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AGE AND ISOTOPE STUDIES IN ECONOMIC GEOLOGY

by

J.R. Richards, R.W. Page, L.P. Black

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INTRODUCTION

In the decade since Richards and White (1965) reviewed the use of isotopic dating in ore search, the number of Australian isotope groups has increased fourfold. World-wide, there has been marked progress in the isotopic explanation of known ore occurrences. By contrast there is just as much difficulty now as then in applying isotopic data to the prediction of mineralized areas.

The former review treated only the stratigraphic aspects of isotope work. The correct timing of a mineralizing event within a regional sequence will always be of interest. Often this must be by inference, but sometimes a mineral clearly related to an ore-forming episode will permit a direct dating; examples are a uranium-rich mineral such as pitchblende, mica in a hydrothermal vein, or a potassium silicate alteration envelope in a porphyry copper environment. The methods have often been described (e.g., Joplin et al., 1972, York & Farquhar, 1972; Doe, 1970; Faure & Powell, 1972; Dalrymple & Lanphere, 1969). It should be sufficient here to illustrate some of the main points with examples.

Time is, however, but part of the story. Almost as a by-product, the isochron method also reveals the isotopic make-up of the Pb and Sr which were present as the rock formed - information which bears on both the origin of the rocks and their possible relationship to neighbouring ore. Although comparatively new to ore studies, this approach will become an essential part of all future investigations.

Further, the light elements (S, O, C, H) are not related to radioactive decay, but do suffer isotopic variations which depend not on time, but on those physical parameters such as temperature and atomic weight which affect rates of chemical reaction, of ion or molecule migration, or of exchange across phase boundaries. Their relevance to ore studies is also shown, although local examples are as yet few.

Selected age and isotopic studies from Australia and Papua New Guinea have been quoted as examples; the list is deliberately incomplete. Comprehensive collections of available dates have been prepared by White (1962), Harding (1969), Bennett & Page (1973) and Thomas (1973). No complete list of purely isotopic studies is known to the authors, although Stanton (1972) and Smith (1974) give good introductions to work on sulphur.

CONSEQUENCES OF RADIOACTIVE DECAY

DATING

"Mineral ages": The individual sample

Joplin and others (1972) give a simple summary of the theory of how ages are deduced from the radioactive decay of an unstable parent isotope to a more stable isotope of a different element (daughter).

Table 1 summarizes the parent-daughter relationships which are of major interest for dating rocks. Two isotopes of rubidium occur naturally in trace amounts in most rocks. That of mass 85 is stable; that of mass 87 steadily decays to ^{87}Sr, one of the four isotopes of natural strontium, at a rate described by the Radioactive Decay Law. Allowing for a slight uncertainty in the proportionality (decay) constant, the observed $^{87}Sr/^{87}Rb$ in a sample should be exactly related to the time required for generation of the observed ^{87}Sr from the parent rubidium. This time is the age of the sample (rock or mineral) provided that (i) none of the observed strontium was present at the beginning, and that (ii) there had been neither gain nor loss of either rubidium or strontium during the whole period ("closed system" requirement). Similar statements apply to the other decay systems listed in Table 1.

If both conditions were true, measurement of the concentration ratio (daughter/parent) in a mineral concentrate or crushed whole-rock sample should yield one independent date. However, almost every sample contains some initial Sr and Pb; initial Ar is usually unimportant, but adsorbed atmospheric argon interferes. Recognition is simple; the mass spectra of the extracted Sr, Pb, or Ar reveal isotopes which do not result from radioactive decay. Correction is also easy, provided we know or can estimate the isotopic makeup of the contaminant. Thus ^{36}Ar indicates an air component in the measured argon; correction of the ^{40}Ar peak is derived from the known air ratio, $^{40}\mathrm{Ar}/^{36}\mathrm{Ar} = 295.5$. For Sr or Pb, this simple approach works only for samples, like some micas, where the initial component is so low that errors in its estimation do not matter; sometimes it may be wrong, even for Ar.

The second condition is less easily controlled. All samples are examined, before analysis, for any sign of strain or re-crystallization. Even so, discordant ages are often the first evidence for disturbance. It is imperative, therefore, that multiple samples should be taken, over the widest extent of a rock unit, with preference for samples which yield at least two datable minerals. In some cases whole-rock samples may be analyzed. Close control from good prior mapping is essential.

Almost all K-Ar dates are measured in this way. Minimum sample size is governed by mineral homogeneity, since K and Ar must be measured on separate fractions. In a new method which obviates this drawback, and has some other advantages, some of the ^{39}K in the sample is converted by fast neutrons to ^{39}Ar; a single argon extraction yields the ratio $^{40}\mathrm{Ar}/^{39}\mathrm{Ar}$ which is proportional to Ar/K. This leads to the age, by comparison with a simultaneously-irradiated sample of known age. The method is less than ten years old; the first Australian results appeared recently (Green, 1973).

Application. K-Ar ages on minerals and/or fresh holo-crystalline whole-rock samples normally reflect the last major geological event, be it time or igneous intrusion or the time of a later superimposed metamorphism. A dramatic example of this latter kind of response is seen near the Broken Hill orebody (Richards & Pidgeon, 1963), where Rb-Sr and K-Ar ages of biotites (450 -500 m.y.) markedly postdate the 1700 m.y. of the dominant metamorphism in the country rocks and related ores (Shaw, 1968). The heating event reflected by the mica ages may have been associated with concentration of the secondary Thackaringa-type orebodies out of the country rock (Reynolds, 1971).

Several porphyry copper deposits in Papua New Guinea and Bougainville have been dated by both whole-rock and mineral K-Ar analyses (Page & McDougall, 1972). The age of mineralization was obtained either from an andesitic hornfels which was reheated and altered at that time, or from potash-enriched materials within the mineralization envelope; the original intrusives proved to be distinctly older. Thus, at Panguna, Bougainville, the mid-Pliocene mineralization at 3.4 ± 0.3 m.y. compares with intrusives 4 to 5 m.y. old. The porphyry copper prospect at Yanderra, in the Highlands of New Guinea, was emplaced in the late Miocene (7 to 8 m.y.), about 5 m.y. later than the main mid-Miocene (about 13 m.y.) intrusives. The prospect at Mt Fubilan (Ok Tedi), Western Papua New Guinea emphasizes the extreme youthfulness of this porphyry copper environment, with hydrothermal alteration and mineralization at 1.1 to 1.2 m.y. (Pleistocene).

Several other mineralized porphyry bodies in this region have now been dated (Page, unpublished data) at between 2 and 5 m.y. i.e., they are of Pliocene age.

This work has complemented other geological evidence for a close temporal relationship, in the southwest Pacific area, between porphyry copper mineralization and calc-alkaline magmatism during the latter half of the Tertiary. Page & McDougall (1972) regarded the mid-Miocene ages in Papua New Guinea as signalling the onset of interaction and collision between the Pacific Plate and the northward-moving Australian Plate, which has New Guinea as its leading edge.

Isochron Method: Grouped Samples

Normally, when initial Sr or Pb (or even Ar) is expected, it is better to use the Isochron Method. In this the observed daughter isotope ratio is treated as a function of the parent/ daughter abundance ratio. The variables used may be:

a) $^{87}Sr/^{86}Sr$ versus $^{87}Rb/^{86}Sr$
b) $^{206}Pb/^{204}Pb$ versus $^{238}U/^{204}Pb$
c) $^{207}Pb/^{204}Pb$ versus $^{235}U/^{204}Pb$
d) $^{208}Pb/^{204}Pb$ versus $^{232}Th/^{204}Pb$
e) $^{40}Ar/^{36}Ar$ versus $^{40}K/^{36}Ar$

Further manipulation of the Age Equation shows that a series of related samples should define a straight line. Points may range along the line because, for example, different feldspar contents result in different values of the Rb/Sr ratio but they should adhere closely to the line, provided the samples have the same age and the same initial $^{87}Sr/^{86}Sr$ ratio, and the system has remained closed. The intercept on the ordinate (Fig. 1) represents the initial ratio; the slope of the line defines the age.

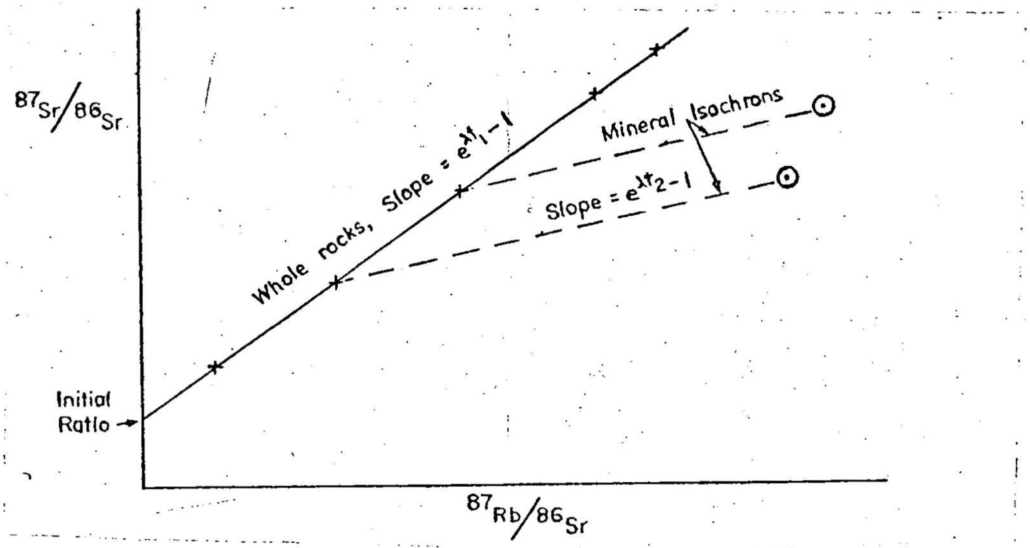

Fig. 1. Isochron Diagram

The isochron approach is very powerful. It is standard for Rb-Sr dating, and has more recently been applied to the U-Pb, Th-Pb, and K-Ar systems. Statistical analysis can expose departures from the assumptions of common age, lack of contamination, or closed system behaviour, and help choose between them. The method can provide both the original time of formation, from a series of whole-rock samples, and the time of a later disturbance to the same set of rocks, from the slopes of the tie-lines between points for separated minerals and the corresponding whole rocks. The method breaks down if there is extreme open-system behaviour (as in a metasomatic event) or if the system is a partial mixture, with end members of very different chemistry (Black et al., 1972). For this method, sampling is somewhat different. Experience has shown that in a sedimentary or metamorphic sequence, and even in a single granitic intrusive, there may be large variations of initial $^{87}Sr/^{86}Sr$; this volates the isochron assumptions. To reduce this uncertainity, between 6 and 10 samples are collected from each sampling site. The widest possible range in felsic-femic proportion is chosen so as to increase the possible Rb/Sr dispersion. Sites must be carefully selected so as to keep the work load within reasonable limits without impairing the result.

Examples. In a study or gold mineralization and granite emplacement in the Kalgoorlie-Norseman area, Western Australia, Turek (Compston & Arriens, 1968; Turek & Compston, 1971) used samples from gold-quartz reefs, and both biotite and sericite from altered wall-rocks, to show that the age of gold mineralization is 2400 ± 40 m.y. This event was much younger than the granites (at least 2600 m.y.), thus excluding earlier proposals that the granites had played a role in the genesis of the gold lodes. Turek's results seem to show that it is more likely that the mineralization could have been genetically associated with emplacement of mafic dykes 2420 ± 30 m.y. ago.

Other dated mineralizations are in northwestern Tasmania (McDougall & Leggo, 1965; Brooks, 1966; Richards, 1968) and eastern Queensland (Webb, 1969).

The age of the important Precambrian deposits near Mount Isa-Cloncurry, northwest Queensland, is still not clear, despite combined attack by K-Ar, Rb-Sr, and U-Pb methods. Unpublished data (Farquharson, pers. comm.) suggest that direct dating of the ore host, the Mount Isa Group, may prove difficult. Complexity of geological history, and the lack of certain critical relationships, have resulted in divergent inferences from the available data. Thus, Farquharson and Wilson (1971) and Wilson (ms. pers. comm.) believe on structural grounds that the Mount Isa Group is older than the southern part of the Sybella Granite (i.e., pre-1650 m.y.). On the other hand, stratigraphic extrapolations (Wilson et al., 1973; Plumb & Derrick, this volume) suggest that at least the northwest part of the Sybella Granite (1580 m.y.; Page & Derrick, 1973) is unconformably overlain by rocks correlated with the Mount Isa Group, and that therefore these sediments (and by extrapolation the ore) are younger than 1580 m.y. Resolution of the enigma must await more detailed evaluation of the field, structural, and isotopic evidence.

Concordia: Open System Correction

If the evidence is that the Rb-Sr system has not remained closed, there is very little that can be done with the results. However, in the U, Th-Pb system, there is one possible further step. There are three separate decay systems ending with isotopes of lead. Hence three independent ages may be calculated. Further, since there is no detectable variation of the $^{238}U/^{235}U$ ratio, a fourth, but not independent, age may be calculated solely from measurement of the $^{207}Pb/^{206}Pb$ ratio in a lead sample. All four ages (denoted as t) should be the same, but frequently are not. Two discordant sequences in the U-related ages have been observed:

a) $t(^{206}Pb/^{238}U) > t(^{207}Pb/^{235}U) > t(^{207}Pb/^{206}Pb)$
b) t(") < t(") < t(")

The first has been ascribed to loss of lead from the system, the second to loss of U. In each case the $t(^{207}Pb/^{206}Pb)$ seems to be the least erroneous.

The data are interpreted by plotting on a Concordia Diagram (Fig. 2), with axes $^{207}Pb/^{235}U$; $^{206}Pb/^{238}U$.

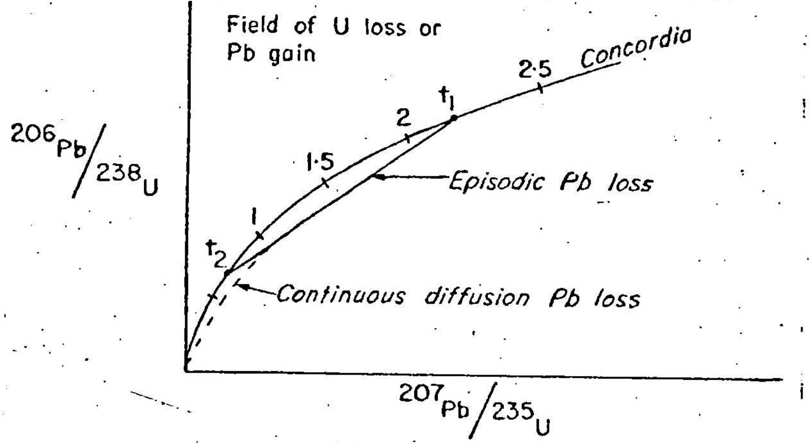

Figure 2. Concordia Diagram

Samples yielding concordant ages (closed system) should appear as points on the curved line, called Concordia. Disturbed samples normally plot below the line (Pb loss or U gain). Loss of U (gain of Pb) drives points above the line. A series of related samples plot linearly, and may be extrapolated to cut Concordia at two points, t_1 and t_2. Point t_1 corresponds to the time of mineral formation; t_2, if real, to the time of a short event during which all the observed lead loss occurred. On the other hand, if the loss were continuous, say by diffusion at ambient temperature, the points would lie along the dotted curve (Fig. 2) which meets Concordia at t_1 and at the origin; t_2 is then an artefact. It is rare to find samples plotting sufficiently close to the origin (i.e., high Pb loss) to permitan objective choice between the two models. Most reported results deal with U-rich accessory minerals such as zircon or sphene; pitchblende samples have also been studied.

Application. Only a few Australian zircon dates have ever been published (Richards et al., 1966). The only other study was in an unpublished report by Fander (1961) who used the old Pb-alpha method, which formally yields results akin to a $^{206}Pb/^{238}U$ age except that no correction is made for possible initial Pb; if that is absent, Pb loss would yield too young an age. The only concordia studies have been on pitchblendes from South and

Western Australia and the Northern Territory (Greenhalgh & Jeffery, 1959), and some further preliminary results by Hills and Richards (1972) and Cooper (1973). These indicate two possibly significant events in the history of uranium deposition in two widely separated localities in northern Australia- late Precambrian (700-800 m.y.) and early Palaeozoic (400-500 m.y.). Further unpublished results point to even older uranium mineralizations in the Alligator River area (Hills, pers. comm.).

Lead Model Ages

The so-called lead model ages are based upon an assumed model of grand-scale earth processes which does not correspond to reality (see Richards, 1968; 1971). None of the published equations is therefore good enough for more than the broadest age inference. These "ages" cannot, therefore, be used legitimately for detailed regional comparisons with other dating methods. This does not, however, discredit the use of lead isotopes as very powerful geochemical tracers (see later, p. 17).

Limitations

Detailed assessment of dates from a number of methods, or laboratories, should be tempered by awareness that there remain some intercomparison problems. These lie both in the calibration of the spikes used for measurement of concentrations by isotope dilution, and in the choice of isotopic parameters such as half life; authors should, and normally do, quote the values used. The situation is at present under review. It seems likely that present inconsistencies, which may be as large as 6 percent, will eventually be resolved; for the moment this value may be taken as an upper limit to the possible disagreements between sets of published data.

ISOTOPIC CORRELATIONS

We now shift our focus to the factor which was a complication in the age calculations, the isotopic proportions of the Pb, Sr (or Ar) initially in the sample, in amounts quite unrelated to the present concentrations of the U and Th or Rb (or K). These proportions were in their turn the result of daughter product addition to an already-existing stock, but at one incarnation further removed; they can reveal important clues to the location of the earlier material, and its duration in that earlier environment. The ability thus conferred on the geochemist to penetrate to the otherwise inaccessible past is one of the fascinations of isotope work.

Two complementary approaches are possible. The first employs the same analytical results used for dating. Once again the isochron diagram (Fig. 1) gives the best values, but initial ratios may also be derived for each individual sample, given an acceptable independent age estimate. The second method, essentially for Pb only, applies to those minerals which contain Pb but virtually no U or Th. The ratios now represent the average of the material acquired by the concentrating and transporting medium from which the minerals formed.

With each choice it is possible to paint a picture in a variety of styles. In the past a moderately broad brush has been popular. The analyzed samples were obtained, more or less at random, from many regions of all ages, and the general trends interpreted in terms of grand-scale Earth processes. Whilst the resulting canvasses do help to illustrate many features of ore-formation processes they could sometimes mislead, because their scale does not permit the testing of vital assumptions about each sample. The same limitation restricts their power to delineate the features of an individual mining district. For this the work must be more detailed, with closer geological control. It is our view that future progress requires the multiplication of such local studies, as a necessary check on the validity of the broader interpretations.

Earth-scale Studies

Initial Ratios. The main emphases of Sr and Pb studies have been on the origin and emplacement of igneous rocks, the relative contributions of Crust and Mantle to continental growth, the properties of the Mantle as deduced from rocks of presumed mantle origin, the sedimentary cycle, and sea water as an averaging medium for crustal material (Faure & Powell, 1972; Doe, 1970).

For ores, the dominant question has been of origin; what part, if any, have mantle rocks played? Discussion for Sr centres on the host rocks; it is based on the observation (Hedge & Walthall, 1963; Faure & Hurley, 1963) that the initial ratios of mafic rocks are low, and vary only slowly with time, whereas in flesic rocks they are often higher and more variable. From this it was inferred that Rb has strongly fractionated, relative to Sr, into the Crust. Hence any rock with a high initial ratio must have had long residence in, or at least some contact with, high-Rb crustal material, whereas a felsic rock with ratio close to the basalt value points to a source with a long history of low-Rb abundance; i.e., even if it were not of direct mantle origin, the source can have had little contact with average continental crust (Faure & Hurley, 1963; Compston, 1963).

In north Queensland (Black & Richards, 1972a) tin in exploitable quantities is closely related to felsic rocks with initial ratio 0.710, as compared to about 0.703 for "mantle" material. Thus since the rocks must have at least partly derived from continental material, we would do well to look for mechanisms which concentrated the tin also from the same source, into either the magma or the related hydrothermal fluids.

Lead is more revealing. Both U and Th have shorter half-lives and tend to concentrate towards felsic rocks (Doe, 1970, p.51). Thus long residence in a continental environment should be signalled by relative enrichment in the three radiogenic Pb isotopes (206, 207, 208). But now there is a further compilation, because Lambert and Heier (1968) have noted that granulite-facies rocks are depleted in U. Lead derived from highly-metamorphosed rocks should therefore be recognizable by its relative depeltion, especially, in the 206 isotope (Armstrong & Cooper, 1971). The study by Black and Richards (1972b) is one of the few so far where such lead has been recognized as a contaminant of the ore fluid.

Study of the systematics of rock-lead variations was inhibited for some years by marked analytical difficulties, not the least of which were high contamination levels, both from reagents and from motor-car exhausts, and comparative insensitivity of the mass spectrometry. Galenas pose fewer analytical problems, and the general principles should be the same as for rock leads. Study of their isotopes has generated a boluminous literature.

Ore Leads. The abundances of the three radiogenic isotopes are expressed as ratios relative to ^{204}Pb, which is assumed to be unchanged in abundance since the birth of the Solar System. The most popular display uses the ratio ^{206}Pb/^{204}Pb as abscissa on two graphs; the first, with dependent variable ^{207}Pb/^{204}Pb, involves only the decay of uranium, with U/Pb and time the only parameters. In the second, with vertical axis ^{208}Pb/^{204}Pb, decay of Th and abundance ratio Th/U are also involved.

Two clear isotopic classes of ore lead emerge. The first involves orebodies of every age, from all continents; many are of major economic size, and are geologically conformable with the surrounding strata. Typical examples are Broken Hill and Mount Isa (Cooper et al., 1969). Data points from these leads define with remarkable fidelity a single, time-sequential curve (Fig. 3) which is best called the "ore-lead growth curve"; other terms used in the literature carry genetic implications which have tended only to confuse discussion.

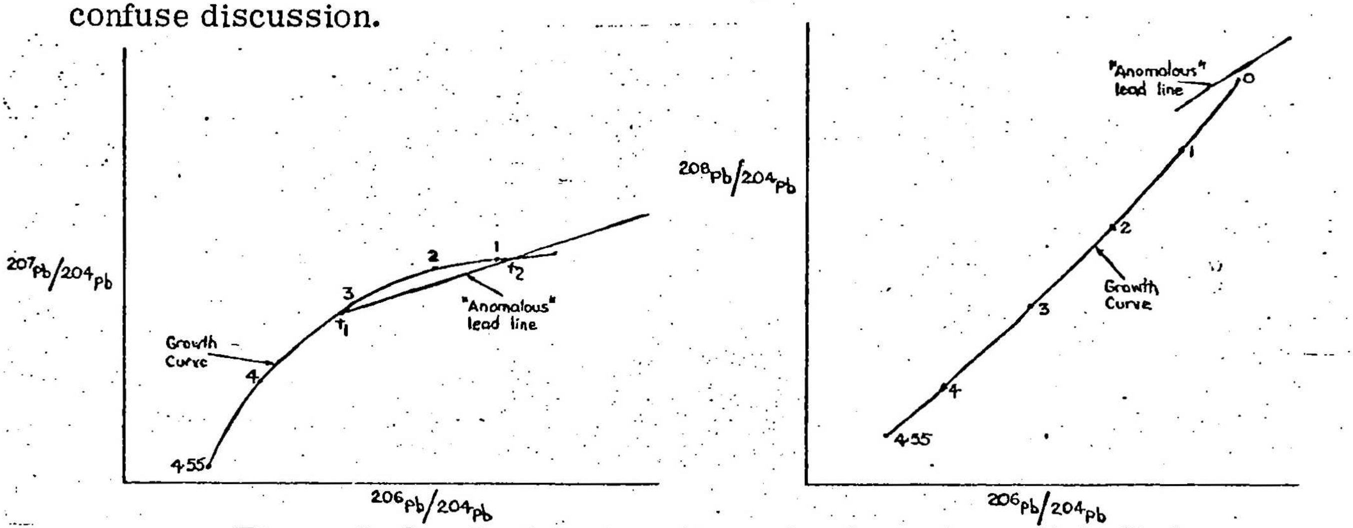

Figure 3. Ore lead systematics; simple systems. Labelled ages on the growth curves are given in 10^9 yr.

The second class comprised initially those samples which did not fit the first group, and were therefore called "anomalous". A series of geologically-related samples yields strongly-divergent data which closely define a straight line; on the ^{206}Pb-^{207}Pb diagram this line usually passes through the point for the dominant ordinary lead (class 1) of the region (Kanasewich, 1968). These deposits are frequently smaller, occurring in fissures and veins intruding the country rock; the Thackaringa-type deposits near Broken Hill are a good example (Reynolds, 1971). It should be noted, however, that this class includes the major producers of mid-continental USA (Mississippi Valley; Richards et al., 1972).

Both the growth curve and the linear distribution may be described quite closely by simple relationships derived from the Radioactive Decay Law.

This, and the apparent concordance between broad regional trends and the ages thus derived, have misled many authors into reading far more than is really justifiable into these simple models. In particular, the so-called "model ages" depend very markedly upon the validity of a number of assumptions, which include that of a hypothetical source region of constant U/Pb and Th/Pb ratios which seem not to be affected by the periodical withdrawal of the lead required to form orebodies. It is not really surprising, therefore, that as more detailed knowledge accumulates, marked divergences are found (Richards, 1971) between these "model ages" and the actual ages of mineralization. For example, the estimated model ages for Mount Isa and Broken Hill range from 1600 to 1500 m.y. (Kanasewich, 1968; Cooper et al., 1969). Broken Hill ore is unlikely to be younger than the 1700 m.y. of the main metamorphism (Shaw, 1968); for Mount Isa the situation is still not clear (see p. 8). Therefore, we recommend that published lead model ages be treated with great reservation.

Recent attempts by Russell (1972), Russell and Birnie (1973), Armstrong (1968), and Armstrong and Hein (1973) to erect models better suited to current geological concepts have not resulted in agreement. At their present stage of development, however, they fit many of the known isotopic facts at least as well as the simple model, emphasising that none of the present interpretations is unique. The study by Armstrong and Hein, in particular, gives warning in strikingly visual form that the linear data-trends may not always relate simply to ages, thus reinforcing a suspicion already expressed by Richards and others (1972). Perhaps this is the clue to resolving the difficulty Reynolds (1971) found with his Thackaringa data; these leads could represent an admixture from at least two different linear sets, each with a different initial value, and thus no direct time correspondence.

Region-scale Studies

Because of their dependence on the timing and size of parent concentrations, the isotope ratios, of Pb in particular, will differ from sample to sample. They can thus serve as a very powerful supplement to a more general trace-element study, in particular to test possible genetic relationships between the rocks and ores of a district. This is illustrated with a few examples of this more detailed application:

Pb and Sr Tracer Studies: Examples. In a purely petrological application, Pidgeon and Compston (1965) showed that the Cooma Granite, NSW, and the nearer high-grade metasediments of its aureole are indistinguishable in age and initial strontium. Rocks from the more remote zones differ significantly, but are in complete accord with the field evidence in suggesting that the Cooma Gravity was derived anatectically from the adjacent sediments as the ultimate step in the regional metamorphic process.

In another example Ewart and Stipp (1968) examined the Pleistocene to Recent Taupo Volcanic Zone of the North Island, New Zealand, and the neighbouring Mesozoic greywacke-argillite sediments. The Sr isotope evidence by itself was not very compelling, but in combination with a comprehensive trace-element study it led the authors to conclude that the dominant rhyolitic magmas were indeed derived by partial melting of the Mesozoic sediments, as had been suggested by previous field workers. The data for the less-abundant andesites were somewhat ambiguous, although they did allow the elimination of several genetic models. Minor basalts were isotopically distinct.

Cooper and Richards (1969) studied lead isotopes in volcanics and associated ores in the adjacent Coromandel-Te Aroha region. The isotopic evidence linked hydrothermally-altered ("propylitized") andesite, rhyolite, and galena-bearing veins with the Mesozoic basement sediments (Armstrong & Cooper, 1971), in clear distinction from unaltered andesite. From this, the field relations, and comparison with results of the prior work farther south by Ewart and Stipp (1968), Cooper and Richards deduced a genetic sequence as follows: 1. Energy from a suitable heat source accompanies the formation of at least some of the andesites, together with mobilization from the Mesozoic sediments of fluids which produced the propylitization effects and formed the mineralized veins. 2. Possible later formation of the unaltered andesites? 3. Partial melting of the Mesozoic sediments to form the rhyolitic magma. The relevance of such an inferred sequence to problems of ore genesis is selfevident. Note that the isotopic data suggest absence of direct chemical connection between the andesite magma (the possible heat source) and the mineralizing solution.

Among the Precambrian granites near Mount Isa, Farquharson and Richards (1970) deduced from colinearity on the Rb-Sr, U-Pb, and Th-Pb isochron diagrams that the Sybella Microgranite and the Mica Creek Pegmatite may be cogenetic, with similar age (1550 m.y.) and initial ratios. The initial lead is distinct (especially in ^{206}Pb) from that in the nearby Ag-Pb-Zn orebody, thus excluding any direct genetic relationship between these rocks and the ore.

In the Mount Garnet-Chillagoe-Herberton area of northeastern Queensland extensive remnants of Upper Palaeozoic magmatic activity occur where the Precambrian Gerogetown Inlier abuts agsinst the northern end of the Palaeozoic Tasman Geosyncline (Oversby et al., this volume). There has been general agreement that the Upper Palaeozoic granites were responsible for the mineralization, and that the Sn, W, and Mo are related to the Elizabeth Creek Granite, one of the two genetic types proposed by Branch (1966).

However, there is no consensus on the origin of the other known metals. In the Chillagoe area Best (1962), de Keyser and Wolff (1964) and Branch (1966) argue for introduction of Cu, Pb, Ag, and Au into metasomatic deposits by a variant of the Herbert River Granite. By contrast, Blake & Smith (1970) and Blake (1972) report evidence for zonation of W, Sn, Cu, and Pb about a mass of Elizabeth Creek Granite in the Emuford-Irvinebank area.

The Pb and Sr isotopic evidence of Black and Richards (1972a & b) suggested genetic connection between both granite types proposed by Branch and the nearby Featherbed Volcanics. However, the "contact metasomatic" ore near Chillagoe contains lead quite distinct from that in the nearby supposedly-related granite. The lead ores near Irvine-bank resembles the magma lead, but part of the zonation noted by Blake and Smith lies within the Featherbed Volcanics, apparently 30 m.y. younger, this throwing into doubt a direct connection. However, further work (Blake, unpublished) has now revealed quite complex age relationships within these volcanics; whereas some of the lead deposits within them (e.g., around Bambord Hill) could not have been formed by the older granite; those farther east, alongside the district zonation may well have been. At least three distince Upper Palaeozoic events may be discerned; all three involve granite intrusion, at 328 m.y., 300 m.y., and about 280 m.y. All that is revealed by the available evidence is that the first, Elizabeth Creek Granite, cannot have been the sole contributor to the mineralization.

The orebody lead isotopes do, however, yield evidence for at least two distinct lead components, one related to the Upper Palaeozoic igneous rocks, the other to the Precambrian craton. Confirmation of the latter component is suggested by a marked correlation between ore-lead isotopic composition and distance from the boundary of the exposed cratonic rocks.

The overall evidence, then, suggests a model in which the Upper Palaeozoic felsic magmas provided at least a heat source for lead mobilization, and probably also fluids containing lead isotopically related to the galenas and igneous rocks of the Irvinebank-Emuford area. During their migration to depositional sites these highly-active fluids leached additional amounts (up to 80 percent at Dargalong) of lead from the traversed country rock.

Argon. There have been comparatively few reported cases of initial argon. This is largely because of its high volatility; most of the argon in the magma escapes during cooling. Exceptions are found usually in young or low-K materials, where the remaining initial argon is not completely masked by the later decay product. If found, it marks either the presence of incompletely-digested xenolithic material (McDougall et al., 1969) or a set of external conditions which inhibited argon escape (Dalrymple & Moore, 1969). Deep rock cover is one such condition (Hart & Dodd, 1962; McDougall & Green, 1964; Lovering & Richards, 1964). An example quoted by Richards and Pidgeon (1963) may well be associated with one of the mineralization events at Broken Hill. Zhirov and others (1972) have shown that gases are enclosed within the ore minerals themselves; they and Pushkarev (1971) have attempted some preliminary calibrations, but many more are needed before the implied pressure-temperature conditions can be established with confidence.

LIGHT ELEMENTS

In this section we turn away from age and metallic components of the ore to the non-metallics, to the contaminating and transporting medium, and to the temperature, pressure, pH, and salinity of formation. These all involve the lighter elements, none of which takes part in nuclear transformations;

their isotopes show separation in any process involving motion because the fractional differences in mass are big enough to have significant effect on reaction or translation velocity.

S, O, and H are all relevant to ore studies. Useful reviews are by Hoefs (1973), Taylor (1967), and Jensen (1967). Carbon, too, can tell about coals and carbonate minerals (Jeffery et al., 1955; Taylor et al., 1967). Other elements such as F, Si, Al, and Mg may warrant attention, should sufficiently exact measurement be possible.

All results are presented as parts per thousand difference from arbitrarily-chosen standards: mean ocean water (SMOW) for O and H, a standard carbonate (Chicago PDB) for C, or the troilite phase in Canyon Diablo meteorite for S.

e.g., $$\delta^{34}S = \left(\frac{\text{Sample } {}^{34}S/{}^{32}S}{\text{Standard } {}^{34}S/{}^{32}S} - 1\right) .1000 \text{ per mil. (mille)}$$

The data may be approached in two ways. The first is qualitive, in terms of an average and range of per mil. values established for each rock type or genetic class. In the second, the per mil. differences between co-existing minerals are used to estimate formation temperature, degree of equilibrium between minerals, and the nature and origin of a hydrothermal fluid or of the non-metallic components.

Qualitative Correlations

Ultramafic and mafic rocks - and seawater sulphate - all show a marked world-wide uniformity in $\delta^{34}S$, $\delta^{18}O$, and $\delta^{13}C$, which implies that each group is derived from a well-mixed source (Gast, 1967). For the rocks this should be the Mantle; the seawater value, which is different, should represent an average of continental material with a small component from sea-floor emanations. In that seawater shows an irregular time variation, some crude dating even has seemed possible (Solomon et al., 1971). The per mil. value, and the range, increase slightly as rocks become more silica-rich, with the trend continuing with metamorphics. But these are all in marked contrast to sediments, where the values range widely, especially $\delta^{34}S$. Many, but not all, authors argue from this for biological activity in the reduction of sulphate to sulphide (contrast Solomon & Jensen, 1967, with Warren, 1972). S evidence has been used in this way by Creevy and Donnelly (1973) to argue for three different times and modes of Cu mineralization at Pernatty Lagoon, South Australia.

This descriptive approach has had its greatest success in the study of water. In fresh (meteoric) waters there is a linear relationship between D (= ^{2}H) and ^{18}O (Taylor, 1967), which depends on latitude; the strongest effect is close to the Poles, or near large masses of snow and ice. Neighbouring thermal waters show signs of exchange with the 0 of the host rocks. Craig (1969) has used the slightly different seawater trend to argue that Red Sea hot brine water was derived from nearby seawater; Sheppard and others (1971) use the fresh-water line, and the parallel lines for the clay products of surface weathering, to argue for surface water involvement in formation of the western USA Porphyry copper deposits.

There must be caution, however, in the detailed application of these arguments. Coexisting minerals do show the regular differences, both in 0 and in S (Rafter, 1965; Taylor, 1967), which considerations of mass-dependent particle velocity might lead us to expect. Indeed Ryznar et al. (1967) have shown the possibility of quite marked isotopic shifts during depletion of a non-replenished pool. Their example emphasises that the effect can be purely inorganic; the more popular case, of biological H_2S production (in sediments), is discussed by Kemp and Thode (1968). It is important to notice from this, however, that wide isotopic range is not by itself sufficient evidence for organic activity (Rafter, 1965).

Quantitative studies

The second approach has been applied both to chemical equilibria in fluid systems and to mineral formation, in oxygen (Taylor, 1967), carbon (e.g., Hulston & McCabe, 1962), and sulphur (Solomon et al., 1969; Thode et al., 1971).

Clayton and Epstein (1958), for 0, and Stanton and Rafter (1967), for S, recognized that a temperature-dependence in the per mil. difference between mineral pairs opened the way to a potentially useful thermometer. Many laboratory calibrations later, the method is beginning to show increasing internal consistency, and agreement with other geothermometers (Taylor, 1967; Groves et al., 1970), although complete success has yet to be achieved (Sheppard et al., 1971).

There are still many cases where different mineral pairs yield conflicting temperatures. The cause could be imperfect mineral separations, particularly with sulphides (Groves et al., 1970), or post-solidification reaction with the surroundings (Wilson & Green, 1971), or non-attainment of equilibrium between two components of different origin (Kusakabe, oral communication, Nelson, New Zealand, May, 1973). Sheppard et al., (1971) show that quartz in particular is resistant to later alteration; therefore 0 in quartz-sericite, for example, from an altered rock, would be a very poor guide to the temperature of that alteration.

Finally, the laboratory calibration experiments permit isotopic recognition of the hydrothermal fluid which altered a host rock, and was thus probably the vehicle for the ore. This should then lead to an estimate of its origin, and the degree of its interaction with the wall rock (Taylor, 1967). A future goal is direct isotopic assay of the liquids in small vesicles in the minerals, as representatives of the fluids present at the time of crystal growth.

CONCLUSION

From this brief summary it can be seen that isotopic studies, both of light elements and of the products of radioactive decay, are worthy partners of age measurements in yielding much of direct value to the economic geologist.

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TABLE 1: MAJOR ISOTOPIC AGE METHODS FOR GEOLOGY

Method	Generalized Decay Scheme	Half-life of radioactive isotope	Decay Constant	Materials that can be dated	Effective Age Range
U-Pb	$^{238}U \rightarrow {}^{206}Pb$	4.5×10^{9} y.	1.54×10^{-10} y.$^{-1}$	Uraninite, pitchblende, zircon, sphene, apatite, epidote, whole rock	10^{7} years to t_o
U-Pb	$^{235}U \rightarrow {}^{207}Pb$	0.71×10^{9} y.	9.72×10^{-10} y.$^{-1}$		
Th-Pb	$^{232}Th \rightarrow {}^{208}Pb$	13.9×10^{9} y.	0.499×10^{-10} y.$^{-1}$		
Rb-Sr	$^{87}Rb \rightarrow {}^{87}Sr$	5.0×10^{10} y.	1.39×10^{-11} y.$^{-1}$	Mica, feldspar, amphibole, whole rock, glauconite	5×10^{6} y. to t_o
K-Ar	$^{40}K \xrightarrow{(11.2\%)} {}^{40}Ar$ $^{40}K \xrightarrow{(89.8\%)} {}^{40}Ca$	1.31×10^{9} y.	0.585×10^{-10} y.$^{-1}$ 4.72×10^{-10} y.$^{-1}$	^{40}K - ^{40}Ar : mica, feldspar, amphibole, pyroxene whole rock, glauconite ^{40}K - ^{40}Ca : dating not practicable	5×10^{4} y. to t_o
	(Note: for $^{40}Ar/^{39}Ar$ variation on technique, see text)				

t_o = age of Earth, about 4.56×10^{9} y.