the total range of these electrons was equal to $0.40+0.37\pm0.03=0.77\pm0.03$ mm of Al. Their maximum energy computed from the energy-range graph should be 0.320 ± 0.005 Mev. By substituting this value into formula (1) it follows immediately that the initial energy of the gamma-rays accompanying the radioactive decay of Be^7 is 0.485 ± 0.005 Mev.

CONCLUSIONS

It should be mentioned that the presence of 0.28 Mev gamma-rays of N^{13}, reported by several authors,[16] could not affect our calculations based upon the maximum range of secondary electrons from radio-nitrogen gamma-radiation. On the other hand it is apparent from the experimental absorption curve for these electrons that there is a very little if any amount of incident gamma-rays with energy above 0.510 Mev.

[16] J. R. Richardson, Phys. Rev. **55**, 609 (1939); E. M. Lyman, Phys. Rev. **55**, 1123 (1939).

Owing to the large value of the slope of the energy-range curve for soft beta-rays the accuracy of our range determination permits us to evaluate the corresponding energies and therefore the energies of the incident gamma-rays within 1 percent. The reasonably close agreement between our experimental result and the mean value of the low excited level of Li^7 deduced from other reactions provides considerable support for the original suggestion to associate the Be^7 gamma-rays with the energy separation of the two states of Li^7 built up by electron capture from the parent body.

ACKNOWLEDGMENTS

Density of Sodium Chloride

The Atomic Weight of Fluorine by Combination of Crystal Density and X-Ray Data

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The densities of carefully purified crystals of sodium chloride have been determined by the method of "crystal flotation" in pure ethylene dibromide. The results yield

$$d_{27.634°C} = 2.16165 \pm 0.00002 \text{ g/ml},$$

which reduces to

$$d_{20°} = 2.16366 \pm 0.00003 \text{ g/ml}.$$

It is found that six successive precipitations of NaCl in the manner employed by Richards and Wells in their determination of the atomic weights of sodium and of chlorine are required to effect purification to constant density ($\pm$ about 4×10^{-6} g/ml), and also that exposure to air produces surface contamination sufficient to cause erratic changes in apparent density that may amount to as much as 5×10^{-4} g/ml within a few minutes. Combination of our value for d_{NaCl} with that of C. A. Hutchison and H. L. Johnston for d_{LiF} and with Straumanis, Ievins, and Karlsons' value for the lattice constant of LiF relative to the Siegbahn value for NaCl yields 0.443640 ± 0.000025

for the ratio of the molecular weights of LiF and NaCl, respectively. With the adoption of 22.997 (International Atomic Weight Committee) for the atomic weight of sodium, this ratio yields 18.994 ± 0.001 for the atomic weight of F. With the adoption of 22.994 (Birge) for sodium, the F atomic weight comes out 18.992. Either of these figures is in reasonable agreement with the value 22.995 ± 0.002, based on densities and lattice constants of fluorite and calcite, and with the gas density determinations for compounds of F, but are somewhat lower than the mass spectrograph value of 18.999 ± 0.001 for F^{19}. It appears that the determination of relative molecular weights by combination of x-ray and density data are as reliable, in favorable cases, as by other standard atomic weight methods. In calcite, fluorite, and rock salt crystals, used to obtain the data underlying these computations, there is no evidence of any appreciable influence of "crystal mosaic" patterns which Zwicky thought might influence crystal densities by as much as 1 percent.

THE most accurate determinations of the density of sodium chloride, reported in the literature, appear to be those of Defoe and

Compton[1] and of Tu,[2] made for the purpose of

[1] O. K. Defoe and A. H. Compton, Phys. Rev. **25**, 618 (1925).
[2] Y. Tu, Phys. Rev. **40**, 662 (1932).

determining standards of x-ray wave-length. Defoe and Compton obtained the value

$$d_{20°} = 2.1632 \pm 0.0004 \text{ g/cm}^3,$$

while Tu obtained the value 2.16418 ± 0.00014 at 18°C. The latter reduces to

$$d_{20°} = 2.16365 \pm 0.00014 \text{ g/cm}^3,$$

when correction is made for the coefficient of thermal expansion of NaCl.[3] Both Defoe and Compton and Tu used natural crystals of rocksalt. Neither made a chemical analysis of the mineral. Defoe and Compton observed differences between different rocksalt crystals that exceeded the error of their density measurements. These differences might be accounted for by traces of chemical impurity or, as we find, by density errors that result from exposure to air.

A recent paper from this laboratory[4] dealt with the accurate determination of the density of chemically pure lithium fluoride, by the method of crystal flotation. We have completed a similar investigation with very carefully purified crystals of sodium chloride. Reduced to 20°C, we obtain the value

$$d_{20°} = 2.16366 \pm 0.00003 \text{ g/ml}$$

$$= 2.16360 \pm 0.00003 \text{ g/cm}^3$$

in good agreement with the value reported by Tu,[2] but with reduced limits of error.

EXPERIMENTAL DESCRIPTION

I. Preparation of Chemically Pure NaCl

Our method of preparing chemically pure NaCl was similar to that employed by Richards and Wells,[5] in their determination of the atomic weights of sodium and of chlorine.

Pure H_2SO_4 was prepared by distillation of Graselli C.P. reagent H_2SO_4 with a distilling flask and condenser made from pure fused quartz. Two successive distillations were carried

out and the middle third of the distillate saved from each distillation.

Pure hydrochloric acid was prepared in a similar manner by double distillation of Graselli C.P. reagent hydrochloric acid, to which a few small crystals of C.P. $KMnO_4$ had been added.

Pure NaCl (preliminary) was precipitated from a saturated aqueous solution of Baker and Adamson's reagent quality NaCl mixed with pure hydrochloric acid (as prepared above) by the addition of HCl gas. The HCl gas for this precipitation was prepared by dropping pure H_2SO_4 (as prepared above) into an aqueous mixture of pure hydrochloric acid (cf. above) and Graselli reagent quality NaCl.

Pure HCl gas, used in subsequent steps in the purification, was prepared by dropping pure H_2SO_4 (cf. above) into an aqueous mixture of pure hydrochloric acid (cf. above) and pure NaCl (preliminary). The evolving gas was passed through two bead towers containing glass beads covered with pure H_2SO_4 (cf. above) and a final drying tube containing Coleman and Bell C.P. P_2O_5. The latter substance had been sublimed into the drying tube.

All aqueous solutions, in the above preparation of reagents, were made up with double distilled water (distilled from a tin still).

The purification of NaCl was continued by dissolving the preliminary preparation of pure NaCl (cf. above) in double-distilled water to which pure hydrochloric acid (cf. above) was added and precipitating with pure HCl gas (cf. above). The precipitated NaCl was centrifuged to remove adhering solution. This treatment was repeated with the precipitate until, in all, six successive precipitations had been made. Sensitive determinations of density revealed that the NaCl produced by this procedure showed small changes in density through five recrystallizations but that NaCl from the sixth, seventh, and eighth crystallizations was of constant density (cf. Table III, seq.).

Following the six successive precipitations by HCl gas and the subsequent centrifugations, the purified NaCl was fused in a platinum crucible to expel HCl.

Finally, the solidified melt was dissolved in warm triple-distilled water in a silver dish and given two recrystallizations from pure water.

[3] $\alpha_l = 40.5 \times 10^{-6}$, Straumanis, Ievins, and Karlsons, Zeits. f. anorg. allgem. Chemie **238**, 175 (1938). Cf. also Eucken and Dannohl.

[4] Clyde A. Hutchison and Herrick L. Johnston, J. Am. Chem. Soc. **62**, 3165 (1940).

[5] T. W. Richards and R. C. Wells, "A revision of the atomic weights of sodium and chlorine," Carnegie Institution of Washington, Publication No. 28 (1905).

TABLE I. Density of ethylene dibromide as a function of the temperature. 28.00°C = 3.843° Beckmann.

Temperature (Beckmann scale)	Densities (g/ml)	
	d_T	$d_{3.471°B}$
0.000	2.16879	2.16158
1.000	2.16673	2.16160
2.000	2.16474	2.16168
2.500	2.16370	2.16167
3.000	2.16265	2.16166
3.500	2.16158	2.16163
4.000	2.16053	2.16162
5.000	2.15843	2.16159

II. The Density Determination

(a) Calibration of the Flotation Medium

The density was determined by floating fragments of pure single crystals in a calibrated flotation medium of pure ethylene dibromide. Details of the procedure have been described elsewhere.[6]

The ethylene dibromide used for this purpose was prepared from Merck's reagent quality $C_2H_4Br_2$ (boiling point range 129°–132°C), first dried over C.P. $CaCl_2$, by distillation in vacuum. The middle third of the distillate was retained. As shown best by the constancy of the temperature of crystal flotation the isothermal density of the purified ethylene bromide did not change by more than experimental limits of error ($\pm 4 \times 10^{-6}$ g/ml) during the several days that measurements were in progress.

The density of the ethylene bromide, as a function of the temperature, was determined by weighing an approximate 9-ml Pyrex sinker immersed in the ethylene bromide. To compensate for possible small effects of surface tension on the 0.004″ suspension wire an auxiliary sinker, of lead, was attached to the lower end of the wire suspension and weighings were taken on: (1) the wire suspension with attached auxiliary sinker immersed in the liquid and (2) the complete suspension assembly, with both the 9-ml sinker and the auxiliary sinker immersed in the liquid.[7]

The exact volume of the 9-ml sinker was 9.10542±0.00002 ml at 27.145°C. This is the average of ten independent determinations, each

consisting of four weighings: (1) suspension +auxiliary sinker in air; (2) suspension+auxiliary and 9-ml sinkers in air; (3) suspension +auxiliary sinker in triple-distilled water at 27.145°; (4) suspension+auxiliary and 9-ml sinkers in triple-distilled water at 27.145°. The density of pure water at 27.145°C was taken to be 0.996502 g/ml.[8] The average deviation from the mean, among the ten determinations of volume, was ±0.00007 ml and the extreme deviation +0.00014 ml. All weighings were corrected to vacuum.

Densities of ethylene dibromide, determined at eight temperatures covering a range of five degrees, are given in the second column of Table I. For convenience, densities were determined at temperatures which corresponded to even divisions of the Beckmann thermometer used with the crystal flotations. The scale of this thermometer was quite uniform, as shown by calibration by the U. S. Bureau of Standards. To convert to degrees Centigrade, the Beckmann was compared with a standard mercury in glass thermometer, with a temperature range 27° to 32°C and graduated at intervals of 0.02°, which was calibrated by the U. S. Bureau of Standards shortly after use. The comparison gave

28.00°C = 3.843° Beckmann.

(b) Preparation of Flotation Crystals

Fragments of single crystals for the flotations were prepared from the purified NaCl (cf. I above) by the method developed by Kyropolous,[9] in the same manner as described previously.[4] It was found necessary to use an annealing period of four hours at a temperature 50° below the melting point of NaCl, and to allow at least four hours for the crystals to cool. It was also found that the densities of the crystals changed erratically when they were exposed to the atmosphere for several minutes. To avoid this erratic behavior it was found necessary to store them over P_2O_5, under vacuum.[10] When

[6] H. L. Johnston and C. A. Hutchison, J. Chem. Phys. 8, 869 (1940).

[7] This is the procedure followed by Osborne, McKelvey, and Bearce, Bull. U. S. Bur. Stand. Reprint No. 197 (1912).

[8] International Critical Tables (McGraw-Hill Book Company, 1928), Vol. 3, p. 25.

[9] S. Kyropolous, Zeits. f. anorg. Chemie 154, 308 (1926).

[10] These observations apply to the crystals at room temperature. Exposure to air during the high temperature annealing had no apparent effect on the density. In the subsequent cooling, which was carried out in the annealing furnace, P_2O_5 was introduced into the furnace, around the platinum crucible that contained the crystals, at about

immersed in the purified $C_2H_4Br_2$, so that they were not exposed directly to the atmosphere, the crystals maintained constant density.

Tables II and III illustrate these observations. Table II is a record of crystal flotations made with crystals that were grown from Baker and Adamson's reagent quality NaCl, without further purification. Table III is a record of crystal flotations with crystals that were grown from purified NaCl (cf. Section I) with varying numbers of HCl precipitations.

It is at once apparent that even C.P. NaCl experiences some manner of surface contamination when exposed to the atmosphere. It is noteworthy that the magnitude of density variations among exposed C.P. crystals is comparable to the density differences among natural crystals reported by Defoe and Compton.[1]

III. The Absolute Density

The average flotation temperature of the last eight crystals recorded in Table III is 3.477°

TABLE II. A record of crystal flotations with crystals that were grown from Baker and Adamson's reagent quality NaCl (unpurified).

Explanation: Crystals designated as 1a, 1b, and 1c were three fragments from a crystal that was annealed for only two hours and that was stored with exposure to the atmosphere. Crystals designated as 2a, 2b, 2c, and 2d were annealed for only two hours but were stored over P_2O_5 under vacuum. Crystals designated as 3a, 3b, and 3c were annealed at a temperature 50° below the melting point for four hours and were stored over P_2O_5 under vacuum.

Crystal designation	Flotation temperatures (° Beckmann)				cf. Footnote[a]
	Newly prepared	After 2 days	After 4 days	After 5 days	
1a	3.682	3.670			
1b	3.249	3.320			
1c	3.506	3.517			
2a	3.427		3.431		
2b	3.399		3.396		
2c	3.437		3.446		
2d	3.510		3.483		
3a	3.422			3.424	3.544
3b	3.497			3.495	3.458
3c	3.477			3.478	3.479

[a] After the five-day flotations made with crystals 3a, 3b, and 3c, crystals 3a and 3b were exposed to the atmosphere for twenty minutes and flotation temperatures redetermined. 3c was not exposed to the atmosphere. The entries in this column are the values obtained in the redetermination.

150°C. At 70°C the crystals were removed from the furnace and placed in the desiccator, which was immediately evacuated. Similar precautions were taken with the original melts prior to annealing. Crystals which had been once altered by exposure to air, at room temperature, were not restored to their original densities by a second annealing.

Beckmann with an average deviation from the mean of $\pm0.002°$—equivalent to $\pm3.6\times10^{-6}$ g/ml in the density of NaCl (cf. infra).

A large scale plot of the experimental densities of ethylene dibromide (column 2 of Table I) against the temperatures gives a near linear relationship. The best straight line through the data has a slope of $-2.074\pm0.01\times10^{-3}$ g/ml/ degree. Making use of this value for the temperature coefficient of density we have corrected each of the entries in column 2 of Table I to the common temperature 3.477° Beckmann. These corrected values are entered in column 3 Table I. The small systematic trend of the values recorded in this column is evidence of a slight curvature in the density temperature plot. By plotting the entries in column 3 against temperature and drawing in a smooth curve we obtain 2.16165 g/ml as the best value for the density of our purified ethylene dibromide at T Beckmann $= 3.477°$. We regard this as reliable to within about ±0.00002 g/ml. Comparison of

TABLE III. Crystal flotations made with crystals that were grown from purified Baker and Adamson reagent quality NaCl, in accordance with the purification scheme outlined in Section I.

Explanation: The crystals that are designated as 4a, 4b, 4c, and 4d were grown from purified NaCl that was given 4 precipitations with HCl gas, followed by fusion in platinum and 2 recrystallizations from triple-distilled water. Those designated as 6a, 6b, 6c, and 6d were given 6 precipitations with HCl gas, followed by fusion in platinum and 2 recrystallizations from triple-distilled water. Those designated as 8a, 8b, 8c, and 8d were given 8 precipitations with HCl gas, followed by fusion in platinum and 2 recrystallizations from triple-distilled water. All crystals were given a four-hour annealing at a temperature 50° below their melting point and were stored over P_2O_5 under vacuum.

Crystal designation	Flotation temperatures (° Beckmann)
4a	3.469
4b	3.456
4c	3.471
4d	3.461
6a	3.477
6b	3.472
6c	3.476
6d	3.476
8a	3.476
8b	3.477
8c	3.482
8d	3.477

Four crystal fragments were selected from the cooled melt of each preparation, and were annealed and floated. The table is therefore a record of all crystals that were floated from these three melts. With NaCl we did not experience the situation reported for LiF (reference 6): namely, that about one-half of the crystal fragments floated at diverse high temperatures, apparently due to internal cracks.

the Beckmann with the standard thermometer calibrated by the U. S. Bureau of Standards, at 28.00°C, yields 27.634°C for 3.477° Beckmann.

We can thus write for the absolute density of NaCl

$$d_{27.634°C} = 2.16165 \pm 0.00002 \text{ g/ml.}$$

Corrected to 20°C by use of the thermal coefficient of expansion for NaCl, this yields

$$d_{20°C} = 2.16366 \pm 0.00003 \text{ g/ml.}$$
$$= 2.16360 \pm 0.00003 \text{ g/cm}^3.$$

IV. Atomic Weight of Fluorine, Lattice Constant of Lithium Fluoride, Structure Constants of Calcite

In an earlier paper by C. A. Hutchison and one of us[11] we made use of the densities of LiF and of calcite, in conjunction with x-ray data to compute a value for the atomic weight of fluorine. The result obtained was 18.994 ± 0.002, based on 100.085 for the molecular weight of calcite. Corrected to the latest values[12] for M_{CaCO_3} this becomes

$$18.995 \pm 0.002,$$

a figure which is a little below the mass spectrograph value of 18.999 ± 0.001[13] but in excellent agreement with gas density determinations. This suggests the possible existence of a trace of some lighter isotope.

We can make a similar calculation from the densities of LiF and NaCl in conjunction with the ratio of their lattice constants and the molecular weight of NaCl. With true a_{LiF} taken to be 2.00904 ± 0.00002A[14] at 25°C; true a_{NaCl}

at 25° set equal to 2.815058A;[15] and with 2.63899 ± 0.0001 and 2.16228 ± 0.00002 taken for the densities of LiF[4] and of NaCl, respectively, the *ratio* of the molecular weights of LiF and of NaCl comes out

$$0.443640 \pm 0.000025.$$

With the atomic weight of lithium taken as 6.9390 ± 0.0002[16] and those of Na and Cl as correct at 22.997[12b] and 35.457,[12b] respectively, the atomic weight of F comes out

$$18.994 \pm 0.001.$$

With Birge's[12b] adoption for the atomic weight of sodium, namely 22.994 ± 0.003, the F value comes out

$$18.992 \pm 0.002.$$

It is apparent that the agreement of the atomic weight values for fluorine from calcite and rock-salt data, respectively, in conjunction with the x-ray and density data for fluorite, is as close as is permitted by the small uncertainties that exist in the atomic weights of Ca and of Na, respectively. It is also apparent that the comparison of molecular weights by combination of accurate x-ray and density data is as reliable as by other standard methods of atomic weight determination. In particular, it is apparent that there is no serious influence of "crystal mosaic" patterns such as postulated by Zwicky,[17] who thought that densities computed from crystal lattice constants might be in error by as much as 1 percent in some cases.

Avogadro's number is not used in this computation of relative molecular weights from x-ray and density data.

[11] C. A. Hutchison and H. L. Johnston, J. Am. Chem. Soc. **63**, 1580 (1941).

[12] (a) 100.09. "Eleventh annual report on atomic weights of the International Union of Chemistry," J. Am. Chem. Soc. **63**, 845 (1941). (b) 100.091 ± 0.005, R. T. Birge, Rev. Mod. Phys. **13**, 233 (1941).

[13] A. K. Brewer, Phys. Rev. **49**, 867 (1936).

[14] Straumanis, Ievins, and Karlsons, Zeits. f. physik. Chemie **B42**, 143 (1939). This value is based on the Siegbahn scale (*effective* $a_{calcite}$ in the first order = 3.02904A at 18°C) through close agreement of the wave-length assumed for copper $K_{\alpha1}$ (1.537395A) with that determined by J. A. Bearden and C. A. Shaw [Phys. Rev. **48**, 18

(1935)] through measurements on calcite (1.537400A) in which they assumed Siegbahn's value for the lattice constant.

[15] Based on the effective first order a calcite = 3.02904A at 18° with Siegbahn's experimental determination of the ratio of the *effective* grating spaces as 0.929007 and corrected for refraction (±0.000260A) and for thermal expansion (reference 3) to 25°.

[16] T. W. Richards and H. H. Willard, J. Am. Chem. Soc. **32**, 4 (1910); Zeits. f. anorg. allgem. Chem. **66**, 237 (1910); Carnegie Inst. Publication No. 25, 1 (1910).

[17] F. Zwicky, Proc. Nat. Acad. Sci. **16**, 211 (1930).