Lead Isotopes

Edna R. Bishop, Margaret Lawrenz and C. B. Dollins, *Alabama Polytechnic Institute*

(Received October 8, 1932)

The magneto-optic method shows that lead has sixteen isotopes of probable mass and order of abundance 208, 206, 207, 205, 212, 210, 204, 202, 203, 211, 201, 209, 216, 215, 214 and 213. All sixteen isotopes of lead were found in C.P. uranium and thorium salts. 206, 210, 202 and 214 predominated in uranium salts, while 208, 216, 204 and 212 predominated in thorium salts. The differential time lag varies directly with the weight of the isotopes of a given element. The amount of isotope present is some unknown inverse function of the concentration necessary to read its minimum so that the order of appearance of minima gives the order of abundance but not the relative abundance of isotopes. The order of abundance of isotopes is also given by the amount of rotation of the analyzing nicol necessary to extinguish their minima.

THE magneto-optic method[1] is useful in the study of isotopes[2] since the number of differential time lags (detected by minima of light intensity) characteristic of any inorganic compounds is equal to the number of isotopes of the cation.

Lead was chosen for further study of isotopic relations obtained by this method of analysis because of the partial separation of its isotopes in nature. Three problems were investigated; the number of lead isotopes, the time lag as a function of the mass of the isotopes, the quantitative relation between the concentration at which the minima appear, and the relative abundance of the isotopes.

For the determination of the number of isotopes, C.P. chemicals were used at a concentration of about one part in 10^{11} of water redistilled from Pyrex. Solutions of each compound were prepared from at least two independent sources and examined on different sets of apparatus by at least three different observers,[3] two working in this laboratory and one at Emory University. The location of minima was reproducible by the same or different observers in both laboratories within 0.01 scale division. If a desired compound was not available, it was obtained by adding a solution of a lead-free compound of the required anion to a nitrate or chloride solution of the cation being studied.

Four different lead compounds were studied and sixteen minima found for each as shown in Table I.

In order to study the time lag as a function of the mass of the isotopes, uranium and thorium compounds were examined for lead minima. One was expected in each, Pb^{206} in uranium and Pb^{208} in thorium. However, concentrated solutions of either gave all sixteen of the lead minima. As the solutions were made more dilute, a concentration was reached at which each solution gave only one lead minimum which is therefore attributed to Pb^{206} and Pb^{208}, respectively, as shown in Table I. This substantiates former evidence[2] that the time lag varies directly with the mass of the isotopes of a given element. It further shows that there are five isotopes lighter than 206 and eight heavier than 208. Consecutive weights have been tentatively assigned to them as being the most probable. This is further substantiated by quantitative work.

For the quantitative study, the method employed consisted of finding the smallest amount of substance which would produce the light minimum characteristic of each isotope. Stock solutions were prepared by dissolving a weighed amount of salt in 100 cc of water. From these a series of dilutions was prepared by successively diluting 10 cc to 100 cc. The most dilute was examined for lead chloride minima and then each consecutive increasing concentration until the minima appeared. Let the concentration in which

---

[1] Allison and Murphy, J. Am. Chem. Soc. **52**, 3796 (1930).

[2] Allison and Murphy, Phys. Rev. [2] **36**, 1097 (1930); Allison, Ind. Eng. Chem. (Anal. Ed.) **4**, 9 (1932); Bishop, Phys. Rev. [2] **40**, 16 (1932).

[3] We acknowledge with thanks the further checking of many of the minima by Dr. Fred Allison and Mr. Roy Goslin.

TABLE I. *Scale readings and differential time lags with respect to carbon disulfide of lead isotopes in different compounds.*

Probable atomic mass of lead isotopes	$PbCl_2$		$Pb(NO_3)_2$		$PbSO_4$		$Pb_3(PO_4)_2$	
	Scale reading	Sec. $\times 10^9$	Scale reading	Sec. $\times 10^9$	Scale reading	Sec. $\times 10^9$	Scale reading	Sec. $\times 10_9$
201	37.90	−22.90	10.52	4.48	25.72	−10.72	48.80	−33.80
202	37.79*	−22.79	10.46*	4.54	25.61*	−10.61	48.62*	−33.62
203	37.68	−22.68	10.39	4.61	25.45	−10.45	48.40	−33.40
204	37.50	−22.50	10.30	4.70	25.36	−10.36	48.22	−33.22
205	37.40	−22.40	10.16	4.84	25.23	−10.23	48.02	−33.02
206	37.30*	−22.30	10.08*	4.92	25.12*	−10.12	47.72*	−32.72
207	37.08	−22.08	9.95	5.05	24.90	− 9.90	47.50	−32.50
208	36.90**	−21.90	9.90**	5.10	24.72**	− 9.72	47.40**	−32.40
209	36.78	−21.78	9.85	5.15	24.60	− 9.60	47.25	−32.25
210	36.68*	−21.68	9.80*	5.20	24.49*	− 9.49	47.10*	−32.10
211	36.59	−21.59	9.71	5.29	24.36	− 9.36	47.00	−32.00
212	36.50	−21.50	9.60	5.40	23.92	− 8.92	46.84	−31.84
213	36.40	−21.40	9.53	5.47	23.82	− 8.82	46.70	−31.70
214	36.28*	−21.28	9.49*	5.51	23.62*	− 8.62	46.53*	−31.53
215	36.19	−21.19	9.44	5.56	23.50	− 8.50	46.38	−31.38
216	36.10	−21.10	9.33	5.67	23.38	− 8.38	46.24	−31.24

* The four most abundant lead isotopes given by a solution of the corresponding uranium compound.

** Most abundant isotope given by a solution of the corresponding thorium compound.

the minima appeared be $1/10^{n+1}$ of the stock solution. A 10 cc burette was then filled with $1/10^{n+2}$ solution which was added in ½ cc portions to a cell containing approximately 50 cc of water and examined for lead chloride minima between each addition until they appeared. This gave the concentration range in which to work. An accurate solution was then prepared corresponding to the most concentrated that did not produce minima. By means of a specially calibrated pipette 49.75 cc of this solution was transferred to a clean cell and the $1/10^{n+2}$ solution was added from the burette drop by drop until each minimum just appeared. The concentration with respect to the stock solution, at which each minimum appears is the concentration placed in the cell increased by as many tenths as there were twentieths of cc added from the burette. The absolute concentration is given by multiplying this value by the g/cc in the stock solution.

A lead chloride solution was determined in this manner. The process was repeated adding a small amount of sodium ammonium phosphate which had been shown to be free from lead by testing with the magneto-optic method. Lead chloride and lead phosphate minima were read simultaneously. The minima became visible in pairs at the same concentration as when the chloride only was present. All minima in these quantitative studies were set on "in the dark." One person set the scale near the known location of a minimum. Another person looked to see whether or not he could detect that minimum in the given concentration. As the observer did not know which of the possible sixteen minima he was trying to set on or which were the pairing ones between the chloride and phosphate, the observations were highly objective. Similar simultaneous determinations of lead chloride and phosphate were made of uranium and thorium salts. The results are shown in Table II.

In ordinary lead, the order of appearance of the first three minima could be determined with certainty and are in agreement with the order of abundance as found by Aston.[4] After that, the minima appeared in such close succession that it was very hard to distinguish their order. They are arranged in their most probable order as given by several different determinations. After the completion of this work, it was found that, if the analyzing nicol were rotated, the minima would disappear in inverse order of abundance of the isotopes. Ordinary lead was rechecked by this method with the phosphate at a concentration of approximately one part of lead in 10^5 of water. Minima were obtained with the circle reading 90° and turned toward 0° to extinguish them so that a minimum circle reading corresponds to a maximum rotation and hence the most abundant isotope. By rotating the nicol it was possible to make

---

[4] Aston, Nature **120**, 224 (1927); **129**, 649 (1932).

TABLE II. *Probable order of abundance of lead isotopes in ordinary lead, uranium and thorium salts.*

Ordinary lead			Uranium salt		Thorium salt	
Atomic mass	Concentration in g/cc × 10^{12} at which minima first became visible	Circle reading at which minima first disappeared	Atomic mass	Concentration in g/cc × 10^4 of uranium at which minima first became visible	Atomic mass	Concentration in g/cc × 10^4 of thorium at which minima first became visible
208	3.15	54°57′	206	4.05	208	4.30
206	3.25	55°5′	210	4.10	216	4.60
207	3.30	55°11′	202	4.10	204	4.70
205	3.34	55°18′	214	4.10	212	4.70
212	3.38	55°24′	207	4.20	206	4.85
210	3.40	55°32′	208	4.20	207	4.85
204	3.40	55°40′	215	4.20	202	4.90
202	3.40	55°43′	203	4.20	205	4.90
203	3.40	55°46′	205	4.30	214	4.90
211	3.40	55°48′	204	4.30	211	4.90
201	3.40	55°49′	209	4.30	209	4.90
209	3.40	55°51′	211	4.30	210	4.90
216	3.40	55°52′	212	4.30	203	4.90
215	3.45	55°58′	213	4.30	213	4.90
214	3.45	56°1′	216	4.30	215	4.90
213	3.45	56°12′	201	4.30	201	4.90

finer discriminations between the appearance of the minima than formerly. The results, shown in Table II, obtained by employing this new method were in good agreement with previous results. Aston[5] has recently found eight isotopes of lead and finds the determination of the abundance of all but the first three uncertain. His most probable order (208, 206, 207, 204, 209, 210, 203 and 205) however, differs from the results of this investigation.

Schüler and Jones[6] reported spectroscopic evidence for Pb^{204} and later suggested[7] that Aston's Pb^{203} and Pb^{205} might be due to thallium with which they are isobaric. Such could not be the case in this work as determinations were made at threshold concentrations, i.e., between 3 and 10 parts of lead in 10^{12} of water redistilled from Pyrex so that any contamination would have had to be present in amounts comparable to that of lead which is highly improbable in several different samples of C.P. salts. Further, isobars do not give coincident minima as the time lag varies *inversely* with the equivalent weight of different cations and *directly* for isotopes of the same element and, since the time lag is a function of equivalent weight, the minima for thallium which is uni- and trivalent are far removed from those of divalent lead.

In uranium salts, the Pb^{206} has a very slight lead over the other fifteen isotopes. It was followed very closely by Pb^{210}, Pb^{202} and Pb^{214} after which the rest appeared in such close succession that no order could be established. Since Pb^{210} and Pb^{214} are known to be transition products in the uranium-radium series, this gives experimental evidence for the assignment of consecutive atomic masses and suggests that Pb^{206} may undergo further decomposition to form Pb^{202}. The first four lead minima that appeared in each uranium compound are shown in Table I.

In thorium salts, on the other hand, Pb^{208} had considerable lead. It was followed by Pb^{216}, Pb^{204} and Pb^{212} which suggests that all of these may be successive products in the decomposition of thorium. They could not be due to contamination of ordinary or uranium lead as their relative abundance is higher than in either of these. The other minima appeared in such close succession that no order could be established with certainty. The first lead to appear in each thorium compound is shown in Table I.

From the concentrations at which lead minima were read (Table II), the concentration of lead in uranium and thorium was approximately one part in 10^8. This is more than could have been formed since the preparation of the respective salts. The amount, however, might easily be present as an impurity in a C.P. salt but the

---

[5] Aston, Nature **129**, 649 (1932).

[6] Schüler and Jones, Naturwiss. **20**, 171 (1932).

[7] Schüler and Jones, Nature **129**, 833 (1932).

uranium and thorium leads would be a more likely source of contamination than ordinary lead. This is substantiated by the very different isotopic composition.

This work is in agreement with former evidence[2] that the abundance of the isotopes is an inverse function of the concentration at which their minima are first detected. Since this function is still undetermined, only the order of abundance and not the relative abundance of the isotopes can be found. It is clear that they exert some kind of mutual influence over each other and appear at concentrations very close together even though their relative abundance may be widely separated.

We wish to express our appreciation to Dr. Fred Allison for the use of his apparatus and for his constant interest and cooperation in the work, and to Dr. C. S. Piggot for critical reading of the manuscript.