Tectonic, geochemical and microstructural context of uranium migration in granites of the Krasnokamensk area, Russia

V.A. Petrov\textsuperscript{1}, M. Lespinasse\textsuperscript{2}, V.V. Poluektov\textsuperscript{1}, S.I. Schukin\textsuperscript{3}, V.N. Golubev\textsuperscript{1}, M. Cuney\textsuperscript{2}

\textsuperscript{1}Institute of Geology of Ore Deposits, Petrography, Mineralogy and Geochemistry (IGEM) Russian Academy of Sciences, Staromonetny per. 35, Moscow, 119017 Russia, vlad@igem.ru
\textsuperscript{2}UMR G2R (7566), Nancy-Université, BP 239, 54506 Vandœuvre-les-Nancy, France
\textsuperscript{3}Priargunsky Industrial Mining and Chemical Operations (PPGKhO), Krasnokamensk, Chita Region, 674665 Russia

The main aim of the contribution is to associate data on fluid flow conditions and radionuclide transport processes, which occur at different spatial-temporal scales in Paleozoic fractured granitic rocks. Objects of examination are three sites at the Krasnokamensk area, Chita region, SE Siberia, Russia including surfaces of the SNF storage potential Sites No. 1 and No. 5 as well as underground openings of the Antey uranium deposit operated by PPGKhO. Data of collaborative four-year in situ observations and subsequent laboratory test are presented. Obtained results are discussed in the context of further modelization of the main features, events and processes existing at different scales arranged in 3D space to quantify the parameters of fluid flow pathways and to assess the dynamics of uranium transfer in variously deformed and altered fractured porous granites.

Introduction

Crystalline rocks, such as granites, are hosted economic uranium deposits and practically viewed as suitable settings for constructing the long-term storage of spent nuclear fuel (SNF) and disposal of high-level wastes (HLW) underground facilities. The evaluation of mineral resources and safety of the facilities directly depends on the identification of the most probable pathways of fluid flow, and spatial-temporal evolution of radionuclide transport conditions. It is known, that permeability of crystalline rocks is controlled by such brittle discontinuities as large fractures and cracks. However for the modelling of fluid flow and transfer of radionuclides in fractured granites it is very important to accurately reflect all scales of the porosity including pores, vacuoles, mineral borders, microcracks, etc. Thus it is necessary to obtain data on “fracture-near fracture” space response on fluid injections as a function of stress, rock alteration processes and time.

Three specific sites at the Krasnokamensk area are of particular interest for studying of fluid flow and uranium migration, namely the SNF storage potential Sites No. 1 and No. 5, and the Antey deposit, located at different places and elevation of the Palaeozoic granite environment outside and inside the Mesozoic Streltsovskaya caldera. The potential Site No. 1 is located in the NW part whereas Site No. 5 is positioned in the SE part of the granitic frame of the caldera\textsuperscript{2}. The Antey vein-type deposit is situated inside the caldera. The deposit is one of the largest in terms of uranium resources in Russia. Uranium mineralization here partly occurs in reducing water-saturated conditions with markings of oxidation and is currently mined by accessible excavations at depth of 700 m from the surface. Antey deposit presents natural analogue of key phenomena and processes in natural systems related to those expected to occur in underground nuclear waste facilities. Thus all three sites offer together an outstanding possibility to provide comparative analysis of tectonic, geochemical and microstructural data for a better understanding of fluid flow conditions and radionuclide transport processes in various granitic environments.

DOI 10.1051/names2007040

Article available at http://names.edpsciences.org or http://dx.doi.org/10.1051/names2007040

115
Methods and Results

The fieldwork procedures applied at the sites and deposit were unified as follows: 1) structural-geological survey including identification of faults and fractures, recording their strike, dip, morphology of planes, aperture, density, origin of their mineral filling, etc.; 2) microtectonic analysis of faults and fractures in outcrops and underground openings; 3) rock sampling for petrographic, mineral-chemical and isotopic-geochemical studies of rock varieties, nature and intensity of hydrothermal-metasomatic and oxidizing transformations; 4) sampling of oriented lumps for petrophysical tests (density, effective porosity, dynamics of water saturation, elastic properties) and microstructural analysis including impregnation with color resin and statistical studies of 2D digital images of pore space structure; 5) sampling of materials for fission-track radiography to search uranium distribution and concentration in mineral components and microcracks of the rocks.

The total investigated area at the Site No. 1 was 42 km². On the whole 20 reference points were studied. The total of 24 oriented samples were taken for lab tests. Three deepest mining horizons (9 - 11) of the Antey deposit were investigated with searching at profiles (up to 50 m) arranging 12 reference points at each horizon. The reference points and 35 oriented samples are located at regular distance from the core of the largest uranium ore-bearing fault. Besides, 3 reference outcrops were investigated at the Site No. 5 positioned across elongation of the main fault pattern where at different places 3 oriented samples were taken. The main preliminary results of field studies and lab tests are as follows.

The area under investigation is situated within the Archaean-Proterozoic anticline that was repeatedly intruded by magmatic melts during Late Proterozoic time with formation of a granitic gneiss cupola subjected to subsequent granitic injections during Caledonian and Variscian tectonic-magmatic cycles. Later on, during Early Jurassic to the Early Cretaceous period of regional tectonic-magmatic activation the central part of the cupola underwent subsidence due to eruption of a large volume of acid volcanic products from several palaeovolcanoes, devastation of the magmatic chamber and, as a result, formation of the piecemeal-type caldera with three main units of volcanic and sedimentary rocks. Concerned granitic Sites No. 1 and No. 5 stayed at the opposite flanks of the caldera and their tectonic positions were determined by the long-lived interblock regional NE- and NW-extended faults bounding the caldera area. Contrary to that the tectonic position of the Antey’s host granites is connected with intrablock N-S fault zones developed within the caldera basement.

Hence, one can expect the evident difference in geochemistry of the rocks compared. According to XRF and ICP-MS measurements provided by three labs on the same sample collection show difference in content of the major constituent oxides (SiO₂, Al₂O₃, CaO, Na₂O, K₂O, Fe₂O₃, MgO) in rocks which form at least three groups in respect to SiO₂ (wt. %): <66, >80 and 74-80. First and second groups are connected with caldera granitic frame (Sites No. 1 and 5) while third one is related to the caldera basement (the Antey deposit). Reasoning from isotopic (U-Pb and Rb-Sr) dating it is possible to preliminary conclude, that first group offers oldest (~800 Ma) biotite-enriched melanocratic gneisses or relics of highly metamorphosed rocks. Preliminary electronic microscopy observations show presence of well-shaped uraninite crystals here. Comparative analysis of geochemical data show that these rocks are notably enriched (ppm) in U (34), Th (220), Rb (845), Zr (834) and La (141), and depleted in Sr (12), Sb (3), Mo (2) and As (30).

Granites of second group are connected with cores of ancient shearing zones. The rocks are enriched (ppm) in As (2043), Rb (373) and Sb (124) while depleted in Th (15), Sr (12), Zr (58), La (12) and Mo (<1). Uranium content is relatively small (~13 ppm). However fission-track radiography data clearly indicate mobilization of significant part of uranium into the shearing cores from the near-zone environment where uranium content is ~7 ppm.
The Antey deposit offers a unique occasion to research variations in geochemistry of caldera basement using the same uranium-bearing fault zone at different levels. Core of the fault at the deepest levels are enriched (wt. %) in SiO₂ (80.3) and Na₂O (3.6), and depleted in K₂O (1.5) and Al₂O₃ (9.1). Contrary to that, at the upper levels content (wt. %) of constituent oxides is SiO₂ (73.8), K₂O (6.5), Na₂O (0.22) and Al₂O₃ (13.7). At the innermost levels the core is enriched (ppm) in U (500-7535), Sr (55-70), Zr (247), Sb (158) and Mo (1076) and depleted in Rb (60) and As (69-85). In contrast to that at the upper part of the deposit the fault core rocks are enriched (ppm) in Rb (243) and As (189-208) while depleted in U (24), Sr (41-45), Zr (76), Sb (9) and Mo (81). These differences in geochemistry for the core one of the deepest uranium-bearing fault indicate various types of hydrothermal-metasomatic alteration of rocks and, probably, diverse redox conditions of uranium migration and accumulation at different levels of the caldera basement.

Since hydrothermal-metasomatic alterations and oxidizing transformations change the entire structure of the rock pore space in transition of stress field then uranium transport mechanisms may vary essentially. In this context, microstructural data provide an additional link between evolution of fluid flow pathways and uranium transfer in variously deformed and altered granites. The best witnesses of palaeofluid circulation are fluid inclusions in healed microcracks within and outside the rock-forming minerals, the Fluid Inclusion Planes (FIP). FIP provide an excellent record of successive episodes of crack initiation and fluid migration. However repeated microfracturing and healing of the rock-forming minerals yield complex superimposed patterns of infilled microcracks. Such patterns are often difficult to interpret due to a lack of suitable chronological criteria. These problems can be documented and solved by coupling deformation studies, detailed examination scales of the relationships between trapped fluids and their host structures at all, and studies of fluid inclusions. Besides, the rock matrix always contains various families of open or partially mineralized microcracks (OC) that form space accessible for meteoric water filtration and uranium accumulation in present-day conditions.

The Anima video screen method was used to obtain data on FIP and OC parameters such as length, orientation, aperture and density. It allowed us to quantify FIP and OC fissural porosity in 2D space in a number of oriented sections. For example, at the Site No. 1 it was determined that orientation most of the FIP-OC families agrees with elongation of the major fluid conductive faults. This indicates a unified mechanism of formation of such different-scale heterogeneities. However a number of FIP families oriented contrary to the main directions is detected at some locations. Analysis of microtectonic data shows that this repartition is connected with inversion of tectonic movements, which took place in Transbaikalia region at the end of Palaeozoic and the beginning of Mesozoic epochs. Thus chronology and properties of transversal fluid inclusions are necessary to verify circulation of uranium-bearing fluids into the granites during consecutive stages of regional tectogenesis. This is particularly important since, in addition to geological indications, the wall rocks of Site No. 1 and No. 5 are similar to type of collisional granites while Antey’s rock resemble to volcanic-arc type granites according to Rb-(Y+Na) and Rb-(Yb+Ta) discrimination.

Discussion and Conclusions

At present interim results of the structural-geological, tectonic, petrographical, mineralogical, geochemical and microstructural investigations of SNF storage potential Sites No. 1 and No. 5, and the Antey uranium deposit located at various conditions into the granitic environment of the Krasnokamensk area are obtained. However these data are not sufficient for quantification of fluid flow parameters and for revealing evolution of uranium migration and accumulation in variously deformed and altered granites due to lack of suitable chronological criteria. Evaluation of geological and geochemical data shows that granites
outside and inside the caldera most likely belong to different types according to their age and tectonic setting. It means reiterated inflow of uranium-bearing fluids into the granites, migration of fluids along the diverse pathways, and deposition of uranium concentrations at different P-T and redox circumstances.

Associating data on fluid circulation conditions and radionuclide transport processes at different spatial-temporal scales in Paleozoic fractured granitic rocks it needs to validate evolution in architecture of fluid flow pathways due to stress field inversion. In this context three approaches of fault structural analysis have to be coupled: geometrical, kinematic, and dynamic. Geometry of the fault zones is complex therefore it is necessary to mark out the fault core (gouge, mylonite, cataclasite, breccia), damaged zone (extensive fracture and vein network) and protolith (wall rock with very low fracture density) in each fluid conductive fault. It helps to organize obtained data in 3D space using, for instance, AutoCad-based computer programs.

Oriented sample profiling across the major fault zones and subsequent lab tests show the high potentiality of combining data of petrography, microstructural (FIP - OC measurements), color resin impregnation (verification and visualization of OC networks) and fission-track radiography techniques for revealing linkage between fracture repartition and uranium distribution in spatial-temporal context. In addition, isotopic dating (U-Pb, Rb-Sr) of rock-forming minerals is needed to obtain time schedule of rock formation and transformation. Besides isotopic-geochemical (\(^{87}\text{Sr}/^{86}\text{Sr}\) and \(^{238}\text{U}/^{234}\text{U}\)) data to assess nature of «water-rock» interactions in different parts of fluid conductive faults is urgent. All these data have to be incorporated into the 3D conceptual and computation geological and filtration-transport models.

Thus, the example of granite environment of the Krasnokamensk area shows long-term circulation of uranium-bearing fluids from Late Proterozoic and Palaeozoic to Mesozoic and probably Cenozoic. With a view to reveal this phenomena it is necessary to provide further research coupling the stress field evolution (microtectonic) studies, estimation of P-T conditions (depths) of the fluid inclusion capture, 3D visualization of architectures of the main pathways at different (from metric to micrometric) scales, and determination of uranium transport mechanisms at site specific oxidizing-reducing conditions. This combination will help us to gather essential data for environmental and natural resource applications.

Acknowledgements

We gratefully acknowledge our colleagues form IGEM RAS, Moscow, CRPG, Nancy and BGR, Hannover for the XRF and ICP-MS measurements as well as for useful comments and critical review of the contribution. This work has been partially supported by grant from Rus. Fund Bas. Res. No. 05-05-64094, CNRS project No. 19079 and ARCUS.

References