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Part II: Models for Uranium Ores in Sandstones and in Carbonaceous Pelites

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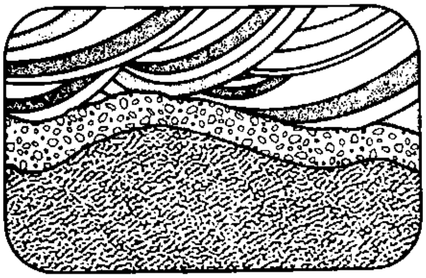

Ore Deposit Models - 3. Genetic Considerations Relating to Some Uranium Ore Deposits,

Part II: Models for Uranium Ores in Sandstones and in Carbonaceous Pelites

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Introduction

Part I of this discussion (Tilsley, 1980a) reviewed the changes in chemical behavior of uranium and uranium minerals as the earth evolved and related these changes to the kinds of deposits that may be expected in various geologic environments through time. Consideration was given to some aspects of a model for the formation of uranium ores in Proterozoic conglomerates and the limiting factors that confine conglomerate ores to sediments older than about 2,200 Ma.

Part II presents two more models. A genetic model for sandstone-hosted uranium ores is reviewed and several areas where deviation from conventional wisdom may be useful are suggested. A model for concentration of uranium in carbonaceous pelites younger than 2,200 Ma is proposed including the mechanism that may contribute to the development of protores and ores in these rocks.

Sandstone-Hosted Ores

This class of uranium ore is of major importance on a worldwide basis. Over 300,000 tonnes U_3O_8 has been produced from sandstone-hosted ores in the United States of America alone. Significant deposits are also known in Argentina, South America, Australia (Westmorland) and Africa (Niger and Gabon). The subclasses include classical 'roll-fronts', 'tabular bodies' and channel-type ore deposits, most commonly developed in permeable sandstones.

The geometry of these bodies is variable depending on a number of sedimentary and hydrological controls. The concentration mechanism is the same in all cases. Electrochemically oxidizing solutions carrying dissolved uranium enter an environment more reducing than the upper stability limit of uraninite (UO_2-UO_3) and the uranium in solution is deposited at the so-called oxidation/reduction front. Each mineral complex has some critical voltage (Eh) above which it is oxidized and below which it is reduced. The most visibly obvious oxidation/reduction front is that of iron, since the reduced minerals are grey-green and the oxidized minerals are reddish-brown. Uranium is reduced and precipitated from solution at an Eh close to that of the Fe^{+2}/Fe^{+3} boundary, so it is visibly associated with the iron oxidation/reduction front. Other elements that may be associated with uranium in this sort of environment form minerals that have oxidation/reduction boundaries slightly displaced from those of uranium or iron. This is reflected by the position of their maximum concentrations at some distance ahead or behind the Fe^{+2}/Fe^{+3} boundary.

The oxidation/reduction boundaries migrate within the sandstone at a velocity which is a function of the rate at which free oxygen is supplied to the system by ground water and the oxygen-demand of the sediments through which the ground water flows. Organic carbon and ferrous iron are the chief oxygen demanding agents in the system. The low capacity of water to transport free oxygen (8 to 10 gms/tonne) results in the rate of advance of an oxidation/reduction front being orders of magnitude less than the rate of ground water flow. For example, the oxidation of 1% FeO to Fe_2O_3 would require about 10,000 gms of oxygen per cubic metre of sediment, or approximately one thousand tonnes of water passing through that volume of sandstone. Given a ground water flow velocity of 0.5 metres/year and a porosity of 20%, this would require 10,000 years for the front to advance one metre, 0.1 millimetre/year.

The rate at which oxidation/reduction fronts move in sediments is relatively slow but, in terms of geologic time, these fronts are transient. Economic deposits tend to be confined to rocks younger than Carboniferous and, although there is some evidence of possible 'fossil roll-fronts' in rocks up to about 2000 Ma, the Mesozoic and Cenozoic sediments contain the bulk of the mineable reserves.

The literature contains extensive information on the American sandstone-hosted ores and some data are available on similar deposits in other parts of the world. Papers by Adler (1974) and Rackley (1976) are particularly informative.

Source of Metal. Most of the uranium now concentrated in sandstones probably originated in source areas topographically higher (i.e., up ground-water flow) than the sediments at time of ore formation. In these highlands, uranium could be derived by weathering from acid igneous rocks, pyroclastics, lavas and, occasionally, from uranium-enriched metamorphic rocks or older sediments. In many situations, tuffaceous components of the host sediments are believed to have provided much of the metal.

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Release of Metal. Chemical weathering processes, at the surface and under special topographic conditions at some substantial depth, are responsible for release of metal from its source rocks. Devitrification of volcanic material is considered to be an important factor also, where extrusive rocks appear to be a likely source.

Transportation. Metal is carried in solution in surface and ground waters into and within the sediments along aquifers that may extend to great depth below the erosion surface.

Concentration of Metal. Uranium is removed from electrochemically oxidizing solutions by reduction at the oxidation/reduction interface mentioned earlier. The interface develops in permeable rocks containing ferrous iron, e.g., as pyrite, and perhaps some carbonaceous material. Although this type of deposit may form at considerable depth, the oxygen is derived from the surface where the water that acts as transportation medium for both the oxygen and metals is collected.

Modification. As stated previously oxidation/reduction boundaries are notably mobile within the host sediment during the time that significant volumes of ground water are available to them. Through time, such concentrations may be completely flushed from the section.

Changes in the recharge-discharge system may curtail ground water flow through the host rocks. Mineralization on the oxidation/reduction front remains stable although the oxidized portion of the sediment through which the front has moved may have been reduced again, leaving little visible indication of its previously oxidized condition.

Preservation. Survival through time of mineralization concentrated on an oxidation/reduction boundary requires that the hydrological system that was active during formation is changed, so that free oxygen is not brought into contact with the mineral concentration at depth. The interface must be protected from weathering and erosion. Metamorphism of the section beyond some critical level is likely to cause dispersion of the metal values. There are no commercial uranium deposits of obvious oxidation/reduction front origin in metamorphosed rocks. It would therefore appear that this type of mineralization is unlikely to persist through any but the lowest grade metamorphic event.

Discussion

While in the past there has been rather strong emphasis on the geometry of this type of deposit, e.g., 'roll-front'; 'tabular deposits', et cetera, consideration of the genetic model should not be limited by geometric constraints. The shape of the oxidation/reduction front and the form of associated mineralization are the result of variations in attitude, permeability and chemical composition of the host rocks, and with the volume, pressure and chemistry of the ground water flowing through the system.

The term 'sandstone-type' deposit has been applied to any uranium deposit developed in sandstone-host rock. Not all the deposits included in this classification have formed in the epigenetic manner discussed. Syngenetic concentration in stream channels during the time of active sedimentation, or at least prior to the onset of diagenesis, is not uncommon. Recognition of this variant in the early stages of exploration is important to effective prospecting. The concentration process is by reduction, usually initiated by decaying organic material in the stream bed sediment and commercial grade mineralization is usually confined to the active channel sediments, as opposed to the finer argillaceous overbank or laterally-deposited material. Commonly however, when associated with mineralized channels, these finer sediments are enriched in uranium as compared to argillaceous material at similar stratigraphic positions but remote from the main drainage axis. This latter factor can be used in exploration.

In Canada, exploration success of this type of deposit has not been remarkable. In spite of poor success in this country, sandstone-hosted epigenetic 'geochemical cell' deposits are an excellent exploration target in much of the world.

Carbonaceous Pelite-Hosted Uranium Deposits

Uranium deposits hosted by post-oxygenation Aphebian age pelitic sediments have been discovered in North America and Australia during the past ten years. Some of these have proven to contain particularly important uranium reserves. In many other localities, pelitic sediments of comparable age laid down in similar depositional environments have been found to be uranium-enriched locally.

In some classifications these deposits have been called vein-type. In the Northern Territory, Australia, discoveries have been made close to the exposure of an unconformable contact between metamorphosed pelitic sediments and younger, undisturbed continental clastic rocks. This led to the conclusion that the deposits were genetically related to the unconformity, and resulted in the term 'unconformity-related veins'.

Various genetic models have been proposed. Some involve hydrothermal processes (e.g., Morton and Beck, 1978; Little, 1974; Munday, 1978); others suggest that uranium has been released from the pelites into solutions derived during metamorphism and that these solutions have migrated towards and along the unconformity to locations where they were trapped and the contained metal precipitated (Eupene, et al. 1976; Ryan, 1977.)

Results of uranium-lead and lead-lead dating of these deposits indicate ages that, as expected, are generally significantly younger than the host and cover rocks. This has led to the belief that mineralization post-dates deposition of both (e.g., see Hoeve, 1978; Hoeve and Sibbald, 1978; Kirchner and Tan, 1978).

There is mounting evidence that many of these deposits have only an accidental spatial relationship to the unconformity, and suggests a polygenetic origin reflected by the model presented herein. This model assumes a primary syngenetic concentration of the metal in carbonaceous pelitic sediments to produce protores and, sometimes, ores. Subsequent modification due to metamorphism is probable in many cases and protores may become sufficiently enriched to be economically recovered. Autogenic processes may produce thermal anomalies that, under certain conditions, can be responsible for significant enrichment (Fig. 1).

Environmental Constraints. This sort of uranium concentration can take place only subsequent to oxygenation of the atmosphere at about 2200 Ma.

The mechanism envisaged could operate at any time subsequent to this. Exploration evidence, however, indicates that Aphebian age sediments are the major collectors and, to date, the only ore hosts, although sections of the Cambrian Kolm shales near Ranstad, Sweden, show some economic potential.

Source of Metal. The source of uranium for syngenetic concentration in carbonaceous pelites is generally accepted to be moderately to strongly enriched granites or metamorphic rocks lying within the drainage basin that produced the sediments (Hegge, 1977; Hegge and Rowntree, 1978). Alternatively, in some areas uranium-bearing volcanics, which are penecontemporaneous with the graphitic sediments that host ores or protores,

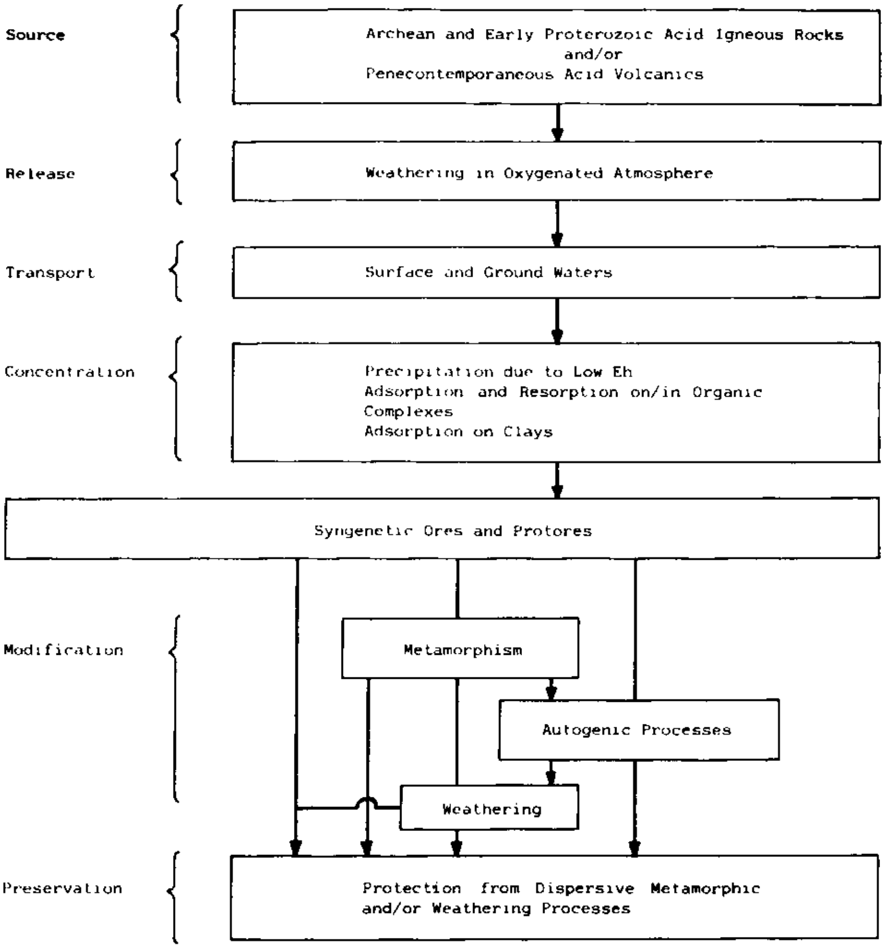

Figure 1 A Genetic *Model for Carbonaceous Pelite Hosted Uranium Deposits.*

appear to be likely source rocks (McMillan, 1976; Gandhi, 1978). The preferred provenance area would have, ideally, gentle topography and a moist, mild to tropical climate without extreme variation in rainfall and runoff.

Release of Metal. Uranium was released to percolating rain water by destruction of uranium-bearing minerals unstable in the prevailing oxidizing environment. Uranium contained in uraninite would be almost totally available, as would most of the metal bound in silicate minerals. Uranium present in certain refractory minerals, such as zircon and monazite, would not have been released in any appreciable quantity and is not considered 'available' or 'labile'.

Uranium released by weathering probably appeared in solution as positively charged complexes in environments having a pH of less than 5 and as uncharged or negatively charged uranyl carbonate complexes at pH greater than 5 (Langmuir, 1978).

Transport. Transportation of the metal from point of release to point of concentration was via surface and ground water flow to the surface drainage systems that also served to transport the clastic sediments which act as host rocks.

Concentration. Uranium in solution in surface waters was carried into a shallow marine, lagoonal environment, where mixing of fresh and salt water would be relatively efficient. In order that precipitation and collection of metal take place, the dissolved metal species must come into contact with organic complexes and clays or migrate into an environment where the Eh is more negative than the upper stability limit of the compatible solid phase, usually uraninite (Langmuir, 1978; Hostetler and Garrels, 1962).

Muds that contain decaying organic material characteristically produce anoxic conditions due to a very high oxygen demand, accompanied by low Eh and low pH (Baas-Becking *et al.*, 1960). These conditions are ideal for the fixation of uranium as UO_2 and in organic complexes. The low Eh of the muds contrasts with relatively more oxidizing Eh of fresh surface waters. A relatively steep potential gradient will exist between the carbonaceous muds and the waters passing above and the mixing of surface runoff and salt or brackish water produces a weak electrolyte. Migration of positively charged metal-bearing complexes from the water into the sediment may account for a significant portion (50% to 60%) of the uranium collected in this environment.

The ideal concentration locality would be a back-reef or barrier island lagoonal environment in which large volumes of surface water could flow slowly at shallow depth across an extensive zone at intertidal and sub-tidal mud flats.

The degree of concentration of uranium in these sediments from solution is a function of the volume of water exposed to the hosting material through time, the amount of metal in solution and the efficiency of the collecting process. Since these factors are likely to vary from one part of a sedimentary basin to another, local variation in the degree of uranium enrichment is to be expected.

The clarke of a carbonaceous pelite probably reflects the level of absorption of uranium on clays during the weathering and transportation process. It is unlikely that significant additional uranium is introduced into the clays immediately after deposition or during early diagenesis because of low permeability in argillaceous sediments at these stages of development.

Enrichments of metals in clay-rich carbonaceous sediments probably indicate rather special conditions in both the local drainage area and in the area of metal accumulation. Concentrations of economic interest will not necessarily be found in pelitic horizons that have an anomalously high abundance of uranium. Conversely, similar rocks shown to contain only 'average' uranium abundances may have local areas of enrichment worthy of exploration attention.

If the constraints and requirements discussed above are satisfied, syngenetic ores or protores may develop.

Modification. Sediments, ores and protores peculiar to the environments under consideration, may be preserved through time relatively unchanged. Others may be modified by a variety of processes and some may be destroyed.

Weathering. Carbonaceous pelite-hosted uranium concentrations may be exposed at surface and undergo weathering at any time subsequent to their deposition. Enrichment is possible below the limit of free oxygen in moist environments.

In wet-dry or arid environments, stable phosphate minerals may develop at surface and sometimes also at considerable depth. While economic concentrations apparently due to weathering of carbonaceous pelites are not known, these rocks would appear to be particularly favourable sources for weathering-related secondary enrichment.

Metamorphism. Metamorphism to greenschist facies grade will produce graphite from organic material in the sediment (Stapleton, 1978). Although organic complexes may contain appreciable quantities of uranium, the crystal lattice of graphite will not accommodate any foreign elements other than boron and uranium will be expelled. Dehydration of clays during metamorphism will also tend to expel metals and, in addition, produce aqueous fluids suitable as transportation media. These fluids may carry uranium (and other metals) into zones of lower confining pressure where precipitation and, possibly, concentration can take place.

Metamorphism strong enough to cause local development of pegmatoids will result in remobilization and possibly concentration of metals in the mobile phases (e.g., Charlebois Lake, Sask., Rössing, South West Africa). However, dispersion of major syngenetic concentrations of uranium by metamorphism, short of complete re-melting of the sedimentary pile, is unlikely.

Low-temperature thermal metamorphism resulting from heat flow anomalies also holds most intriguing possibilities. Such thermal anomalies may be related to steeply inclined graphitic zones and these may play some role in redistribution or enrichment of ores and protores.

Autogenic Processes. Remobilization of minerals that have been stable in any geologic environment requires expenditure of energy which may be supplied by the processes noted earlier.

There is an additional source of energy available for modification of radioactive ores, the heat of radiogenic decay. Historically, consideration of this energy has been confined largely to relatively small deposits of the sandstone type. The volume of ore available in these deposits is restricted and the geometry of the ore zones is not conducive to temperature rise.

In other situations, however, such as the high-grade deposits of the Athabasca basin type, or in large lower grade bodies that are steeply inclined, radiogenic heat becomes important. Convection cells driven by the heat of radiogenic decay may be responsible for redistribution of uranium into overlying rocks as at Midwest Lake, Saskatchewan (Tilsley, 1980b).

If one considers a situation where a tabular syngenetic uranium concentration has been folded into a nearly vertical position, the cross-section through which the thermal energy of radioactive decay can be transmitted toward surface would be greatly reduced. Temperature rise would result. The magnitude of this temperature rise is a function of the volume of uranium present, the horizontal cross section of the mineralized zone and the depth of cover. For example, the Jabiluka II ore body contains about 200,000 tonnes U. It is steeply inclined in cross section. Assuming 1,000 metres of cover, a temperature in the central part of the ore zone of 60° to 90° C above ambient is possible. The local temperature anomaly could be responsible for initiation of a pore water convention cell (Fig. 2).

If we assume an ambient temperature of about 275° C and an internal temperature rise of 60° C, the centre of the zone might attain a temperature of 335° C. In higher temperature environments, such as develop during regional metamorphism, say greenschist facies grade, the solubility of UO_2 in aqueous solutions reaches a maximum. Temperature rise above about 270° C at 750 bars results in a rather rapid drop in solubility. Reference to Lemoine's data (1975) suggests a drop in solubility of UO_2 at 335° of about 40% from the maximum solubility at 265° C (Fig. 3).

Fluids circulating from the fringes of the ore body would therefore tend to collect uranium in the low temperature zone and deposit UO_2 toward the centre of the ore body with rising temperature. Depletion of UO_2 might also occur in the upper part of the mineralized zone as fluid temperature fell toward 270° C. If additional temperature drops, say to 200° C, were experienced outside the ore zone, a reprecipitation of UO_2 would be expected.

There is a multitude of possibilities in regard to redistribution/concentration by this mechanism. Leaching or precipitation of UO_2 in this model is strictly temperature dependent. Location of the minerals redistributed in this manner is simply a function of fluid flow path and palaeo-temperature patterns.

The temperature of the ore body is the critical factor in this concentration/dispersion model. In any given ore body,

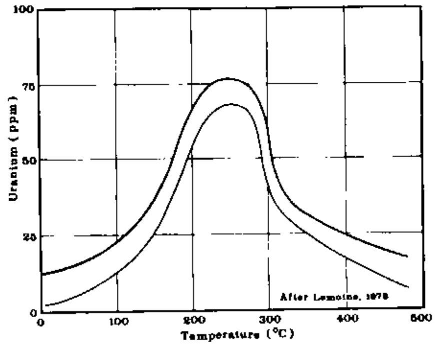

Figure 3 *Solubility of UO_2 at 750 bars*

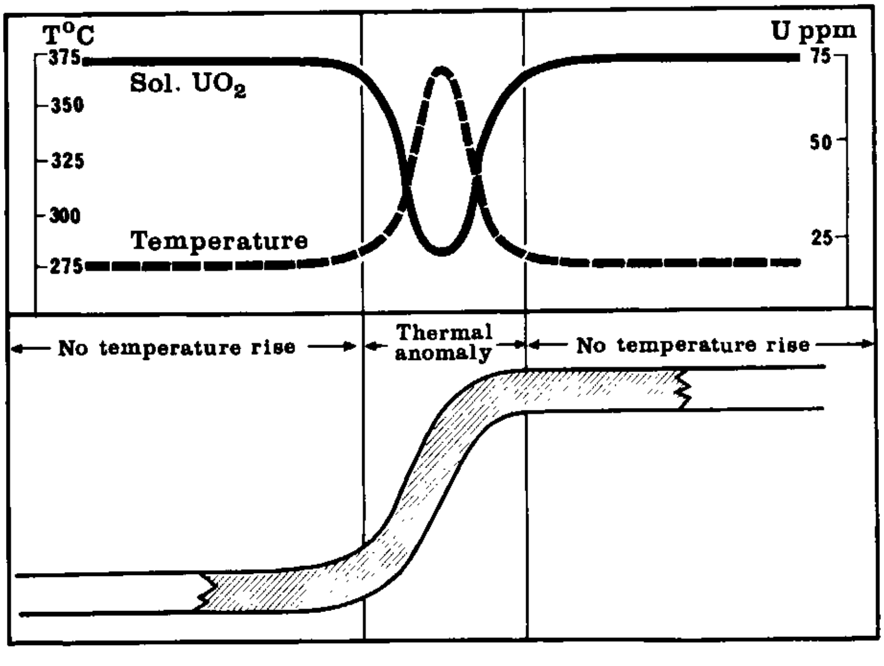

Figure 2 *Relationship between attitude of mineralized body temperature rise and uranium solubility.*

variation in temperature due to the process described is a function of depth of cover. The periodic redistribution of uranium within these ore zones as suggested by the spread of radiogenic age determinations, may reflect changing cover thickness due to sedimentation and erosion rather than assumed regional metamorphic or intrusive events for which there is often little evidence (Table I).

Preservation. Preservation of this type of deposit and any of its sub-types depends upon protection from the extremes of the modification processes discussed. Weathering and metamorphism to the level of anatexis are the most efficient destructive mechanisms.

Fluid convection driven by geothermal anomalies of one type or other could conceivably cause total dispersion of a mineral concentration. However, we know of no examples of either total or partial depletion of mineralization attributable to such activity. Autogenic temperature rise that might be responsible for driving a convection cell at temperatures compatible with redistribution of UO_2 could not be maintained beyond some minimum residual grade. Total self-destruction is not possible.

Discussion

This proposed class of uranium deposit includes such major ore bodies as Jabiluka II and Koongarra in the Northern Territory, Australia, near-ores at Ranstad, Sweden, and numerous enrichments in carbonaceous pelites of post-oxygenation Aphebia age and younger.

Syngenetic concentration of uranium in carbonaceous pelites deposited in relatively restricted shallow-water environments appears to be a significant ore and protore forming process. Protores may be subsequently enriched more or less in situ due to regional or local thermal metamorphism or by secondary concentration related to weathering or to autogenic processes.

Exploration experience has shown that post-oxygenation Aphebian age carbonaceous pelites are the most favourable rocks for this type of uranium concentration. Younger carbonaeous pelites which have been subjected to conditions similar to those that produced ores in Aphebian age rocks would appear to be attractive exploration environments. But, to date, there are no discoveries in younger rocks of economic mineralization similar in scale to those known in the Northern Territory, Australia.

Table I *Autogenic Enrichment of Syngenetic Uranium Ores*

Genetic Sequence:	
Stage I	Syngenetic concentration of U in carbonaceous pelites
Stage II	Metamorphism and folding: Ambient T ≃ 250° C
Stage III	Development of thermal anomaly due to radiogenic heat in steeply-dipping mineralized body. Internal temperature, say, 335° C
Stage IV	Circulation of pore fluids. Collection of UO_2 from fringes of temperature anomaly. Precipitation of UO_2 toward interior of mineralized zone
Stage V	Reduction of ambient temperature due to cessation of orogeny responsible for metamorphism or reduction of cover rock thickness by erosion
Subsequent Acitivity	Possible re-initiation of activity due to increasing depth of cover. Cessation due to erosion of cover. May be repeated several times.

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