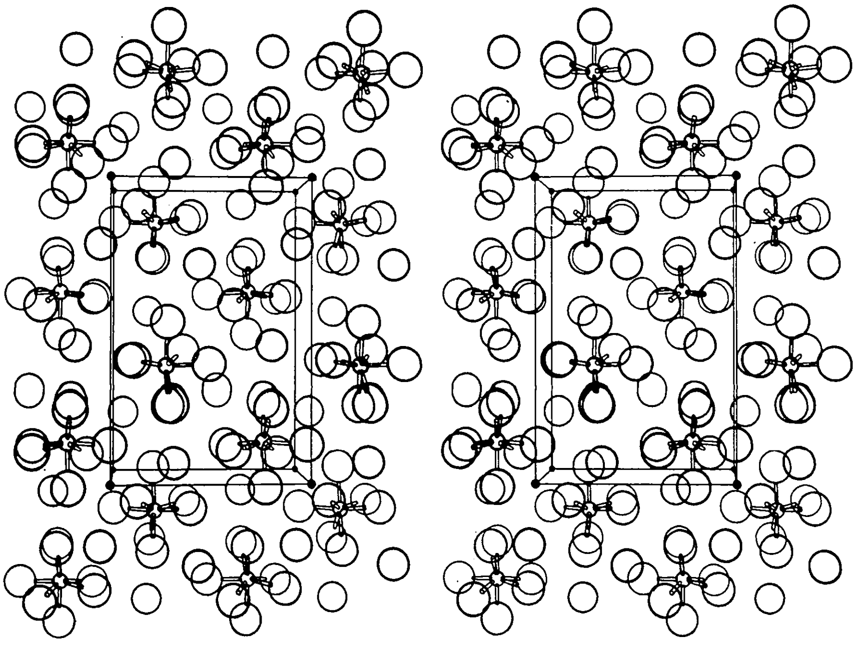

Fig. 3. Stereoscopic view of the $K_2NbF_7$ crystal structure. The direction of view is that of the vector $-\mathbf{a}$. The axis $\mathbf{b}$ is vertical from top to bottom and the axis $\mathbf{c}$ is horizontal from left to right in the view. The reference asymmetric unit is the one within the cell outline at the upper left.

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[*] In comparing our Fig. 3 with Hoard's Fig. 3(*b*), one should note that the latter drawing is based on a left-handed coordinate system.

[†] The program of Busing, Martin & Levy (1964) was used in calculating distances and angles and in analyzing the thermal parameters. The least-squares covariance matrix was used in calculating standard errors.

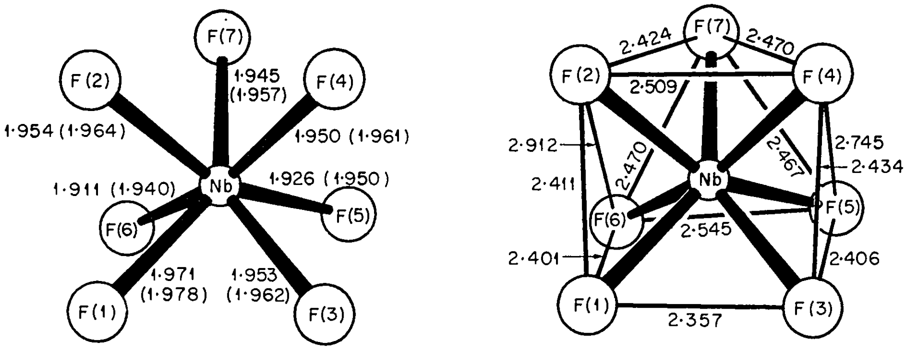

Fig. 4. Interatomic distances (Å) in the $NbF_7^{2-}$ ion. The Nb–F distances in parentheses have been corrected for thermal motion ('riding' model). The computed standard errors are 0·002 Å to 0·003 Å for the uncorrected Nb–F bond lengths and 0·003 Å to 0·004 Å for the F–F distances. The corrected bond lengths are thought to be accurate within 0·01 Å.

(73·86°–97·74°) describe the irregular shape of the ion. The irregularities are, presumably, the result of electrostatic interactions between the fluorine atoms in the anion and the potassium ions which surround it. Fig. 5 shows the arrangement of the potassium ions about the anion, with all of the K–F near-neighbor distances given. Atoms F(1), F(2), F(3), and F(4) are each in contact with three potassium ions in an approximately trigonal arrangement about the axis through niobium and fluorine. Atoms F(5), F(6), and F(7) are each in close contact with only two potassium ions, although F(5) and F(6) each have an additional potassium neighbor at a distance distinctly greater than the average K–F distance. The shortest three of the seven Nb–F bonds are those involving these fluorine atoms having only two potassium near neighbors.

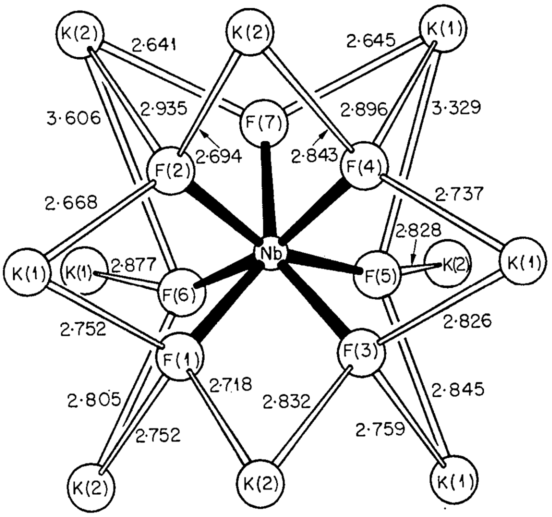

Fig. 5. The $NbF_7^{2-}$ ion and surrounding $K^+$ ions in the $K_2NbF_7$ structure (parallel projection). The computed standard errors of the K–F distances are 0·003 Å to 0·004 Å.

Table 6. *Angles F(i)–Nb–F(j) in the* $NbF_7^{2-}$ ion

The standard errors of these angles are all about 0·1°

| *i* | *j* | Angle |
|---|---|---|
| 7 | 2 | 76·9° |
| 7 | 4 | 78·7 |
| 7 | 5 | 79·2 |
| 7 | 6 | 79·7 |
| 2 | 6 | 97·7 |
| 4 | 5 | 90·2 |
| 2 | 4 | 80·0 |
| 5 | 6 | 83·1 |
| 1 | 2 | 75·8 |
| 1 | 6 | 76·4 |
| 3 | 4 | 77·2 |
| 3 | 5 | 76·7 |
| 1 | 3 | 73·9 |

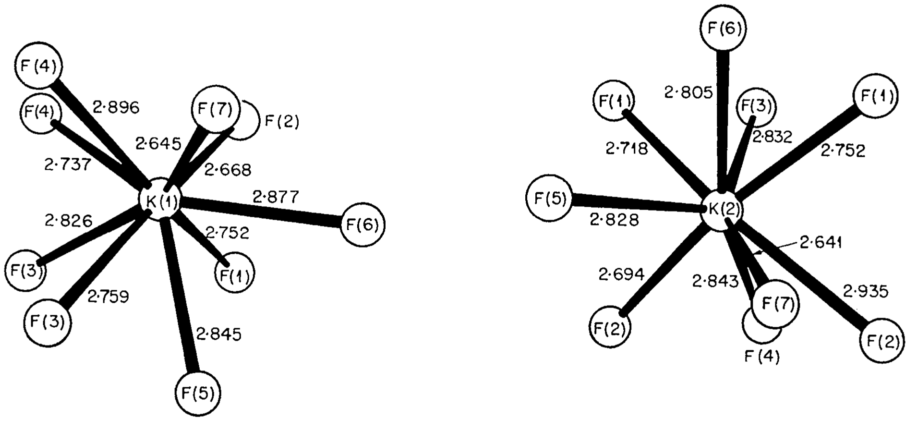

Fig. 6. Perspective views showing the coordination about K(1) and K(2) in $K_2NbF_7$.

The ninefold coordination about each of the two potassium ions (Fig. 6) may be conceived as derived by considerable distortion from an arrangement in which eight fluorine atoms are at the corners of a cube and a ninth atom projects from one cube face. The range of K–F distances is much the same for the two potassium ions.

The shortest interionic F–F distances are: 3·07 Å between F(2) and F(4) in glide-related ions; 3·08 Å between F(7) and F(7) centrosymmetrically related (compare with Hoard's distance 2·91 Å); and 3·18 Å between F(6) and F(7) in centrosymmetrically related ions). There are many F–F distances near 3·30 Å. The range of F–F distances is quite normal.

*Thermal motion*

Principal-axis transformations have been performed on the thermal-parameter ellipsoids of the ten atoms of the asymmetric unit. The root-mean-square axial displacements and the axial orientations are given in Table 7. The thermal motion of the $NbF_7^{2-}$ ion and the 10 $K^+$ ions which surround it can be visualized readily from the stereoscopic drawing of Fig. 7, in which the direction of view is almost the same as that in Figs. 4 and 5. In the drawing the thermal motion of each atom is represented by a thermal-motion probability ellipsoid (Johnson, 1965) centered at the mean position of the atom. The lengths of the principal axes represent the r.m.s. vibrational displacements in the principal-axis directions on a scale 2·25 times that of the internuclear separations. The ellipsoids are, therefore, the 83% probability ellipsoids.

Originally we had supposed that the thermal parameters of the atoms of the $NbF_7^{2-}$ ion would likely be interpretable readily in terms of rigid-body motion of the ion. One can see, however, merely from inspection of Fig. 7 that the large r.m.s. displacements of F(5) and F(6) are inconsistent with the rigid-body model. Detailed analysis (Cruickshank, 1956) does show that the thermal parameters of the other six atoms describe fairly well the motion of a rigid structural unit. The analysis is of doubtful utility, however, because its ultimate purpose, the correction of bond lengths, can be achieved in a more direct manner in this particular case through use of the 'riding' model (Busing & Levy, 1964).

That the riding model is appropriate for calculation of corrections to the Nb–F bond lengths is a direct conclusion from consideration of data in Table 7. The motion of the niobium atom is almost exactly isotropic. For each fluorine atom F(*i*) the principal axis along which the r.m.s. displacement is least is nearly parallel (within 8°) to the corresponding vector Nb → F(*i*). The r.m.s. displacement along this axis is in each case very

Table 7. *Description of atomic thermal-parameter ellipsoids: orientations of the principal axes and root-mean-square axial displacements*

The orientation of each principal axis is specified with respect to the crystal axial vectors **a**, **b**, **c** and, for each fluorine atom F(*i*), with respect to the vector Nb → F(*i*). Standard errors appear in parentheses.

| | | | Angle (°) to reference axis | | | |
|---|---|---|---|---|---|---|
| Atom | Principal axis | R.m.s. displacement | **a** | **b** | **c** | Nb → F(*i*) |
| Nb | 1 | 0·110(3) Å | 77(15) | 141(13) | 54(9) | — |
| | 2 | 0·116(3) | 37(15) | 61(17) | 70(15) | — |
| | 3 | 0·123(3) | 56(14) | 113(10) | 137(9) | — |
| K(1) | 1 | 0·134(4) | 81(5) | 161(6) | 107(7) | — |
| | 2 | 0·162(4) | 97(17) | 108(7) | 19(10) | — |
| | 3 | 0·173(4) | 11(11) | 84(7) | 81(16) | — |
| K(2) | 1 | 0·140(5) | 88(17) | 110(3) | 160(4) | — |
| | 2 | 0·153(5) | 178(15) | 90(7) | 92(16) | — |
| | 3 | 0·191(4) | 91(5) | 160(3) | 70(3) | — |
| F(1) | 1 | 0·125(3) | 40(3) | 130(3) | 85(3) | 172(3) |
| | 2 | 0·165(3) | 53(15) | 41(11) | 75(52) | 83(4) |
| | 3 | 0·168(3) | 77(33) | 82(40) | 165(50) | 87(7) |
| F(2) | 1 | 0·126(3) | 131(6) | 139(6) | 90(3) | 172(6) |
| | 2 | 0·146(3) | 135(6) | 52(6) | 111(2) | 82(6) |
| | 3 | 0·214(3) | 75(2) | 104(2) | 159(2) | 90(1) |
| F(3) | 1 | 0·125(3) | 44(2) | 76(3) | 130(2) | 172(2) |
| | 2 | 0·172(3) | 113(16) | 24(20) | 95(19) | 86(4) |
| | 3 | 0·177(3) | 125(12) | 108(24) | 139(4) | 96(3) |
| F(4) | 1 | 0·120(4) | 123(4) | 81(2) | 145(3) | 178(3) |
| | 2 | 0·155(3) | 145(3) | 108(2) | 61(4) | 91(4) |
| | 3 | 0·217(3) | 80(2) | 160(2) | 107(1) | 92(1) |
| F(5) | 1 | 0·122(3) | 102(2) | 166(1) | 96(2) | 5(1) |
| | 2 | 0·175(3) | 138(1) | 85(3) | 49(1) | 89(3) |
| | 3 | 0·296(3) | 129(1) | 77(1) | 138(1) | 95(1) |
| F(6) | 1 | 0·132(3) | 104(3) | 97(3) | 165(1) | 5(3) |
| | 2 | 0·174(3) | 135(1) | 46(1) | 85(4) | 86(4) |
| | 3 | 0·328(3) | 132(1) | 135(1) | 75(1) | 92(1) |
| F(7) | 1 | 0·132(3) | 30(2) | 106(3) | 115(2) | 4(3) |
| | 2 | 0·181(3) | 90(4) | 32(5) | 122(5) | 94(3) |
| | 3 | 0·201(3) | 120(2) | 117(6) | 137(5) | 89(2) |

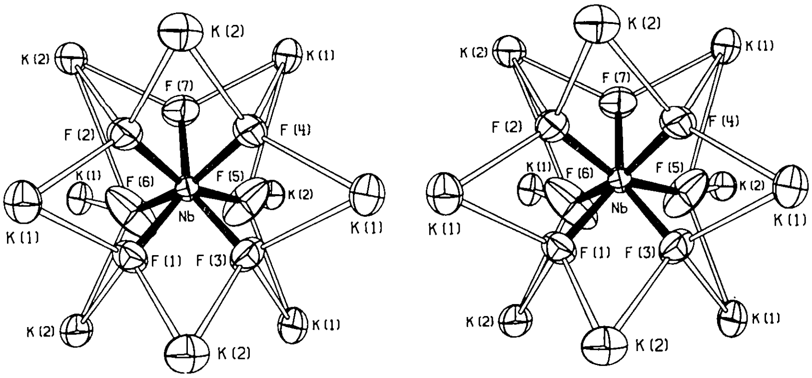

Fig. 7. Stereoscopic view showing thermal-motion probability ellipsoids of the $NbF_7^{2-}$ ion and the surrounding $K^+$ ions.

close in magnitude to the isotropic displacement of the niobium atom. The two larger displacements of each atom F($i$) are nearly perpendicular to the Nb → F($i$) vector. Evidently, the motion of each fluorine atom is to a close approximation a librational motion about the central niobium atom compounded with the translatory vibrational motion of the niobium atom. Thus, the $NbF_7^{2-}$ ion is an almost ideal case for use of the riding model in computing corrections to bond lengths.

The exceptionally large vibrational displacements of F(5) and F(6) seem plausible in view of the packing about these atoms. As already noted, the arrangement of potassium ions about F(5) and F(6) is different from that characteristic of most of the other fluorine atoms. The large displacement, for F(5) and for F(6), is nearly perpendicular to the plane of the fluorine atom and its two close potassium neighbors. The relation of two F(7) atoms across a symmetry center at the unusually short distance 3·08 Å is just such as to inhibit the similar large displacement that might be expected for atom F(7).

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