A further three-dimensional U-Pb method for solving the two-stage episodic model

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As a continuum of the preceding studies on the three-dimensional U-Pb method (Zheng, 1989), a further U-Pb discordia plane model is developed by taking the measured isotope ratios 235U/207Pb, 206Pb/207Pb and 204Pb/207Pb for X, Y and Z axis respectively. Its geometry and applicability are elucidated in order to show the advantage of the three-dimensional U-Pb isotopic data presentation.

INTRODUCTION

So far three types of three-dimensional U-Pb method have been established: (1) Pb-Pb isochron plane (Chu, 1975; Neymark and Levchenkov, 1979), (2) Wetherill U-Pb discordia plane (Levchenkov and Shukolyukov, 1970), and (3) Tera-Wasserburg U-Pb discordia plane (Wendt, 1984 and 1989). Their uses enable us to solve the U-Pb two-stage episodic model and to obtain a unique solution to two ages t1 and t2 responsible for two geological events without any assumption about the isotopic composition of initial (common) lead for correction (Zheng, 1989). An application of the Pb-Pb isochron plane model to the whole-rock U-Pb dating has been presented by Zheng (1990) for the high-grade metamorphic rocks from the Lewisian basement in Northwest Scotland. Geometrically, a further three-dimensional U-Pb discordia plane method can be introduced by taking the measured isotope ratios 235U/207Pb, 206Pb/207Pb and 204Pb/207Pb for X, Y and Z axis, respectively. Correspondingly, the third kind of concordia diagram can be drawn to show the relationship between discordia line and concordia curve. Within the framework of the U-Pb two-stage evolution, some constraints can be put on correction of common lead in conventional discordia dating and on U/Pb differentiation factors at the two episodic events.

DERIVATION

Physically the U-Pb two-stage model equation has the following form (e.g., Gale and Mussett, 1973):

\[
\begin{align*}
\frac{206\text{Pb}}{204\text{Pb}}_3 &= \frac{206\text{Pb}}{204\text{Pb}}_1 + \left(\frac{238\text{U}}{204\text{Pb}}_3\right) (\exp \lambda t_1 - \exp \lambda t_2) \\
&\quad + \left(\frac{235\text{U}}{204\text{Pb}}_3\right) (\exp \lambda t_2 - 1) \quad (1A) \\
\frac{207\text{Pb}}{204\text{Pb}}_3 &= \frac{207\text{Pb}}{204\text{Pb}}_1 + \left(\frac{235\text{U}}{204\text{Pb}}_3\right) (\exp \lambda' t_1 - \exp \lambda' t_2) \\
&\quad + \left(\frac{235\text{U}}{204\text{Pb}}_3\right) (\exp \lambda' t_2 - 1), \quad (1B)
\end{align*}
\]

where subscript 1 denotes the initial system prior to time t1, and 2 and 3 denote time t2 and the present day respectively; \(\lambda\) and \(\lambda'\) are the decay constants of 238U and 235U, respectively.

An expression for \(\frac{235\text{U}}{204\text{Pb}}_3\) can be derived from Eq. (1B), then substituting it into Eq. (1A), we obtain another form of Pb–Pb isochron plane equation:

\[
\begin{align*}
\frac{206\text{Pb}}{204\text{Pb}}_3 &= a \left(\frac{235\text{U}}{204\text{Pb}}_3\right) + b \left(\frac{207\text{Pb}}{204\text{Pb}}_3\right) + c. \quad (2)
\end{align*}
\]
Let Eq. (2) be divided by \((^{207}\text{Pb}/^{204}\text{Pb})_3\) and thus the third type of U-Pb discordia plane equation is yielded:

\[
\frac{^{206}\text{Pb}}{^{207}\text{Pb}} = a \cdot \frac{^{235}\text{U}}{^{207}\text{Pb}} + b + c \cdot \frac{^{204}\text{Pb}}{^{207}\text{Pb}}
\]

with

\[
a = 137.88(\exp \lambda t_2 - 1) - b(\exp \lambda' t_2 - 1)
\]

\[
b = \frac{137.88(\exp \lambda t_1 - \exp \lambda t_2)}{\exp \lambda' t_1 - \exp \lambda' t_2}
\]

\[
c = \left(\frac{^{206}\text{Pb}}{^{204}\text{Pb}}\right)_1 - b\left(\frac{^{207}\text{Pb}}{^{204}\text{Pb}}\right)_1
\]

where the subscript 3 is omitted for simplification because it refers to the isotopic ratios measured at the present day; \(^{238}\text{U}/^{235}\text{U}\) ratio is taken to be 137.88 after Steiger and Jäger (1977).

The quantities \(^{235}\text{U}/^{207}\text{Pb}\), \(^{206}\text{Pb}/^{207}\text{Pb}\) and \(^{204}\text{Pb}/^{207}\text{Pb}\) can be experimentally determined for a suite of cogenetic samples. These quantities can then be used to fit the plane parameters \(a\), \(b\) and \(c\) in terms of the least squares procedures (e.g., Wendt, 1984; Zheng, 1989). When the numerical values of \(a\) and \(b\) are known, we can find \(t_2\) and \(t_1\) through Eq. (4) and (5). In this way, both \(t_1\) and \(t_2\) can be simultaneously obtained from the experimentally measured U–Pb isotope data without any assumption about the isotopic composition of initial (common) lead for correction. The fitness of data points to the discordia plane equation can be graphically checked in a diagram of \((^{206}\text{Pb}/^{207}\text{Pb}) - c(^{204}\text{Pb}/^{207}\text{Pb})\) versus \((^{235}\text{U}/^{207}\text{Pb})\). The degree of coplanarity of the data points can be expressed by the polycorrelation coefficient defined by Zheng (1989). This is a good criterion to judge whether or not a fitted plane is likely to yield significant information on the ages.

**Geometric Analysis**

Within the framework of the third U–Pb discordia plane model, as shown in Fig. 1, the transformation of Eq. (1) into Eq. (3) is equivalent to constructing a discordia plane ABCD which cuts the \(^{206}\text{Pb}/^{207}\text{Pb}\) versus \(^{235}\text{U}/^{207}\text{Pb}\) plane along a straight line AB whose equation corresponds to the third kind of discordia:

\[
\frac{^{206}\text{Pb}}{^{207}\text{Pb}} = a \cdot \frac{^{235}\text{U}}{^{207}\text{Pb}} + b,
\]

\[
Y = \frac{^{206}\text{Pb}}{^{207}\text{Pb}}
\]

\[
X = \frac{^{235}\text{U}}{^{207}\text{Pb}}
\]

\[
Z = \frac{^{204}\text{Pb}}{^{207}\text{Pb}}
\]

*Fig. 1. A further three-dimensional U–Pb discordia plane diagram. Line AB is in the \(^{206}\text{Pb}/^{207}\text{Pb}\) vs. \(^{235}\text{U}/^{207}\text{Pb}\) plane.*
Three-dimensional U-Pb method

where superscript * denotes the radiogenic lead corrected for the initial lead prior to time \( t_1 \). Thus a further concordia curve is defined by

\[
x = \frac{1}{\exp \lambda' t - 1}
\]

\[
y = \frac{137.88(\exp \lambda t - 1)}{\exp \lambda' t - 1}.
\]

The curve starts from \( x = \infty \) and \( y = 137.88 \times \frac{\lambda}{\lambda'} = 21.718 \) for \( t = 0 \), and both \( x \) and \( y \) decrease with increasing \( t \), opposite to the Wetherill concordia curve. Apparently, the third discordia line also intersects the concordia curve at \( t_1 \) and \( t_2 \) respectively, as depicted in Fig. 2. Any discordant data point on the discordia line can be regarded as a mixture of the concordant U–Pb system of age \( t_2 \) (point \( t_2 \) in Fig. 2) and a certain amount of “inherited” radiogenic lead produced between \( t_1 \) and \( t_2 \) (point \( b \) in Fig. 2). If a sample has subjected itself to lead loss (or uranium gain) at time \( t_2 \), the data point lies between \( b \) and \( t_1 \); if excess \( ^{206}\text{Pb} \) was produced by the decay of \( ^{230}\text{Th} \) (with a half-life of 75200 yr) or if the radiogenic lead was lost after \( t_2 \) due to incipient weathering, the data point lies on the discordia line above concordia curve at time \( t_2 \) with the \( ^{206}\text{Pb} / ^{207}\text{Pb} \) ratio greater than 21.718.

The projection of the discordia plane in \( xyz \)-space on \( xy \)-plane shows the data points located inside the triangle \( t_2, b \) and \( O \), and yields the discordia line defined by Eq. (7) after corrected for the initial lead. In the case of a concordant age for a suite of cogenetic samples, the data points uncorrected for the initial lead are linearly distributed in the triangle \( t_2, b \) and \( O \) to yield a primary discordia (isochron):

\[
\frac{^{206}\text{Pb}}{^{207}\text{Pb}} = a' \cdot \frac{^{235}\text{U}}{^{207}\text{Pb}} + b' \tag{10}
\]

with parameters:

\[
a' = 137.88(\exp \lambda t - 1) - b' (\exp \lambda' t - 1) \tag{11}
\]

**Fig. 2.** The projections of the three-dimensional U-Pb discordia plane in \( xyz \)-space onto \( xy \)-plane and \( yz \)-plane, respectively.
where subscript $i$ denotes the initial system prior to time $t$. The primary discordia intersects the third concordia curve only one time with the concordant age. The intercept $b'$ with $y$-axis yields the $^{206}\text{Pb}/^{207}\text{Pb}$ ratio of the initial lead. In this respect, Eq. (10) is equivalent to the representation first reported by Tatsumoto et al. (1972) in plotting $^{206}\text{Pb}/^{207}\text{Pb}$ versus $^{238}\text{U}/^{207}\text{Pb}$ for the Apollo 14 lunar samples. Unfortunately, the authors mistreated the “primary” discordia as an isochron for their isotope data on density fractions of lunar sample 14310 while a Wetherill U-Pb concordia diagram was used by themselves to indicate a two-stage evolution for the lunar sample. A reappraisal of their U-Pb isotope data using the present three-dimensional discordia plane method yields a unique solution of $t_1=4550\pm 80$ Ma and $t_2=3700\pm 50$ Ma, corroborating the existence of the two-stage history for the sample 14310. However, because the third concordia curve is nearly linear in pre-Archean time, the resulting discordia chord is nearly parallel to the concordia curve so that the recognition of discordance was hampered for the lunar sample.

The projection of the new discordia plane onto the $yz$-plane lies inside the triangle $t_2$, $b$ and $d$, and yields a lead-line for uncorrected isotopic ratios in the case of the two-stage model:

$$\frac{^{206}\text{Pb}}{^{207}\text{Pb}} = b + c \cdot \frac{^{204}\text{Pb}}{^{207}\text{Pb}} \quad (13)$$

which connects the initial lead point $d$ with the radiogenic lead $b$ produced between $t_1$ and $t_2$ (Fig. 2). Within the framework of the U-Pb two-stage evolution, recent uranium loss only induces poorly fitted U-Pb isochrons which frequently give spuriously old ages, whereas ancient uranium loss would have introduced a scatter into the lead-line. So would do ancient or recent gain of lead. In this context, when the lead-line can be treated as a Pb-Pb isochron with an age which is in agreement with the $t_1$ age of the discordia method within the error limits, uranium mobility can be suggested to occur at time $t_2$ without alteration to the Pb isotopic composition. If the lead loss took place due to metamorphism at time $t_2$, or if there exists inherited radiogenic lead in samples (e.g., zircons from young granitoids), the intercept $h$ of the lead-line with the $y$-axis will lie between $b$ and $t_2$ and a secondary Pb-Pb isochron could be yielded for the involved systems. A true Pb-Pb isochron will exist if the investigated system has a concordant age or if the system is only fractionated recently with $t_2=0$. A geological event (e.g., metamorphism, partial melting etc.) before Mesozoic would fractionate the U-Pb system so that the Pb-Pb isochron would give an age greater than the true $t_1$ age.

**IMPLICATIONS FOR DISORDIA DATING AND THE NATURE OF INITIAL LEAD**

Because the third concordia diagram is essentially a simple transformation of the concordia diagram of Wetherill (1956) or Tera and Wasserburg (1972), a given set of samples will yield identical age results within the analytical errors, provided that the investigated U-Pb isotope system did undergo a strictly two-stage evolution starting at time $t_1$ and having the same type of initial lead (i.e. following the linear relationship between $^{207}\text{Pb}/^{206}\text{Pb}$ and $^{206}\text{Pb}/^{207}\text{Pb}$). Having the same advantage as the Tera-Wasserburg concordia diagram (Wendt, 1989), in the newly introduced concordia diagram the error in $x$ and $y$ directions are essentially uncorrelated, which makes their mathematical treatment much easier than the Wetherill concordia diagram when fitting a chord or a plane through the discordia data points. The third concordia curve is also expanded relative to the Wetherill concordia at young time, so that a stronger curvature of the concordia curve can be obtained to show the intersection relationship between discordia line and concordia curve, as illustrated in Fig. 3 for young torbernite samples from the uranium mineralization in the basement rocks at Grosschloppen, NE Bavaria, Germany.

Comparing with the Tera-Wasserburg con-
Fig. 3. Modified U–Pb concordia diagram for eight torbernite samples from the uranium mineralization in the basement rocks at Grosschloppen, NE Bavaria (data from Carl and Dill, 1985). The present results are identical to those obtained from the Tera-Wasserburg concordia diagram by the original authors and from the other three 3-dimensional U-Pb methods by Zheng (1989).

Fig. 4. Modified U–Pb concordia diagram for nine whole-rock samples from the Gortdrum deposit in Ireland (data from Duane et al., 1986). The present result ($t = 357 \pm 23$ Ma) is well compatible with the Pb–Pb isochron age of $359 \pm 26$ Ma and the Wetherill concordia upper intersection age of $340 \pm 25$ Ma given by the original authors.
cordia diagram, the present one is just an "upside down" presentation of the former. The new discordia has a positive slope rather than the negative slope obtained by Tera and Wasserburg (1972) if \( t_2 \) value is greater than zero. When \( t_2 \) value calculated is less than zero, which can be caused by incipient weathering due to the effect of initial radioactive-daughter disequilibrium on the U–Pb system (Ludwig, 1977; Wendt and Carl, 1985), a discordia with a negative slope can be yielded on the third concordia diagram and its intersection with the concordia curve yields the true age. This is illustrated in Fig. 4 for whole-rock samples from the Gortdrum deposit in Ireland.

From Eq. (1) it can be seen that in conventional discordia dating the experimentally measured Pb isotope ratios have to be corrected for the initial lead responsible for that prior to time \( t_1 \) rather than \( t_2 \). Unfortunately, this has been overlooked in dealing with the inheritance of radiogenic lead in zircons from young granitoids by reverse discordia defined by Gulson and Rutishauser (1976). Apparently, the correction for common lead at time \( t_2 \) is incorrect in the dating concept of discordia method, although the miscorrection does not cause large uncertainties in calculating the ages of discordant samples which have high \( ^{206}\text{Pb}/^{204}\text{Pb} \) ratios (e.g., > 1000).

The three-dimensional U–Pb method has a potential application to the restricted three-stage model of Ulrych (1969). If a given set of starting parameters is assigned to the three-stage evolution, e.g., either the meteoritic model given by Tatsumoto et al. (1973) or the plumbotectonic model proposed by Zartman and Haines (1988), a \( \mu_1(=^{238}\text{U}/^{204}\text{Pb}) \) value can be calculated for the time interval from \( t_0 \) to \( t_1 \):

\[
\mu_1 = \frac{c - \alpha_0 + b \beta_0}{(\exp \lambda t_0 - \exp \lambda t_1) - b(\exp \lambda' t_0 - \exp \lambda' t_1)}/137.88,
\]

given that \( \alpha_0, \beta_0 \) and \( t_0 \) are constants with the definitions: \( \alpha = ^{206}\text{Pb}/^{204}\text{Pb} \) and \( \beta = ^{207}\text{Pb}/^{204}\text{Pb} \) as well as \( \alpha_1 = \alpha_0 + \mu_1(\exp \lambda t_0 - \exp \lambda t_1) \) and \( \beta_1 = \beta_0 + \mu_1(\exp \lambda' t_0 - \exp \lambda' t_1)/137.88 \) (where subscript \( o \) denotes the primordial source). The \( \mu_1 \) value can be used to infer the nature of the source region of initial lead incorporated at time \( t_1 \) into the investigated samples. Then U–Pb differentiation factors at the two episodic events can be evaluated quantitatively through \( f_1 = \mu_1/\mu_2 \) and \( f_2 = \mu_2/\mu_3 \).

**APPLICATION TO SAMPLES WITH HIGH COMMON LEAD**

The great merit of the three-dimensional U–Pb method is that it can treat a suite of co-genetic samples with relatively high amounts of initial (common) lead, which make it impossible to use either the usual concordia diagram approach due to the critical importance of correction for common lead incorporated at time \( t_1 \), or a straightforward U–Pb isochron approach that assumes either negligible or isotopically constant initial lead incorporated into the samples at the formation time. This can be illustrated by the U–Pb system in the Schwartzwalder hydrothermal uranium deposit, Colorado. The U–Pb isotopic analyses of uranium ore samples from this deposit showed high amounts of initial (common) lead with the total Pb isotope ratios of \( ^{206}\text{Pb}/^{204}\text{Pb} = 30.078 - 49.647 \) and \( ^{207}\text{Pb}/^{204}\text{Pb} = 16.631 - 17.437 \). The initial lead at the time of uranium mineralization was both variable and relatively radiogenic in its isotope ratios \( ^{206}\text{Pb}/^{204}\text{Pb} = 26-30 \). Using a sophisticated iteration procedure Ludwig et al. (1985) obtained concordant U–Pb isochron ages of 69.3 ± 1.1 Ma \( ^{206}\text{Pb}/^{204}\text{Pb} \) versus \( ^{238}\text{U}/^{204}\text{Pb} \) and 69.9 ± 2.3 Ma \( ^{207}\text{Pb}/^{204}\text{Pb} \) versus \( ^{235}\text{U}/^{204}\text{Pb} \) for timing of the uranium mineralization. After subtracting the radiogenic lead produced from the 69.3 Ma to the present day for the uranium ore samples, the initial Pb isotope systematics at \( t = 69.3 \) Ma defined a secondary Pb–Pb isochron with a \( t_1 \) age of 1730±130 Ma, which was interpreted by the authors to repre-
Fig. 5. Modified U-Pb concordia diagram for 25 whole-rock samples from the Schwartzwalder uranium deposit in Colorado (data from Ludwig et al., 1985).

sent the age of the source of the metals in the Schwartzwalder deposit. With the present three-dimensional U-Pb discordia plane method, a reevaluation is made on the U-Pb isotopic data for 25 ore samples utilized by the original authors and the identical results are obtained with $t_2 = 69 \pm 2$ Ma and $t_1 = 1800 \pm 100$ Ma, as shown in Fig. 5. Furthermore, within the framework of the restricted U-Pb three-stage evolution, it is calculated for the uranium ores that $\mu_1$ value is 7.73 for the time interval from 4570 to 1800 Ma, and that $\mu_2$ values are 35–50 for the time interval from 1800 to 69 Ma, given that the meteoritic lead is taken for the starting parameters (i.e., $a_0 = 9.307$, $b_0 = 10.294$, and $t_0 = 4570$ Ma; after Tatsumoto et al., 1973). The relatively high $\mu_2$ values indicate a modest enrichment of uranium relative to lead at the regional metamorphic time of $t_1 = 1800$ Ma, as previously recognized by Ludwig et al. (1985). The obtained $f_1$ values are 0.15–0.21 and $f_2$ values are 0.02–0.89, indicating that uranium was further markedly enriched relative to lead at the mineralization time $t_2$ with respect to the metamorphic time $t_1$. In formal terms, if $f_1 < 1$, uranium gain or lead loss are both admissible (Gale and Mussett, 1973). Conversely, $f_1 > 1$ implies that uranium is depleted relative to lead at event $t_1$. This has been observed for high-grade (e.g., granulite facies) metamorphism of a whole-rock system in the Lewisian basement (Moorbath et al., 1969; Zheng, 1990).

**CONCLUSIONS**

A third three-dimensional U-Pb discordia plane method was developed and its geometric natures and applicabilities were illustrated. Although the newly introduced method does not give new age results as to those derived previous-
ly by either two- or three-dimensional approaches, it contributes a supplement to the three-dimensional U-Pb data presentation. Apparently, all the four forms of the three-dimensional U-Pb method established so far can be unified within the framework of a U-Pb two-stage episodic evolution. Using experimentally determined U-Pb isotopic ratios one can not only obtain a unique solution to two ages \( t_1 \) and \( t_2 \) responsible for two episodic U/Pb differentiation events without any assumption about the isotopic composition of common lead incorporated, but also shed some light on the feature of the source region of the initial lead incorporated at time \( t_1 \) and on the U/Pb differentiation factors at the two episodic events. The common lead for correction in conventional discordia dating is clarified to be responsible for the initial lead prior to time \( t_1 \) rather than \( t_2 \). Any addition of common lead to the samples at time \( t_2 \) would cause alteration of lead isotopic ratios owing to simple mixing of leads that have been evolved in different \( \mu \) environments for the preceding stage. This would violate the U-Pb two-stage evolution described by Eq. (1). It may nevertheless occur in nature.

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