The Relative Abundance of the Isotopes of Lithium, Potassium and Rubidium

A. Keith Brewer and P. D. Kueck, *Bureau of Chemistry and Soils, Washington, D. C.*

The abundance ratios of the isotopes of lithium, potassium and rubidium have been measured with a Dempster type mass spectrograph. Artificial aluminum silicates of these alkalis were used as the source of ions. An abundance ratio of 12.14 ± 0.4 was obtained for Li^7/Li^6. On the basis of this ratio the masses of the two isotopes are 7.016 ± 0.002 and 6.016 ± 0.002 and the packing fractions are 23 ± 3 and 26 ± 3, respectively. The observed abundance ratio for potassium is $K^{39}/K^{41} = 13.88 \pm 0.4$. This value gives 38.96 ± 0.003 and 40.96 ± 0.003 for the masses of the two isotopes, and -9.85 ± 0.9 and -9.37 ± 0.9 for their respective packing fractions. A ratio of $Rb^{85}/Rb^{87} = 2.59 \pm 0.04$ was obtained for rubidium. On the basis of 85.44 for the atomic weight the masses of the isotopes come out to be 84.88 and 86.88 while the corresponding packing fractions are -14.1 ± 0.7 and -13.8 ± 0.7. The values for the packing fractions given above fall fairly closely on the packing fraction *vs.* mass number curve given by Aston for lithium and potassium but are about 70 percent low for rubidium.

T HE reports in the literature of the relative abundance of the isotopes of lithium and potassium differ widely, not only for various methods of measurement but between different investigators. In the case of lithium for mass-spectrographic measurements Harnwell and Bleakney[1] obtained a ratio for $Li^7/Li^6 = 8.4$, Bainbridge[2] gives 11.28 as the most probable value, Morand[3] found 14.9, while Dempster[4] obtained ratios ranging from 37.0 to 4.8 with an average value between 7 and 10. Using band spectra data, Wijkand and Koeveringe[5] found a ratio of 7.2 while Nakamura[6] obtained 8. Measurements of the abundance ratio of the potassium isotopes are limited. Dempster[4] observed a ratio of 18, while a published curve by Bainbridge[2] shows the ratio to be near 12.5. The best direct determination for rubidium is that of Aston[7, 8] who found $Rb^{85}/Rb^{87} = 3$ as the most probable ratio.

The results for lithium, potassium and rubidium obtained with a Dempster type of mass-spectrograph are presented in the present paper.

Apparatus

The spectrograph was made of brass with a 5 cm focusing radius. The analyzing chamber was designed for rapid evacuation. Various slit arrangements were tried, but the simple accelerating slit proved the most satisfactory. The slit openings were arranged to give flat topped peaks.

The resolved current was measured with an FP-54 Pliotron; the scale was checked for non-linearity by the application of known potentials after each series of runs.

In the case of lithium and potassium the filament aperature was a round hole, varying from time to time between 0.2 to 0.5 mm in diameter, located in the bottom center of a cup-shaped magnetic shield surrounding the filament. The slit in front of the Faraday cage was 1.5 mm wide and 5 mm long. The ion sources were artificially prepared aluminum silicates of the alkalies.[9] The filaments were of the short hairpin type with a straight section over the slit about 4 mm in length. The position of the filament was varied from 3 mm to 25 mm above the slit. Tests were made with and without a cylindrical shield 8 mm in diameter and 6.0 cm long surrounding the filament to concentrate the ion beam over the slit. When the shield was used the ion beam could be so focused that at 2.5 cm filament-slit separation the resolved current was one one-hundredth of the total ion current.

It was found necessary to change the slit arrangement slightly for a complete resolution of the rubidium isotopes. In this case the collector slit was 0.5 mm wide and 5 mm long, while the filament slit was 0.1 mm wide and 4 mm long. The filament, of the Hertz type, was a tungsten

[1] Harnwell and Bleakney, Phys. Rev. **45**, 117 (1934).
[2] K. T. Bainbridge, J. Frank. Inst. **212**, 317 (1931).
[3] Morand, Comptes rendus **182**, 460 (1926).
[4] Dempster, Phys. Rev. **18**, 415 (1921).
[5] Wijkand and Koeveringe, Proc. Roy. Soc. **A132**, 98 (1931).
[6] Nakamura, Nature **128**, 759 (1931).
[7] Aston, Proc. Roy. Soc. **A134**, 575 (1932).
[8] *Note:* For a general discussion see Aston, *Mass Spectra and Isotopes*, 1933 edition.

[9] Jones and Hendricks, Phys. Rev. **44**, 322 (1933).

hairpin on which a platinum disk 2 mm in diameter was spot-welded; the artificial rubidium aluminum silicate was coated on the disk.

The mass peaks were located by varying the field current of the magnet, the accelerating potential between the filament and slit being kept constant for each series of runs. Since the magnetic field could be varied through an appreciable range without changing the maximum of the resolved ion current the curves were all flat topped; values of these maximum currents therefore were taken as a measure of the relative abundance of the ions of the isotopes.

RESULTS

A typical mass vs. abundance curve for lithium is shown in Fig. 1. The mass is plotted as a function of the magnetic field current.

It will be noted that the flat topped peaks eliminated the difficulty in setting on the maximum of the sharp pointed peaks generally obtained. The resolution, as shown by the curves, was sufficient for a complete separation of the peaks.

The deviations in the measurements for a single filament are shown in Table I. The results

TABLE I. *Abundance ratio Li^7/Li^6 obtained with a single filament.*

No. of run	Li^7/Li^6	Deviation
1	12.28	+0.07
2	12.24	+0.03
3	12.18	−0.03
4	12.14	−0.07
5	12.12	−0.09
Average	12.21	

in general do not show any trend with time. A set of runs on a given filament usually extended over a period of about an hour. Since slightly coated filaments were used the life of a filament for constant emissivity was not materially longer than this.

The ratios determined from four separate filaments are shown in Table II. V_a is the accelerating potential, i_p the unresolved positive ion current, i_c the maximum resolved Li^7 currents to the collector, and n the number of tests made on each filament, average deviations are used in fixing the limit for the abundance ratio.
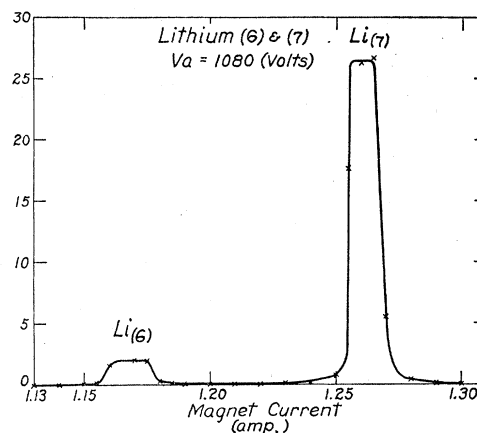
The data indicate a small trend towards higher

FIG. 1. Mass-spectrogram of lithium ions.

TABLE II. *Abundance ratio Li^7/Li^6 obtained with four different filaments.*

V_a (v.)	i_p (amp.)	i_c (amp.)	n	Li^7/Li^6
1080	5×10^{-8}	1.74×10^{-10}	5	12.21
1080	5×10^{-8}	1.7×10^{-10}	7	12.31
1080	3×10^{-8}	1.7×10^{-10}	7	11.36
1080	4.25×10^{-7}	9.0×10^{-10}	7	12.67
			Average	12.14 ± 0.4

ratios for increasing positive ion currents but it is not felt that much significance can be placed on this trend.

A typical curve for the isotopes of potassium is shown in Fig. 2. In general the resolution was complete, the deflection returning close to zero between the peaks.

The average results from different filaments are shown in Table III. The third value was ob-

TABLE III. *Abundance ratio K^{39}/K^{41}.*

V_a (v.)	i_p (amp.)	n	K^{39}/K^{41}
450	1.1×10^{-8}	8	13.26
450	6.0×10^{-8}	9	14.16
540		1	14.22
		Average	13.88 ± 0.4

tained from a special run made with a filament-slit separation of 3 mm and with no concentrating shield. The first two values were obtained with a separation of 2.5 cm The mass vs. abundance curves were identical in shape in both instances.

A representative curve illustrating the separation of the isotopes of rubidium is shown in Fig. 3.
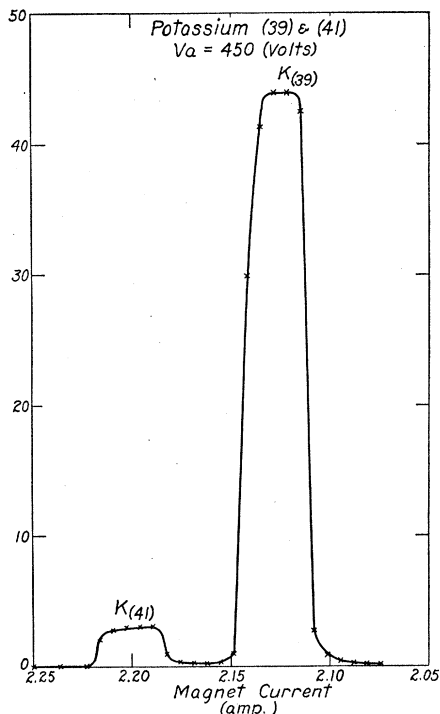
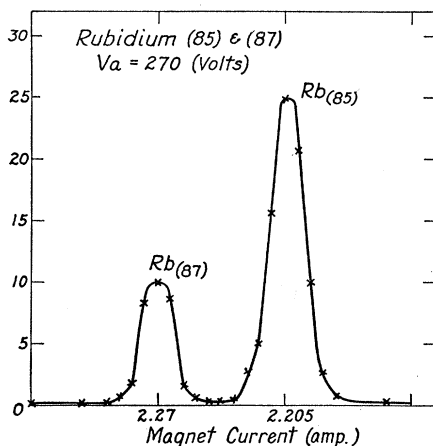FIG. 2. Mass-spectrogram of potassium ions.

FIG. 3. Mass-spectrogram of rubidium ions.

The curves are flat topped for small changes in the magnetic current; the separation of the two isotopes is complete.

The results of a series of runs for several filaments are shown in Table IV. These values were taken with the position of the filament over the slit such that a maximum resolved current was

TABLE IV.

V_a (v.)	i_p (amp.)	i_c (amp.)	n	Rb^{85}/Rb^{87}
270	5×10^{-8}	1.85×10^{-10}	10	2.54 ± 0.04
"	"	2.31	4	$2.55\pm\ .02$
"	"	1.5	5	$2.65\pm\ .03$
"	"	1.5	5	$2.63\pm\ .03$
"	"	1.5	7	$2.59\pm\ .04$
			Average	2.59 ± 0.04

obtained. The homogeneity of the primary ion beam was tested by measuring the isotope ratio when the filament was rotated to either side of the maximum; no change in the ratio was detected.

DISCUSSION OF RESULTS

An estimate of the abundance ratio from the ion emission ratio obtained in these experiments necessitates an understanding of the mechanism of diffusion of the alkali ions through the aluminum silicate. If perfect mixing takes place at the source, the movement of the alkali in the emitter will obey the laws of free diffusion; the ion ratio, therefore, must be corrected for the isotope effect in free evaporation by multiplying by the square root of the ratio of the heavy to the light isotope to give the abundance ratio. On the other hand, if the diffusion is of an electrolytic type in which the positive ions move step by step through a fixed negative ion lattice, the alkali ions in each underlying lattice layer moving up to take the place of those emitted, there will be no free diffusion effect and the ion ratio will represent the abundance ratio. It is impossible to state definitely which of the two mechanisms is correct, but the facts that the filaments were operated below the fusion temperature and that no change in the ion ratio could be detected throughout the entire life of the filament strongly support the latter alternative.

Assuming for lithium that the ion ratio represents the abundance ratio $Li^7/Li^6=12.14\pm0.4$ the mass of the isotopes on the basis of 6.94 for the atomic weight is $Li^7=7.016\pm0.002$ and $Li^6=6.016\pm0.002$. From these values the packing fraction expressed in parts per 10,000 is 26 ± 3 for Li^6 and 23 ± 3 for Li^7. Bainbridge,[10] measuring the mass of the isotopes directly, obtained

10 K. T. Bainbridge, Phys. Rev. 44, 56 (1933).

$Li^7 = 7.0146$ and $Li^6 = 6.0145$ on the basis of O^{16}. His values for the packing fraction, therefore, are $Li^7 = 20.85$ and $Li^6 = 24.2$. The two sets of data check each other fairly well, the agreement being within the experimental error in measuring the abundance ratio.

In the case of potassium the abundance ratio of 13.88 ± 0.4 is in fair agreement with 12.5 taken from the mass *vs.* abundance curve of Bainbridge, but is materially lower than the ratio of 18 found by Dempster and the value of 17.5 assumed by Aston (reference 8, p. 236). With 39.096 for the atomic weight of potassium this ratio gives 38.96 ± 0.003 and 40.96 ± 0.003 for the mass of the isotopes K^{39} and K^{41}, respectively, while the corresponding packing fractions come out to be 9.85 ± 0.9 and 9.37 ± 0.9. Since a small error in the abundance ratio necessarily makes a large error in the packing fraction it is significant that these values do not deviate from Aston's packing fraction-mass number curve (p. 167) by more than 20 percent; this is less than that found for many other elements.

The ratio $Rb^{85}/Rb^{87} = 2.59$ is materially lower than the value of 3 assigned by Aston for rubidium. On the basis of the atomic weight being 85.44 the masses of the isotopes 85 and 87 come out to be 84.88 and 86.88, respectively, while the corresponding packing fractions are -14.1 ± 0.7 and -13.8 ± 0.7. Aston has shown that the packing fractions for mass numbers 84 and 86 are -8.5 and -8.2, respectively; it seems probable, therefore, that the present values are in error. A packing fraction of -8.2 corresponds to an abundance ratio of 2.92 which is well outside the limits of experimental error in these experiments. While it does not seem possible to account for the discrepancy at this time, it is interesting to note that an abundance ratio of 2.59 ± 0.04 corresponds to an atomic weight for rubidium of 85.48 ± 0.005 in place of the accepted value of 85.44.

The Genesis of the Elements

GILBERT N. LEWIS, *Department of Chemistry, University of California*
(Received September 26, 1934)

The hypothesis is suggested that a great part of the matter in the universe is composed chiefly of iron and nickel, like the metallic meteors, and that such material, which is thermodynamically stable with respect to all spontaneous transmutations, except at extremely high temperatures, is superficially attacked by cosmic radiation to produce the material represented by the earth's crust and by the stony meteors. As a test of this hypothesis of the genesis of stony meteors from metallic meteors, a comparison is made of the relative abundance of the chief atomic species in the two types of meteors. The striking results of this comparison strongly indicate a genetic relationship. Three main disintegration processes to which the iron and nickel nuclei are subjected seem to be the splitting off of oxygen from these nuclei; the splitting of the nuclei into two identical parts; and the splitting of helium from the products of the preceding process.

IN spite of the extraordinarily interesting information regarding nuclear structure which is now coming week by week from experiments on atomic disintegration, it seemed to me that it might still be possible to extract useful information from the older data, especially those related to the relative abundance of the elements. It was Harkins[1] who first called attention to the striking connection between the atomic weights of the elements and their abundance, not only in the earth's crust, but in the meteors. For the latter he made use of analyses collected by Farrington[2] for hundreds of meteors of the stony and of the metallic type. These results, and Aston's[3] data concerning the relative abundance of isotopes, form the basis of this paper.

---

[1] Harkins, J. Am. Chem. Soc. **39**, 856 (1917).

[2] Farrington, Publications 120 and 151, Field Columbian Museum, Chicago.

[3] Aston, *Mass-Spectra and Isotopes*, Longmans, Green and Co., New York, 1933.