

# Recent mobilization of U-series radionuclides in the Bernardan U deposit (French Massif Central)

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## Abstract

Several nuclides of the  $^{238}\text{U}$  ( $^{234}\text{U}$ ,  $^{230}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{210}\text{Pb}$ ) and  $^{235}\text{U}$  ( $^{231}\text{Pa}$ ,  $^{227}\text{Ac}$ ) decay series have been measured through gamma spectrometry in the U-bearing episyenites of the Bernardan mine (northwest Massif Central, France), in order to study recent mobilizations in the U-deposit and their time-scales. ( $^{230}\text{Th}/^{238}\text{U}$ ) and ( $^{231}\text{Pa}/^{235}\text{U}$ ) ratios show that U has been little affected during the last 350 ky, especially in the U-rich samples. In contrast,  $^{226}\text{Ra}$  was subjected to a general redistribution, with a clear tendency for Ra to be lost from the U-rich samples and added to U-poor samples. The highest Ra excesses are present in low-U samples, and are related to adsorption onto iron hydroxides. Ra appears to have been redistributed within the mineralised episyenitic bodies, which seem to remain in  $^{226}\text{Ra}$ – $^{238}\text{U}$  radioactive equilibrium, a result consistent with the earlier study of Leroy [1984. Episyénitisation dans le gisement d'uranium du Bernardan (Marche): Comparaison avec des gisements similaires du Nord-Ouest du Massif Central français. *Miner. Depos.* 19, 26–35]. As shown by the  $^{226}\text{Ra}$  and  $^{210}\text{Pb}$  data, the Ra redistribution was very active between 8 and 0.1 ky, and this might be a consequence of the hydrological changes in groundwater circulation accompanying the Holocene climate warming.

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## 1. Introduction

The Limousin district (NW French Massif Central) is well known for its disseminated-type U deposits and was one of the most productive area of U ore in France. The Bernardan mine near the village of Jouac (Haute-Vienne) was the last U-mine still exploited in France until its closure in 2001. The genesis of the Bernardan and similar

U-ores located in episyenitic rocks (resulting from extensive hydrothermal alteration of granite) has been studied by several groups (e. g. Leroy, 1984; Respaut et al., 1991; Lancelot et al., 1995; Turpin and Leroy, 1987; Ahamdach et al., 1990; Patrier et al., 1997). All these studies documented recent redistributions of uranium or U-series radionuclides (mainly  $^{226}\text{Ra}$ ) in the mineralised episyenites and their surroundings. Here we present a further example of these mobilizations, and we include data on all nuclides of geological interest of the  $^{238}\text{U}$  and  $^{235}\text{U}$  decay series, which can be measured by non-destructive high-resolution gamma-ray spectrometry ( $^{238}\text{U}$ – $^{234}\text{U}$ – $^{230}\text{Th}$ – $^{226}\text{Ra}$ – $^{210}\text{Pb}$ ;  $^{235}\text{U}$ – $^{231}\text{Pa}$ – $^{227}\text{Ac}$ ).

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The existence of radioactive disequilibria among nuclides in the decay series is an indication of recent fractionation events, usually related to gain or loss of the more mobile nuclides. This open-system behaviour can result from instantaneous or continuous processes. Moreover, in a closed system, radioactive disequilibria can only persist for a limited period of time, about 5 half-lives of the daughter nuclide ( $T_D$ ) in a parent–daughter pair where the parent has a much longer half-life than the daughter, as indicated by the classical radioactive decay equation:

$$(D) = (P) \cdot (1 - \exp(-\lambda_D t)) + (D)_0 \cdot \exp(-\lambda_D t) \quad (1)$$

where  $t$  is the time since parent–daughter fractionation,  $(P)$  and  $(D)$  are the present day activities (number of decays per unit time) of the parent and daughter nuclides respectively,  $\lambda_D$  the decay constant and  $(D)_0$  the initial activity of the daughter nuclide.

By measuring activity ratios for daughter–parent pairs with different half-lives, information on the time-scales of the mobilization processes can be obtained. The existence of radioactive disequilibrium in a given daughter–parent pair allows definition of an upper age limit ( $\sim 5T_D$ ) for the fractionation, if mobilization was caused by an instantaneous process. For a continuous process, the presence of radioactive disequilibrium does not constrain initiation of the mobilization event, but does indicate that open-system behaviour persisted during the last  $5T_D$  interval.

## 2. Geological setting and sampling

The Bernardan district is located in the Western Marche complex in the northwest part of the French Massif Central, and is limited to the north by the roughly West–East trending Marche fault (Fig. 1). The geology of the area has been studied in detail by several authors and is summarised in Patrier et al. (1997, and references therein). U-ores are found in episyenitic bodies, developed within hercynian leucogranites, and usually forming very irregular pipes (Grandprat, 1989). Episyenites are porous rocks resulting from extensive dissolution of granitic quartz by hydrothermal fluids (Michel, 1983; Leroy, 1984). Several authors showed that these episyenitic bodies have undergone multiple alteration episodes characterised by crystallisation of different minerals (Michel, 1983; Leroy, 1984; Grandprat, 1989; Pailhous et al., 1993; Patrier et al., 1997). According to Patrier et al. (1997), three successive stages can be distinguished in the Bernardan episyenites. The first one corresponds to a high-temperature (up to 360°C)

hydrothermal alteration immediately following the formation of episyenites, some 310My ago, and is marked by crystallisation of secondary minerals like K-feldspar, quartz, dolomite and fluorite. The second stage consists of the argillisation of the episyenites, which took place at a lower temperature ( $\sim 100^\circ\text{C}$ ) during the Dogger Epoch (170–140Ma). Argillisation formed in response to infiltration of fluids that originated in sedimentary basins and resulted in concentrating U in the episyenites as U silicates such as coffinite. The last stage corresponds to a remobilization of the U mineralization by alteration during an extended period of time, up to the present day. It resulted in oxidation and formation of high-grade ore near the top of the mineralised episyenitic columns. However, in such a fractured aquifer, the alteration by percolating fluids was very inhomogeneous and associated with fractures and zones of higher porosity within the episyenites. Changes in tectonic stress during the geological history may have caused new fractures to open, offering new fluid pathways and allowing alteration and oxidation of previously unaffected zones. Therefore, late stage remobilization of U mineralization was probably a multi-episodic and strongly heterogeneous process. As a result of this complex evolution, various parageneses of U-bearing minerals are found, either as oxidised (autunite, gummite), or reduced (pitchblende, coffinite) phases including “black products” adsorbed on clay minerals (Michel, 1983).

All samples analysed in this study come from the so-called “amas profond nord”, a northwards dipping complex of episyenitic columns, extending to a depth of more than 700m below the surface (Fig. 2). Most samples come from cores drilled from mining galleries. They have been separated in several groups according to the classification of Patrier et al. (1997), who give a detailed mineralogical description of the samples. It should be noted that all the studied samples were collected from the deep part of the mineralised episyenitic body (more than 250m below the surface), and not from the shallow upper part of the U-deposit.

## 3. Analytical techniques

The samples were crushed via standard grinding techniques. Powdered samples weighing between 15 and 17.6g were sealed in cylindrical Plexiglas boxes (internal dimensions: 54mm diameter and 5mm height), and were analysed using the gamma spectrometry methods described previously by Condomines et al. (1995). The gamma spectrometry instrumentation at the Laboratoire “Magmas et Volcans” in Clermont-Ferrand consists of two CANBERRA Ge detectors facing each

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