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CRYSTAL-CHEMISTRY AND ISOTOPE GEOCHEMISTRY OF ALTERATION ASSOCIATED WITH THE URANIUM NOPAL I DEPOSIT, CHIHUAHUA, MEXICO

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Detailed petrologic studies of weathering or hydrothermal residual products have been used to define ordered distributions of secondary mineral parageneses in space and time. In these alteration systems, clay minerals dominate and the comparison of natural data with experimental results or calculated thermodynamic data may be used to assess the physico-chemical parameters which control these alterations. However, in numerous cases, these estimations are not precise. This is particularly true when secondary alteration minerals are ubiquitous, as for example kaolinite which is stable up to 300°C. Crystal-chemical properties and isotopic compositions of these minerals can be used to precise physico-chemical parameters of alteration. These tracers are specifically useful to study water/rock interactions in alteration systems where clay minerals dominate and where classical tracers like fluid inclusions are absent or too small to be studied.

Such an approach has been applied to an hydrothermal uranium deposit hosted by eocene volcanic rocks in Chihuahua, Northern Mexico (Nopal I deposit).

The Nopal I deposit consists of a "breccia pipe" like orebody (100m high, 20x40 m wide) structurally controlled, at the intersection of several faults. This mineralized breccia pipe intersects the two members of the Nopal formation (44 My) consisting of a lower weakly welded tuff and an upper welded rhyolitic tuff. Detailed petrological study of the deposit has been undertaken to determine the major mineral associations ¹.

Uranium occurs mainly as secondary uranyl silicates which fill the fissures and have been

mobilized from a reduced mineralization made of uraninite associated with pyrite. This uranium concentration is linked to an intense alteration of mineralized rocks and surrounding barren rocks. Two major alteration types have been distinguished in relation with their location relatively to their geological setting :

-a kaolinization and silicification mainly developed in the mineralized zone. Kaolinite occurs as fracture fillings and feldspar pseudomorphs while silicification affects the mesostasis of the tuffs. In the scarce reduced mineralization, the closely association of kaolinite and uraninite indicates that they are comtemporaneous. Kaolinization is also developped in the surrounding barren rocks and is restricted to the upper rhyolitic tuff, where it is less intense at larger distance from the pipe. In the mineralized pipe, ultimate mobilization of silica is expressed as opale coatings in the fissures, where opale recovers all other secondary minerals (clays and uranyl silicates).

- a smectitization affects the glassy matrix while feldspars are weakly altered in the fully argillized sommital part of the underlying tuffs. This smectitization has also been detected in the altered barren rhyolitic tuffs. In this last case, smectite is posterior to kaolinite as smectite particles overlay kaolinite plates.

These secondary alteration minerals are widely encountered in various geological surroundings and cannot be used directly to determine the physico-chemical parameters of the alteration. As clay minerals have large surface area, they easily

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interact with fluids and can memorize some fluids properties in their intimate structure or isotopic composition. Consequently, crystal-chemical and isotope studies of the major secondary alteration minerals have been performed in order to get informations on the origin, temperature and composition of fluids that produced uranium mineralization.

Crystal-chemistry of various kaolinites, defined with respect to their origin (fissure, feldspar pseudomorphs), particle morphology, structural order, has been performed using electron paramagnetic resonance (EPR)². This method allows us to detect paramagnetic impurities, like substituted Fe³⁺, and paramagnetic defect centers. The variations of defect centers concentration are only correlated to the location of the kaolinite in the deposit. High defect center contents correspond to kaolinites in the mineralized area where uranium concentration is maximum. It does not exist any relation between defect center contents and other structural (order-disorder) or chemical characteristics (iron content) of kaolinites. Moreover, two kinds of defect centers, presenting different stabilities, have been detected in these kaolinites : one, present in all kaolinites, is very intense and stable over geological periods; the other one, only present when kaolinites are intimately mixed with uranium bearing minerals, is unstable (destroyed at 200°C). These two types of defect centers have been related to two successive irradiations: the first one occurring during crystal growth of kaolinites from radioactive solutions, the second one arising from uranium-bearing minerals when they are intimately mixed with kaolinite. This knowledge of the parameters governing the formation and stability of the radiation-induced centers in kaolinite indicate that this mineral can be used as an in situ dosimeter for radionuclides contents in alteration fluids 2 . A gradient of concentration in radionuclides at the scale of the deposit is evidenced and it is shown that uranium has mainly circulated inside the breccia pipe.

Isotopic data of oxygen and hydrogen were recorded on major alteration minerals (kaolinite, smectite and opale). Assuming their formation from local meteoric water, we derive formation temperatures of clay minerals. Isotopic signatures of kaolinites indicate an hydrothermal origin with low temperature alteration fluids of about 60°C and a great homogeneity of their composition whatever their origin (fissures or feldspar pseudomorph) and location (inside or outside the mineralized area). These data account for high water/rock ratios inducing a relatively constant isotopic composition of kaolinites. Isotopic compositions of smectites are in agreement with lower temperature crystallization (25 to 50°C), and those of opale give about 35°C. All secondary minerals have constant δD values, inducing a homogeneous composition in hydrogen of fluids during all alteration stages.

These isotopic data reveal a thermal gradient inside the deposit in accordance with the mineralogical successions detected by petrographic data.

In conclusion, petrologic data combined with crystal-chemistry and geochemistry of secondary alteration minerals are interpreted in terms of history and physico-chemical parameters of alteration.

The major alteration stage is a kaolinization, contemporaneous with the reduced uranium mineralization. All kaolinites crystallize from a fluid having the same temperature and origin (meteoric), but radiation-induced defects in these kaolinites indicate different composition in radionuclide contents of these fluids. The source of the mineralization appears not to be magmatichydrothermal in origin, but more likely to have derived from epithermal alteration by a geothermal convective groundwater system.

REFERENCES

1- ILDEFONSE P., MULLER J.P., CLOZEL B. and CALAS G. (1989) - A natural analogue of waste repository in tuff : Evaluation of radionuclide migration by secondary alteration minerals, radiation induced defects and dose rate estimation. GSA Abstracts with Programs, St Louis, 21, 20. 2- ILDEFONSE P., MULLER J.P., CLOZEL B. and CALAS G. (1990) - Study of low alteration systems as analogues of radionuclides release and migration. Engineering Geology (accepted).

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