On the use of a three-dimensional method in solving the U-Pb two-stage model

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(Received April 11, 1989; Accepted May 24, 1989)

The three-dimensional method is described for treating U-Pb isotopic data obeying a two-stage evolution. The U-Pb discordia plane model and the Pb-Pb isochron plane model are developed respectively, to eliminate the effect of the unknown common lead correction. Calculation formulae of best-fit plane parameters are given and some examples from the literature are recalculated to demonstrate the applicability of the method. The plane-fitting can be a convenient means in U-Pb dating.

INTRODUCTION

The decay of the naturally occurring isotopes of uranium to lead gives rise to two independent geochronometers. When the uraniferous mineral or rock sample being dated has remained closed to uranium and all of its daughters and when appropriate corrections are made for the lead incorporated into the sample at the time of its formation, the two geochronometers should give concordant dates. However, discordant ages have often been observed ever since establishment of the U-Pb dating method. Usually it is attributed to lead loss (although it can also be due to uranium gain). The episodic U-Pb models have been suggested by Wetherill (1956) and Gale and Mussett (1973) to decipher such a puzzle. Normally a suite of cogenetic samples are analysed and the results are presented on a concordia diagram where the $^{236}\text{Pb}^*/^{238}\text{U}$ ratio is plotted versus the $^{207}\text{Pb}^*/^{235}\text{U}$ ratio (Wetherill, 1956) or on a modified concordia diagram where the $^{207}\text{Pb}^*/^{206}\text{Pb}^*$ ratio is plotted versus the $^{238}\text{U}/^{206}\text{Pb}^*$ ratio (Tera and Wasserburg, 1972). In this way, a two-stage model can be solved to yield two ages t_1 and t_2 responsible for two episodic events. However, the superscript asterisk in the diagrams indicates that the

radiogenic lead component only has to be used, i.e., the common ^{206}Pb and ^{207}Pb have to be subtracted, which can be done only when the $^{206}\text{Pb}/^{204}\text{Pb}$ and $^{207}\text{Pb}/^{204}\text{Pb}$ ratios of the common lead in the sample suite are known.

A knowledge of the isotopic ratios of common lead is of crucial importance for an exact determination of the radiogenic lead dates, especially when the proportional of the common lead in the sample is large (Ludwig *et al.*, 1985). These can be obtained from other minerals which are syngenetic and contain no uranium. However, these prerequisites are not always present. To eliminate the effect of the unknown common lead correction a three-dimensional discordia plane model has been developed by Wendt (1984) based on the modified discordia equation to solve the two-stage model using the directly measured isotopic ratios $^{238}\text{U}/^{206}\text{Pb}$, $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{204}\text{Pb}/^{206}\text{Pb}$. This method assumes that all of the samples contain common lead of the same isotopic composition. Furthermore, it requires that the true U-Pb apparent ages are variably discordant. It can display all the isotopic information about concordancy and common lead, or discordancy and intersection ages t_1 and t_2 , without a correction for the common lead. Nonetheless, its derivation and calculating pro-

cedure appear to be somewhat sophisticated so that it is not easy for it to be adopted by geochronologists who do not have a good mathematical background. In order to elucidate various aspects of the three-dimensional U-Pb data presentation, another two forms of plane solution of the U-Pb two-stage model are described in the present paper in the light of the Pb-Pb isochron diagram and the concordia diagram. The present derivation seems easier to understand for a computation.

U-Pb DISCORDIA PLANE MODEL

According to the discordia equation (Wetherill, 1956) the concentrations of the radiogenic lead isotopes are linearly related to their parent isotopes in the concordia diagram:

$$\frac{{}^{206}\text{Pb}^*}{{}^{238}\text{U}} = \frac{{}^{207}\text{Pb}^*}{{}^{235}\text{U}} a_1 + a_2 \quad (1)$$

in which

$$a_1 = \frac{\exp \lambda t_1 - \exp \lambda t_2}{\exp \lambda t_1 - \exp \lambda' t_2} \quad (2)$$

$$a_2 = (\exp \lambda t_2 - 1) - a_1(\exp \lambda' t_2 - 1). \quad (3)$$

Where λ and λ' are the decay constants of ${}^{238}\text{U}$ and ${}^{235}\text{U}$, respectively. Within the framework of the two-stage model, Eq. (1) can be rewritten as:

$$\frac{{}^{206}\text{Pb}_p}{{}^{238}\text{U}} - \frac{{}^{206}\text{Pb}_i}{{}^{238}\text{U}} = a_1 \left(\frac{{}^{207}\text{Pb}_p}{{}^{235}\text{U}} - \frac{{}^{207}\text{Pb}_i}{{}^{235}\text{U}} \right) + a_2 \quad (4)$$

where the subscripts p and i denote the present-day and initial, respectively. After introducing the present-day ${}^{238}\text{U}/{}^{204}\text{Pb}$ ratio into Eq. (4) we obtain the following plane equation:

$$\left(\frac{{}^{206}\text{Pb}}{{}^{238}\text{U}} \right)_p = a_1 \left(\frac{{}^{207}\text{Pb}}{{}^{235}\text{U}} \right)_p + a_2 + a_3 \left(\frac{{}^{238}\text{U}}{{}^{204}\text{Pb}} \right)_p \quad (5)$$

in which

$$a_3 = \left(\frac{{}^{206}\text{Pb}}{{}^{204}\text{Pb}} \right)_i - 137.88 a_1 \left(\frac{{}^{207}\text{Pb}}{{}^{204}\text{Pb}} \right)_i \quad (6)$$

Eq. (5) was essentially obtained by Levchenkov and Shukolyukov (1970). The quantities $({}^{206}\text{Pb}/{}^{238}\text{U})_p$, $({}^{207}\text{Pb}/{}^{235}\text{U})_p$ and $({}^{238}\text{U}/{}^{204}\text{Pb})_p$ can be experimentally determined for a suite of

samples. These quantities can then be used to fit the parameters a_1 , a_2 and a_3 in terms of the least squares method of fitting data points to a plane (see Appendix), provided that the U-Pb isotopic data from the analysed samples have experienced the following two-stage evolution:

(1) The lead evolved in an environment with a given U/Pb ratio until a time t_1 . At time t_1 the initial system became closed and was homogeneous with respect both to the U/Pb ratio and to the isotopic composition of each element.

(2) Since time t_1 the system evolved only by the decay of uranium into lead, remaining closed and homogeneous until a time t_2 .

(3) At time t_2 a brief differentiation took place within the closed system, altering the U/Pb ratio from place to place within the system (without any isotopic fractionation of either lead or uranium) and thus dividing the grand system into a number of subsystems. Each subsystem is characterized by the quantity $({}^{238}\text{U}/{}^{204}\text{Pb})_p$.

(4) From t_2 to the present, evolution again proceeds only by the decay of uranium, each subsystem remaining homogeneous and closed within itself, at least as far as uranium and lead are concerned. The specimens selected for analysis will, for this strictly two-stage evolution, be a direct sample of the whole set of subsystems.

In this context, the uranium has been differentiated two times relative to the lead in their evolutionary history. But the most important point is that isotopic signatures of the initial (common) lead and the radiogenic lead from t_1 to t_2 have not been obliterated after the two episodic events in the analysed specimens.

When the numerical values of a_1 and a_2 are known, one can find t_2 at first through Eq. (3) and then t_1 through Eq. (2). In this way, both t_1 and t_2 can be simultaneously obtained from the experimentally measured U-Pb isotopic data without a correction for the common lead. The goodness of fit of data points to the plane equation can be expressed as polycorrelation coefficient R (see Appendix), which can serve as

Fig. 1. U-Pb discordia plane diagram. Line AB is in the $^{206}\text{Pb}/^{238}\text{U}$ versus $^{207}\text{Pb}/^{235}\text{U}$ plane.

a check of the assumptions that all of the specimens have evolved in the two-stage model and remained closed to uranium and lead. Graphically a diagram of $(^{206}\text{Pb}/^{238}\text{U})_p - a_3/(^{238}\text{U}/^{204}\text{Pb})_p$ versus $(^{207}\text{Pb}/^{235}\text{U})_p$ can be plotted to display the correlation of the data points.

Geometrically speaking, as shown in Fig. 1, the transformation of Eq. (1) into Eq. (5) is equivalent to constructing a discordia plane ABCD that cuts the $^{206}\text{Pb}/^{238}\text{U}$ versus $^{207}\text{Pb}/^{235}\text{U}$ plane along a straight line AB whose equation corresponds to Wetherill's discordia equation. The condition $1/(^{238}\text{U}/^{204}\text{Pb})=0$ implies that the specimens did not contain the common lead.

Pb-Pb ISOCHRON PLANE MODEL

Physically the usual form of the two-stage model equation has been given by Gale and Mussett (1973):

$$\left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}}\right)_p = \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}}\right)_i + \left(\frac{^{238}\text{Pb}}{^{204}\text{Pb}}\right)_2 (\exp \lambda t_1 - \exp \lambda t_2) + \left(\frac{^{238}\text{U}}{^{204}\text{Pb}}\right)_3 (\exp \lambda t_2 - 1) \quad (7a)$$

$$\left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}}\right)_p = \left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}}\right)_i + \frac{1}{137.88} \left(\frac{^{238}\text{U}}{^{204}\text{Pb}}\right)_2$$

$$\times (\exp \lambda' t_1 - \exp \lambda' t_2) + \frac{1}{137.88} \left(\frac{^{238}\text{U}}{^{204}\text{Pb}}\right)_3 (\exp \lambda' t_2 - 1) \quad (7b)$$

where the subscripts 2 and 3 denote the time t_2 and the present-day, respectively. In order to preserve consistency with the U-Pb discordia plane model in numbering the stages of episodic U-Pb evolution, it is taken in this section that Eq. (7) still refers to the two-stage model because it involves only two episodic differentiations of uranium relative to lead. Actually it corresponds to a three-stage model equation described by Gale and Mussett (1973). In this regard a further discussion has been given by Chu (1975).

An expression for $(^{238}\text{U}/^{204}\text{Pb})_2$ can be derived from Eq. (7a) and then substituted into Eq. (7b). We thus obtain another form of plane equation:

$$\left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}}\right)_p = b_1 \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}}\right)_p + b_2 \left(\frac{^{238}\text{Pb}}{^{204}\text{Pb}}\right)_p + b_3 \quad (8)$$

in which

$$b_1 = \frac{\exp \lambda' t_1 - \exp \lambda' t_2}{137.88 (\exp \lambda t_1 - \exp \lambda t_2)} \quad (9)$$

$$b_2 = \frac{1}{137.88} (\exp \lambda' t_2 - 1) - b_1 (\exp \lambda t_2 - 1) \quad (10)$$

Fig. 2. Pb-Pb isochron plane diagram. Line A'B' is in the $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ plane.

$$b_3 = \left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}} \right)_i - b_1 \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}} \right)_i \quad (11)$$

A similar form to Eq. (8) was derived by Neymark and Levchenkov (1979), but there is an incorrect expression for the term b_3 in their Eq. (3).

Within the framework of this model, as depicted in Fig. 2, a suite of cogenetic samples will lie on the plane A'B'C'D' which cuts the $^{207}\text{Pb}/^{204}\text{Pb}$ versus $^{206}\text{Pb}/^{204}\text{Pb}$ plane along a straight line A'B' which intersects the Pb-Pb growth curve at t_1 and t_2 , respectively. The condition $^{238}\text{U}/^{204}\text{Pb} = 0$ means that there would not exist measurable uranium in the analysed samples.

Because the quantities $(^{207}\text{Pb}/^{204}\text{Pb})_p$, $(^{206}\text{Pb}/^{204}\text{Pb})_p$ and $(^{238}\text{U}/^{204}\text{Pb})_p$ can be directly measured, least squares fitting applied to Eq. (8) yields b_1 , b_2 and b_3 from which t_1 and t_2 can be derived without a correction for common lead. Not only can the polycorrelation coefficient R serve as a check of the assumptions; graphically the goodness of fit of the data points can be shown on a diagram of $(^{207}\text{Pb}/^{204}\text{Pb})_p - b_2(^{238}\text{U}/^{204}\text{Pb})_p$ versus $(^{206}\text{Pb}/^{204}\text{Pb})_p$ (Chu, 1975).

A modified U-Pb discordia plane model can be obtained by dividing Eq. (8) by $(^{206}\text{Pb}/^{204}\text{Pb})_p$, giving the following form:

$$\left(\frac{^{207}\text{Pb}}{^{206}\text{Pb}} \right)_p = b_1 + b_2 \left(\frac{^{238}\text{U}}{^{206}\text{Pb}} \right)_p + b_3 \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}} \right)_p \quad (12)$$

which was derived by Wendt (1984) in another way. Geometrically speaking, this operation is equivalent to constructing the modified discordia plane which cuts the $^{207}\text{Pb}/^{206}\text{Pb}$ versus $^{238}\text{U}/^{206}\text{Pb}$ plane along a straight line whose equation corresponds to the modified discordia equation of Tera and Wasserburg (1972). The condition $1/(^{206}\text{Pb}/^{204}\text{Pb}) = 0$ indicates that the samples did not contain the common lead.

EXAMPLES AND CONCLUSIONS

Purely as a way of illustrating the three-dimensional method, a computer programme has been used with data published in the literature for seven suites of samples. These include zircons from the pre-Permian basements of the Bohemian Massif (Grauert *et al.*, 1973), from the Precambrian Fay-de-Bretagne eclogite in the Hercynian belt of South Brittany (Paquette *et al.*, 1985), from acidic porphyritic volcanites of Mareuil-sur-Lay Formation in the Armorican Massif (Peucat *et al.*, 1986) and from Hushan granite of the Taishan complex, China (Jahn *et al.*, 1988), and uranium minerals such as primary pitchblende from Ace Mine, Saskatchewan (Russell and Ahrens, 1957) and secondary torbernite from the uranium mineralization in the basement rocks at Grossschloppen, NE Bavaria (Carl and Dill, 1985), as well as a meteorite (Unruh *et al.*, 1979). As shown in

Table 1. Some examples using the three-dimensional method

Occurrence	Mineral	Number of sample	U-Pb discordia plane model		Pb-Pb isochron plane model		Modified U-Pb discordia plane model		Usual concordia diagram		References
			t_2 (Ma)	t_1 (Ma)	t_2 (Ma)	t_1 (Ma)	t_2 (Ma)	t_1 (Ma)	t_2 (Ma)	t_1 (Ma)	
Bohemian Massif	zircon	18	564	2263	562	2254	565	2251	560	2260	Grauert <i>et al.</i> (1973)
Brittany	zircon	6	410	2169	401	2113	406	2148	413	2208	Paquette <i>et al.</i> (1985)
Armorican Massif	zircon	6	397	2108	385	2121	396	2083	392	2103	Peucat <i>et al.</i> (1986)
Taishan	zircon	10	347	2560	327	2556	344	2560	334	2560	Jahn <i>et al.</i> (1988)
Ace Mine	pitchblende	5	252	1781	163	1813	231	1764	190	1800	Russell and Ahrens (1957)
Bavaria	torbernite	8	6.4	244	5.7	218	5.9	225	5.2	205	Carl and Dill (1985)
Barwell	chondrite	10	-53	4530	-91	4549	-176	4541	-170	4536	Unruh <i>et al.</i> (1979)

Table 1, the age values obtained applying the present methods agree well with the results given by the concordia diagram. Therefore, the three-dimensional U-Pb method can yield the t_1 and t_2 ages in the case of a two-stage evolution history without any assumption about the isotopic composition of the common lead present in the U-Pb system of the samples being investigated.

The U-Pb discordia plane model and the Pb-Pb isochron plane model described in the present paper are not independent of each other. Theoretically a suite of cogenetic samples will yield identical results for the three types of 3-dimensional solution if the mathematical treatments are correct and the sample system being investigated did experience the two-stage evolution. In this context there is no reason to prefer one or the other type of presentation. In setting about the task of interpreting a given set of U-Pb data one can nevertheless treat the U-Pb data by the three 3-dimensional models together to check all of the required isotopic information, e.g. not only the ages of the two episodic events, but also the "real" isotope ratios of the initial (common) lead through Eq. (6) or (11). Furthermore, the Pb-Pb isochron plane model can possibly treat a suite of samples with relatively high amounts of initial lead, or with very low uranium content, which makes it impossible to use the usual concordia diagram approach.

Acknowledgments—I wish to thank Profs. Yu-Liang Xia and Quan Tao for introducing me to the basic principles of U-Pb multistage models. I am indebted to Profs. N. H. Gale and C. J. Hawkesworth for their constructive comments.

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APPENDIX

Least Squares Fitting of Plane Equation

For a plane equation:

$$y = ax + cz + b$$

the simplest method of fitting data points is the least squares regression procedure, which consists of minimizing the sum of the squares of the difference between the observed and calculated values, i.e.

$$\sum (y_{\text{obs}} - y_{\text{calc}})^2 = \text{minimum.}$$

Given the constants a , b , c , we can for any given point (x, z) calculate:

$$y_{\text{calc}} = ax + cz + b$$

and the deviation between y_{obs} and y_{calc} is given by

$$\text{deviation} = y_{\text{obs}} - ax - cz - b.$$

If the sum of the squared deviations is given by $F(a, b, c)$, then

$$F(a, b, c) = \sum (y_{\text{obs}} - ax - cz - b)^2.$$

If $F(a, b, c)$ is to be minimized, then the partial differentials should equal zero:

$$\frac{\partial F}{\partial a} = \frac{\partial F}{\partial b} = \frac{\partial F}{\partial c} = 0$$

using the values of y_{obs} , the partial derivations are

$$\frac{\partial F}{\partial a} = \sum 2(y - ax - cz - b)(-x) = 0$$

$$\frac{\partial F}{\partial b} = \sum 2(y - ax - cz - b)(-1) = 0$$

$$\frac{\partial F}{\partial c} = \sum 2(y - ax - cz - b)(-z) = 0$$

which, on multiplying out and summing the individual terms, gives

$$a \sum x^2 + c \sum xz + b \sum x = \sum xy$$

$$a \sum x + c \sum z + nb = \sum y$$

$$a \sum xz + c \sum z^2 + b \sum z = \sum yz$$

where n = the number of data points and the summation signs imply summing from 1 to n . The equations for calculating the parameters (a , b and c) of the best plane are:

$$a = \frac{W_{xy}W_{z^2} - W_{yz}W_{xz}}{W_{x^2}W_{z^2} - W_{xz}W_{xz}}$$

$$c = \frac{W_{yz}W_{x^2} - W_{xy}W_{xz}}{W_{x^2}W_{z^2} - W_{xz}W_{xz}}$$

$$b = \bar{y} - a\bar{x} - c\bar{z}$$

in which

$$\bar{x} = \frac{1}{n} \sum x, \bar{y} = \frac{1}{n} \sum y, \bar{z} = \frac{1}{n} \sum z$$

$$W_{x^2} = \sum (x - \bar{x})^2, W_{z^2} = \sum (z - \bar{z})^2$$

$$W_{xy} = \sum (x - \bar{x})(y - \bar{y})$$

$$W_{xz} = \sum (x - \bar{x})(z - \bar{z})$$

$$W_{yz} = \sum (y - \bar{y})(z - \bar{z}).$$

The polycorrelation coefficient is given by

$$R = \sqrt{1 - \frac{F}{\sum (y - \bar{y})^2}} \quad (0 \leq R \leq 1)$$

A pocket computer is sufficient to perform

the planar regression. The more exact method of calculating the parameters of the best plane must take into account the known analytical errors of the coordinates of the data points on the plane diagram. This can be accomplished by the use of weighting residuals, which has been given by Wendt (1984).