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U-Pb and Pb-Pb Age Determinations on Zircons

Results Obtained with a Direct Ionization Method for the Isotopic Analysis of Lead

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With 3 figures and 2 tables in the text

Résumé

Le présent article décrit et discute une méthode d'analyse isotopique du plomb contenu dans les zircons, cela sans extraction chimique préalable de cet élément, comme il est habituel de le faire. La méthode a été appliquée à deux échantillons de zircons du Col des Gets (Haute Savoie, France), à deux échantillons de zircons du Massif du Mont-Blanc et à quatre concentrés provenant du Groenland. En utilisant la fluorescence des rayons X pour déterminer les teneurs des zircons en uranium et plomb, l'analyse isotopique par spectrométrie de masse peut être utilisée valablement sur des zircons contenant au moins 80 ppm de plomb.

INTRODUCTION

KOSTOLANYI (1964) described a method for the isotopic analysis of lead without prior extraction from minerals. He used this method in order to determine the isotopic ratios of lead in uranium-bearing minerals like pitchblende, autunite, chalcocite and colombo-tantalite (KOSTOLANYI, 1963; KOSTOLANYI and DULLIER, 1966). Later, the same technique was used to date some zircons (ROUBAULT et al., 1967; COPPENS et al., 1964–1965); $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ¹⁾ ages for samples containing less than 5% common lead were reported.

Since we have been dating zircons, using the "total lead" method, since 1962 (BUCHS et al., 1962; CHESSEX et al., 1964; BERTRAND et al., 1965), the uranium, thorium and lead contents being determined by X-ray fluorescence spectrometry according to a procedure described by one of us (BUCHS, 1962), we found it interesting to test KOSTOLANYI's direct ionization technique

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¹⁾ Pb_r = radiogenic lead.

on some of our zircon samples. As the method does not require tedious separation procedures for the isotopic analysis of lead, it was worthwhile to explore its possibilities and limitations.

ANALYTICAL PROCEDURE

The thermoionization is performed with a CH-4 Varian MAT mass spectrometer by using two-filament insert units. The first diagram (a) of Figure 1 shows a portion of such an insert unit with the two filaments, and the second diagram (b) shows the position of the filaments with respect to the plates which are used to extract the ions from the ion source. The insert unit is a separate part of the ion source, i. e., the filaments are mounted on the insert unit outside the ion source and the unit is introduced into the ion source by means of a vacuum lock. The sizes of the filaments are: $0.04 \times 0.7 \times 7.0$ mm. Rhenium filaments give better results than either tungsten or tantalum filaments. It is not too difficult to obtain stable ion beams with tungsten filaments, in contrast to tantalum filaments, but the temperature required for ionization is then much higher than with rhenium filaments.

For each experiment about 0.2 mg of finely powdered zircon is coated on filament B (see Fig. 1) with two drops of a 1 : 1 mixture of HF (48–51%) and H_3PO_4 (85%). The coated filament is then heated outside the ion source with a 1.5 Amp. current; after about 10 minutes the intensity of the current is increased to 2.0 Amp. and maintained so until all the white fumes have disappeared. The insert unit is then introduced in the ion source of the mass

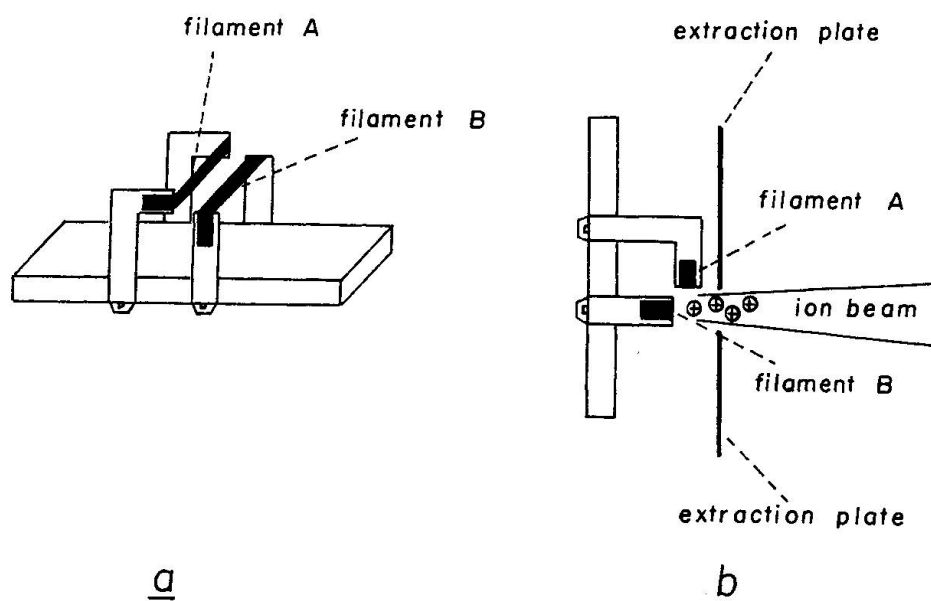


Fig. 1. Diagram of an insert unit.

spectrometer. New filaments are used for each experiment. The preparation of the zircon samples is described elsewhere (CHESSEX et al., 1964).

RESULTS AND DISCUSSION

In order to get reliable results for the age determinations two conditions had to be fulfilled: one had to find a way to eliminate any background spectrum at masses 204, 206, 207 and 208, and it was necessary to be sure that the U-Pb_r ages were not altered by common lead not incorporated in the crystal lattice of the zircon. Common lead which is adsorbed on the surface of the zircon or which is introduced during the preparation of the sample or the coating of the filament will be referred to as "contaminating lead", while lead which is incorporated in the crystal lattice of the zircon (original common lead and radiogenic lead) will be referred to as "zircon lead".

When we proceed in the way described by KOSTOLANYI (1964), i. e., degassing the sample inside the ion source by heating first gradually filament A (see Fig. 1) during about 10 minutes up to approximately 2000° C and then filament B (see Fig. 1) to 700° C, and maintaining these temperatures during 20 minutes before attempting to start the ionization process, we could never obtain a thoroughly clean background. This difficulty can be overcome by heating first filament A with a 4.5 to 4.7 Amp. current, this maximum value being reached in about 10 minutes. Afterwards, filament B is heated with a 1 Amp. current and those two intensities are maintained for at least 12 hours (overnight). The current on filament A is then shut off, and after the ion source has cooled down, the current on filament B is very slowly increased until ionization begins.

Working according to this procedure results in a clean background in almost every case. Nevertheless, even when there is apparently no background spectrum in the mass region between m/e 204 and m/e 208, one has to watch carefully for an eventual hidden background peak at mass 204; an unsuspected background peak at that mass will naturally affect drastically the correction which has to be performed for the occurrence of common lead. Figure 2 shows a plot of the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ratio vs. the ionization current. Intensity readings of the signals at masses 204, 206 and 207 were collected for increasing values of the ionization current. The result of the experiment shows that the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ratio increases until the ionization current reaches approximately 1.85 Amp. The ratio then settles at a fairly stable value which remains unaffected by further increasing the temperature of the filament. Such a behaviour is a clear indication that there was an unidentified background peak superimposed on the ^{204}Pb peak. The number of sets of peaks measured at each value of the ionization current is also reported in Figure 2. From 1.85 Amp. up to 1.98 Amp. the intensity of the ionization current was modified every

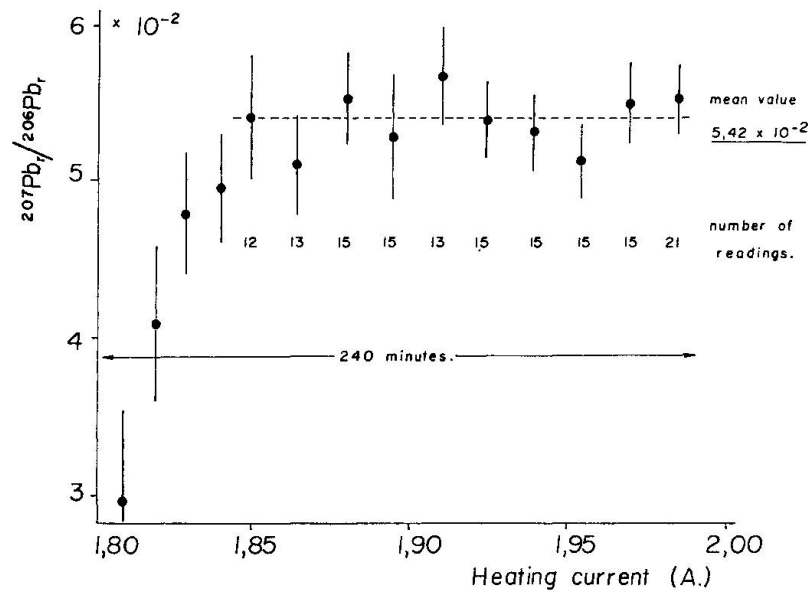


Fig. 2. Plot of the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ratio vs. ionization current.

20 minutes. The error limits indicated in both Figures 1 and 2 are based only on statistical fluctuations. The precision of the reported values is given at 95% confidence level.

It should be stressed that even when no signal is detected at mass 204, i. e., when the intensity of the signal is below the detection limit of our instrument, which is approximately equal to 4×10^{-18} Amp., the same effect is sometimes observed, but to a much lesser extent than for the example illustrated in Figures 2 and 3.

The amount of "contaminating lead" (lead adsorbed on the zircon and lead which is introduced during the coating procedure with the $\text{HF}/\text{H}_3\text{PO}_4$ mixture) cannot be measured with this technique. However, our experiments show that this lead does not behave in the same manner as does the "zircon lead". At any ionization temperature of the filament, the "contaminating lead" is evaporated at a faster rate than the "zircon lead" and can therefore be eliminated during the ionization process. However, we could never observe the appearance of a pure common lead spectrum at the very beginning of ionization; there is first a mixed spectrum of "contaminating lead" and "zircon lead".

The concentration of common lead in the lead evaporating from the zircon sample for which Figure 2 shows the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ratio vs. current intensity, is reported in Figure 3 for increasing values of the ionization current. Since the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ratio shows a monotonous drift between 1.81 Amp. and 1.85 Amp. (see Fig. 2), the first four values reported in Figure 3 (those below 1.85 Amp.) do not represent the actual concentration of common lead. The % of common lead continuously decreases to reach a minimum value toward the end of the ionization process. The only way to ascertain that the minimum

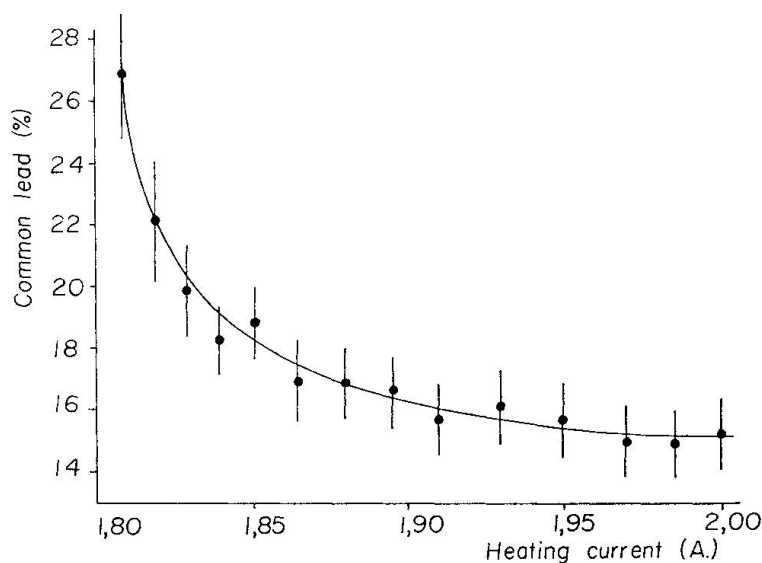


Fig. 3. Plot of the % of common lead vs. ionization current.

value which is reached actually represents the concentration of original common lead (incorporated into the crystal lattice) is to repeat the experiment with the same sample, with different coatings. Two separate runs with the zircon sample for which the results of one of the runs are reported in Figure 3 gave respectively $16.3 \pm 1.2\%$ and $14.9 \pm 1.3\%$ common lead.

In order to test the reliability of the technique we first compared our results with those reported by STERN et al. (1966), for a zircon which had been analyzed according to the usual procedure, i. e., after the lead had been chemically extracted. According to STERN et al., who sent us the sample, the zircon had been washed with concentrated HNO_3 in order to remove as much "contaminating lead" as possible before the lead was extracted and analyzed for its isotopic composition.

Our results, reported in Table 1, indicate that there is less than 0.6% common lead in that zircon, a result which is not in agreement with the one published by STERN et al. But if one subtracts from STERN's results the common lead contributions to isotopes 206, 207 and 208, the agreement between STERN's results and ours is quite good. This clearly indicates that the 4.5% common lead which were reported were not "zircon lead". The "contaminating lead" was completely eliminated during the early phase of ionization. The 0.6% common lead reported in Table 1 represents an upper limit. In actual practice the peak at mass 204 had completely vanished before valid readings were collected. The highest ion current which was obtained amounted to 5.305×10^{-14} Amp. (sum of ion beams 206, 207 and 208). With an electron multiplier having a gain factor of 2×10^6 , with an output resistance of 5×10^6 ohms, this corresponds to a 530.5 mV signal on our recorder. Since the smallest signal which we can differentiate from background noise is approximately

Table 1. *Comparative results obtained with a zircon sample by the direct ionization method and by isotope dilution analysis on chemically separated lead*

	Concentration ppm		Relative atomic abundance %							
	U	Pb	²⁰⁴ Pb	²⁰⁶ Pb	²⁰⁷ Pb	²⁰⁸ Pb	% common lead	²⁰⁶ Pb _r ¹⁾	²⁰⁷ Pb _r	²⁰⁸ Pb _r
STERN et al.	406	139	0.067	70.70	20.11	9.12	4.5	72.88	19.98	7.07
This work	—	—	0.008	—	—	—	0.6	72.76 ± 0.55 ²⁾	20.56 0.41	6.70 0.34

¹⁾ Pb = radiogenic lead.

²⁾ Precision of these values at 95% confidence level.

equal to 4.5×10^{-2} mV, i. e., an ion current of 4.5×10^{-18} Amp., the concentration of ²⁰⁴Pb in STERN's sample cannot exceed 0.008%, otherwise we would have detected a signal at *m/e* 204. The results of our experiment, which are reported in Table 1, represent the averaging of the intensity measurements performed on 73 sets of peaks²⁾.

In Table 2 are reported results for two zircons (samples No. 1 and No. 2) from Col des Gets (Haute-Savoie, France), for two zircons from the Mont-Blanc Massif (samples No. 3 and No. 4) and for four rather old zircons (samples No. 5, No. 6, No. 7 and No. 8) from Greenland³⁾. The ages were calculated by using the values computed by STIEFF et al. (1959); the following physical constants were used:

$$\begin{aligned}
 \text{Decay constants} \quad & {}^{238}\text{U} = 1.53 \times 10^{-10} \text{ yr}^{-1} \\
 & {}^{235}\text{U} = 9.72 \times 10^{-10} \text{ yr}^{-1} \\
 & {}^{232}\text{Th} = 4.88 \times 10^{-11} \text{ yr}^{-1}
 \end{aligned}$$

Atomic ratios

$$\begin{aligned}
 \text{Common lead:} \quad & 204 : 206 : 207 : 208 = 1.00 : 15.95 : 15.27 : 35.34 \\
 \text{Uranium:} \quad & 235 : 238 = 1.00 : 137.7
 \end{aligned}$$

$$\begin{aligned}
 \text{Atomic masses} \quad & \text{Pb:} \quad 203.973 \quad 205.974 \quad 206.976 \quad 207.977 \\
 & \text{U:} \quad 234.041 \quad 235.044 \quad 238.051 \\
 & \text{Th:} \quad 232.038
 \end{aligned}$$

For all the zircons, Table 2 shows also the "total lead" ages which were determined in our laboratory. The lead, uranium and thorium contents were measured by X-ray fluorescence spectrometry; a detailed description of the method is given in one of our previous publications (BUCHS, 1962). In all the

²⁾ Only values which do not show a monotonous drift for increasing values of the filament temperature are taken into consideration for the isotopic ratio calculations.

³⁾ As the present work is primarily concerned with the analytical aspect of the ionization technique, no description of the samples will be given here.

Table 2. Analytical data and ages

Sample No.	Concentration ppm.			Relative atomic abundance %				% common lead	Ages in m. y.			
	U	Th	Pb	204	206	207	208		t 207/206	t 206/238	t 207/235	"total lead"
1	1423	1041	86	0.373 ±0.019	65.61 0.55	9.16 0.28	24.86 0.44	25.2 1.3	—	266 5	298 22	343
1				0.393 ±0.026	64.97 0.71	9.21 0.21	25.40 0.60	26.6 1.8	—	263 5	277 30	
2	1373	963	89	0.231 ±0.014	73.95 0.25	7.35 0.23	18.85 0.60	15.6 1.0	395 68	334 5	343 17	363
3	2292	600	114	0.085 ±0.014	83.45 0.58	5.68 0.23	10.78 0.60	5.7 0.9	352 92	303 5	309 14	319
4	2236	1131	129	0.135 ±0.013	70.52 0.62	5.96 0.29	18.39 0.65	9.1 0.9	343 109	313 5	317 27	345
5	652	123	204	0.087 ±0.017	82.91 0.24	8.85 0.20	8.15 0.27	5.9 1.2	1498 85	1695 5	1610 37	1685
6	314	53	112	0.014* ±	83.27 0.24	10.40 0.23	6.31 0.16	0.9*	2060 93	1915 5	1996 44	1900
7	497	118	169	0.018* ±	83.71 0.27	9.26 0.28	7.01 0.24	1.2*	1840 90	1825 5	1855 42	1810
8	1238	331	320	0.025 ±	82.79 0.24	8.99 0.26	8.19 0.37	1.7	1805 89	1448 5	1600 36	1414

* These values represent upper limits. No peak at mass 204 was detected

samples the isotopic composition of the lead reported in Table 2 represents the average value of two runs. For sample No. 1 the individual results of both runs are given. The error limits indicated in Table 2 are based only on statistical fluctuations. The precision of the reported values is given at 95% confidence level. The analytical error on the lead, uranium and thorium analysis by X-ray fluorescence spectrometry has not been taken into account for the calculation of the ages. For samples No. 6 and No. 7 the common lead content we report represents an upper limit, calculated on the basis of the detection limit of our instrument and the total ion current which was obtained for these measurements; no peak at m/e 204 was in fact observed with these two samples.

The age patterns are concordant except for sample No. 8 for which the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ age is significantly higher than the two U-Pb_r ages. The rather large errors which affect the $^{207}\text{Pb}_r/^{206}\text{Pb}_r$ ages for samples No. 3 ($\pm 26\%$) and No. 4 ($\pm 32\%$) are attributable to marginal ionization conditions during their analysis. As the present work was primarily performed to explore the possibility of using a modified version of KOSTOLANYI's technique for dating zircons, the validity of the ages as well as their geological implications will not be discussed here.

The high content of original common lead in samples No. 1 and No. 2 has to be compared with results reported by other authors. If generally, isotopic ratio determinations on lead extracted from zircons have shown common lead contents well below 10%, various authors have nevertheless found higher contents, ranging from about 12% up to as much as 40% (GRAUERT and ARNOLD, 1968; GRÜNENFELDER et al., 1964; GRÜNENFELDER, 1963; GOTTFRIED et al., 1959; HANSON et al., 1966).

In order to correct the isotopic compositions for common lead we have simply used the ratios given in the Chart of Nuclides (1963). These ratios are certainly too low, especially the $^{206}\text{Pb}/^{204}\text{Pb}$ and the $^{208}\text{Pb}/^{204}\text{Pb}$ ratios. Since we do not extract the lead prior to its isotopic analysis, it would have been useless to choose the isotopic ratios one finds for lead extracted from chemicals used during the separation procedure of lead from the zircons. It is therefore not possible, without other data about the isotopic composition of lead found in associated minerals, free of uranium and thorium, and having approximately the same age as the zircons studied, to make a definite choice for these ratios⁴). At any rate, if one takes into account values which are probably too high, like for instance the following set: $^{204}\text{Pb} : ^{206}\text{Pb} : ^{207}\text{Pb} : ^{208}\text{Pb} = 1 : 19 : 16 : 39$, the ages reported in Table 2 would not be drastically affected. For example, the $^{206}\text{Pb}_r/^{238}\text{U}$ age of sample No. 1 would be lowered by 2% and the $^{207}\text{Pb}_r/^{235}\text{U}$ age by 5%.

⁴) The problem of an appropriate choice for the isotopic composition of common lead is not discussed here; it is beyond the scope of this paper.

CONCLUSION

We agree that there is no direct proof that all the "contaminating lead" is eliminated during the ionization, i. e., that the common lead which is found is truly part of the "zircon lead". Nevertheless, on the basis of the results which have been obtained, especially with STERN's sample and with three of the four zircons from Greenland (samples No. 6, No. 7 and No. 8), we think that it is highly plausible that by using the method described here, the "contaminating lead" can be totally eliminated. Of course, one should be careful not to begin using readings before all the isotopic ratios remain constant at different temperatures of the filament.

It can be concluded that this method, in connection with the determination of U, Th and Pb by X-ray fluorescence spectrometry, provides a useful way to date zircons, at least when they contain more than about 80 ppm of lead. This limitation is imposed by both the necessity of obtaining ion beams with enough intensity and the limited accuracy of X-ray spectrometry for the lead content determination. However it should be stressed that it is sometimes impossible to obtain reliable results, even when the lead concentration exceeds 80 ppm. This was observed mainly with a series of zircon samples from the Mont-Blanc Massif. For about 30% of the Mont-Blanc zircons it was not possible to obtain well controlled ion beams, appearing with enough intensity so as to allow reliable age calculations to be performed. It should also be noted that the best ionization conditions (rate of heating as well as temperature of the ionization filament) change from run to run, even with the same zircon sample. No attempt has yet been made to correlate these difficulties with some physical property of the zircon sample.

The determination of lead, uranium and thorium by spark source mass spectrometry can certainly increase the value of this method. The minimum amount of sample which is presently needed with zircons containing about 100 ppm of lead, is limited by the necessity to have at least 150 to 200 mg of sample for the X-ray fluorescence analysis of U, Th and Pb. The use of spark source mass spectrometry will improve the method in regard both to accuracy and sample consumption. Some experimental work is presently undertaken with a spark source mass spectrometer.

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