TABLE I

VAPOR PRESSURE OF SODIUM

Temp., °C.	Pressure, mm.	Temp., °C.	Pressure, mm.	Temp., °C.	Pressure, mm.
396.5	0.3025	346.0	0.0677	296.5	0.01205
392.0	.2700	339.0	.0538	291.0	.00955
389.5	.2400	331.0	.0438	286.0	.00760
384.0	.2195	329.5	.0381	280.0	.00603
381.0	.1952	323.0	.0332	273.0	.00480
376.0	.1702	319.5	.0289	271.0	.00427
371.0	.1517	312.5	.0235	268.0	.00381
366.0	.1233	310.0	.02045	267.0	.00347
360.0	.1074	302.0	.01520	263.0	.00303
354.0	.0876	300.0	.01385		

given by Rodebush and Walters as the best representation of all previous data. It is evident from Fig. 2 that the new data confirm the correctness of the equation in a most satisfactory manner.

URBANA, ILLINOIS

[CONTRIBUTION FROM THE GEOPHYSICAL LABORATORY, CARNEGIE INSTITUTION OF WASHINGTON]

ISOTOPES AND THE PROBLEM OF GEOLOGIC TIME

BY CHARLES SNOWDEN PIGGOT

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Time can be estimated only by reference to some change, and geologic time is no exception. Any change demands a beginning and an ending condition, and a rate.

The phenomena which fulfil these conditions throughout the great lengths of geologic time are: (1) denudation and sedimentation, (2) the salting of the ocean and (3) radioactive disintegration of uranium into lead.

They are all of the hour-glass principle and if we could measure the amounts involved at the beginning and at the end—and ascertain the rate of change—the problem of geologic time would be a very simple one. But of the three, only the last can be measured with the accuracy necessary to inspire confidence in the result. We can well imagine what a difficult task it was to arrive at any accurate estimate of the amount of material that had been eroded from the highlands and deposited in the hollows to form sedimentary formations, particularly when it was known that these sedimentary formations often became, in turn, highlands from which further erosion took place.

It was not much easier to estimate the rate at which sodium was being leached from the igneous rocks and carried into the sea. It was easy to determine the sodium in a rock, and in a sample of sea water; but to decide on the amount of rock and the amount of sea was not so simple, nor did such minor corrections as the great salt deposits necessitated make the problem any easier for the old-time geologist.

With the discovery of radium and the unraveling of the mysteries of radioactive disintegration, and especially the demonstration of the relationship existing between uranium and lead, the geologist felt that at last he had an accurate timepiece, of relative simplicity, and one capable of most refined measurements.

And so he had, compared with the ones he had been struggling with before, and the new tool was adopted with enthusiasm. However, as refinement of measurement and accuracy increased, factors which at first had been ignored began to assume increasing importance, and it is some of these minutiae that I wish to discuss now.

If it were only a case of ascertaining the amount of uranium and the amount of lead present in a mineral and applying that information to the known rate of change of the uranium–radium–lead series the problem would resolve itself into simply one of refined chemical analysis; but some thorium is almost always present in uranium minerals, which, besides complicating the chemical analysis, makes necessary a correction to take care of that portion of the total lead that came from the thorium. This correction factor (0.38 Th) is shown in the formulas given below.

Simplified Formulas (Approximate) for Calculation of Geologic Time

$$\text{Age} = \frac{\text{Pb}}{\text{U} + 0.38\ \text{Th}} \times 7400 \text{ million years} \tag{1}$$

$$\text{Age} = \frac{\log\left(\text{U} + 0.38\ \text{Th} + 1.55\ \text{Pb}\right) - \log\left(\text{U} + 0.38\ \text{Th}\right)}{6.5} \times 10^{11} \text{ years} \tag{2}$$

It is supposed to represent the amount of uranium which is equivalent in lead-producing capacity to 1 g. of thorium. Unfortunately, this value is not universally accepted, nor is the time required for a given amount of thorium to form its corresponding amount of lead known with satisfactory accuracy. Therefore the inevitable presence of thorium introduces an annoying element of uncertainty into any formula that may be devised. So, if we could eliminate consideration of the thorium altogether, it would be a great relief.

Furthermore, there is no actual measure of that proportion of the total lead which is known to have been produced by the uranium alone. If this latter could be determined by actual experimental measurement, the thorium could be disregarded altogether and the only other uncertainty which would remain inherent and unmeasured in this method would be the existence of possible isotopes of uranium which have disintegrated at a different rate in the past from the uranium which we know today.

We attempted to accomplish all this by the identification and the determination of the isotopes of lead. As this had not been done, we first began work on "ordinary" lead. The problem was to get the lead into some volatile form with the proper polarity to enable it to be swept along in the stream of positive rays in a mass spectrograph.

Lead tetramethyl was made and the technique developed to a point where a workable quantity could be obtained from 5 g. (or less) of lead chloride.[1]

Dr. F. W. Aston,[2] F.R.S., kindly consented to coöperate and in July, 1927, he carried out several experiments with this material. These demonstrated very clearly the existence of the three anticipated isotopes of masses 206, 207 and 208 in the approximate ratios of 4, 3 and 7, respectively, as shown in Table I.

TABLE I

RELATIVE PROPORTIONS OF LEAD ISOTOPES

	Percentage of isotopes Pb 206	 Pb 207	 Pb 208
Ordinary lead	28.6	21.4	50.0
Radioactive lead	86.8	9.3	3.9

The existence of isotope 207 demonstrated that the atomic weight of 207.2 was not a statistical mean of isotopes 206 and 208 only.

The next step was to do the same for "radioactive" lead. This was obtained from a very pure sample of Norwegian bröggerite, a mineral which is high in uranium but contains very little thorium. In Professor Aston's mass spectrograph this lead also revealed isotopes 206, 207 and 208. The presence of isotope 207 in this radioactive lead was somewhat of a surprise, but when it was noticed that the relative proportions were radically different, its presence took on added significance.[3]

These results demonstrate that isotope 207 exists even in lead of radioactive origin. Consequently it must have a corresponding source, and this source is now assumed to be an isotope of uranium which is the parent substance of the actinium series.

To quote from Aston's report in Nature of March 2, 1929: "The line 207 is of peculiar significance. It cannot be due to the presence of lead as an impurity, for in ordinary lead 208 is about twice as strong as 207, neither can it be the product of radium or thorium. It is difficult to resist the natural conclusion that it is the end-product of the only other known disintegration, namely, that of actinium. If this is so it settles the mass numbers of the members of this series, that of protoactinium being 231."

We can now assume that the actino-uranium (parent substance of the actinium series) has a mass of 239 and is converted into a mass 235 in consequence of an alpha change followed by two beta changes, as is the

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[1] This was done for me by Mr. S. C. Witherspoon, then of the U. S. Chemical Warfare Service.

[2] Aston, *Nature*, **120**, 224 (1927).

[3] Aston. *ibid.*, **123**, 313 (1929).

case in the uranium–radium series, or a mass 235 to begin with, which passes through an alpha and a beta transformation to form protoactinium.

Both alternatives are suggested and discussed by Rutherford.[3]

By whatever method they were formed, we now are certain of three leads each legitimately descended from its corresponding parent substance, as is illustrated diagrammatically in Table II.

TABLE II

ORIGINS OF THE THREE LEAD ISOTOPES

$$\begin{tikzcd}
U_{238} \arrow[d, "8\alpha"] & U_{239} \text{ or } {}_{235} \arrow[d, "8\alpha \text{ or } 7\alpha"] & Th_{232} \arrow[d, "6\alpha"] \\
Pb_{206} & Pb_{207} & Pb_{208}
\end{tikzcd}$$

By means of the mass spectrograph and an intensimeter we are now able to determine the amount of Pb_{206} in the lead separated by analysis of a uranium mineral, but as yet we cannot determine the actual U_{238} from which it came. This will require the determination of the isotopes of uranium—as yet not accomplished.

Dr. Moses Gomberg is very generously coöperating with us in an effort to secure some volatile organic compound of uranium which could serve as the tetramethyl did for the lead, but uranium is infinitely more difficult to cope with than lead, because of its multiplicity of valences and its annoying habit of combining with itself.

The field of the organic compounds of uranium is practically untouched.

Should the determination of the isotopes of uranium be accomplished, we would then have a measure of the particular uranium from which the Pb_{206} came, and our formula would be simplified thereby. Further knowledge of the isotopes of uranium would be of great value in many lines of research, but to mention only one having tremendous significance for the problem of geologic time, it is very important to learn whether or not the U_{238}–Pb_{206} series disintegrates at a different rate from that determined for the mixture of isotopes as a whole.

WASHINGTON, D. C.