Nuclear Instruments and Methods in Physics Research A 833 (2016) 15–22

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

Quantitative autoradiography of alpha particle emission in geo-materials using the Beaver™ system

Paul Sardini [a,*], Axel Angileri [a], Michael Descostes [b], Samuel Duval [c], Tugdual Oger [c], Patricia Patrier [a], Nicolas Rividi [d], Marja Siitari-Kauppi [e], Hervé Toubon [b], Jérôme Donnard [c]

[a] IC2MP Equipe HydrASA, 6 Rue Michel Brunet, B35, TSA 51106 Poitiers Cedex 9, France
[b] AREVA Mines, R&D Department, Paris, France
[c] AI4R SAS, Nantes, France
[d] Service Camparis, Université Pierre et Marie Curie, Paris, France
[e] Radiochemistry Laboratory, University of Helsinki, Helsinki, Finland

ARTICLE INFO

Article history:
Received 2 June 2016
Received in revised form
27 June 2016
Accepted 4 July 2016
Available online 4 July 2016

Keywords:
Alpha particles
Uranium
Rocks
Autoradiograph
Radioactive equilibrium

ABSTRACT

In rocks or artificial geo-materials, radioactive isotopes emitting alpha particles are dispersed according to the mineralogy. At hand specimen scale, the achievement of quantitative chemical mapping of these isotopes takes on a specific importance. Knowledge of the distribution of the uranium and thorium series radionuclides is of prime interest to several disciplines, from the geochemistry of uranium deposits, to the dispersion of uranium mill tailings in the biosphere. The disequilibrium of these disintegration chains is also commonly used for dating. However, some prime importance isotopes, such as ^{226}Ra, are complicated to localize in geo-materials. Because of its high specific activity, ^{226}Ra is found in very low concentrations (~ppq), preventing its accurate localization in rock forming minerals.

This paper formulates a quantitative answer to the following question: at hand specimen scale, how can alpha emitters in geo-materials be mapped quantitatively? In this study, we tested a new digital autoradiographic method (called the Beaver™) based on a Micro Patterned Gaseous Detector (MPGD) in order to quantitatively map alpha emission at the centimeter scale rock section. Firstly, for two thin sections containing U-bearing minerals at secular equilibrium, we compared the experimental and theoretical alpha count rates, measured by the Beaver™ and calculated from the uranium content, respectively. We found that they are very similar. Secondly, for a set of eight homemade standards made up of a mixture of inactive sand and low-radioactivity mud, we compared the count rates obtained by the Beaver™ and by an alpha spectrometer. The results indicate (i) a linearity between both count rates, and (ii) that the count obtained by the Beaver™ can be estimated from the count obtained by the alpha spectrometry using a factor of 0.82.

1. Introduction

Natural or artificial geomaterials are likely to contain alpha emitter isotopes. Significant amounts of ^{238}U and ^{232}Th together with their daughter products are commonly found in various rock types. Radioactive disequilibria of uranium decay chains are extensively used as dating tools by geologists [1]. The method is developed for naturally occurring radioactive materials (NORM) but could also be applied for other artificial actinides. For example, homogeneity verification of ceramics (U, Pu pellets), or waste repository in geological formations use radioactive disequilibria studies [2–4].

At the scale of a hand scale sample (a geological thin section), the detailed characterization of the spatial distribution of uranium and thorium series radionuclides is a major objective, especially for some alpha emitters which are commonly undetectable by elemental mapping, such as ^{226}Ra. Such radionuclides are commonly assessed either by bulk measurement (see for instance [5]), or by indirect measurements, e.g. sequential leaching [6,7]. Recent developments regarding ^{238}U decay product measurements in geological samples were performed by ionic probe. Detection and localization of alpha emitters in geomaterials is crucial for mining and environmental applications.

Solid State Nuclear Track Detector (SSNTD) is the main method for the quantitative mapping of alpha particles in a centimetre scale rock section [8–11]. This technique is based on the counting of defects caused by alpha particles in a plastic/polycarbonate film, and the subsequent etching of the film to enhance the defects. This widely used technique is commonly employed for detecting radon

* Corresponding author.

http://dx.doi.org/10.1016/j.nima.2016.07.003
0168-9002/

emanations [12]. SSNTD is rarely applied to map alpha emission from geological rock sections. Firstly, the technique is based on an etching process which has to be stringently controlled to ensure reproducibility. Secondly, due to the structural heterogeneity of geomaterials, alpha emission takes place in clusters [13]. This leads to an overlapping of nuclear tracks coming from emitter-rich grains, resulting in counting problems.

Autoradiography by phosphor imaging was mentioned by [14] as a method for quantitatively mapping alpha emission. However, this technique has not yet been used for mapping alpha particles emission in rocks, because phosphor imaging is also sensitive to beta emission. This is why the use of phosphor imaging to map natural radioactivity in rock remains qualitative [15], even though [16] used phosphor imaging for the quantitative mapping of uranium in corallite thanks to a specific calibration step.

More recently, the use of scintillation-based detectors for mapping alpha emission was described for the surface deposits of tracers [17] and for the study of tracer dispersion in biological cryo-sections [18]. This technique uses the scintillation light generated by interaction between the alpha or beta particle and a scintillating solid (here Eu-doped ZnS) for evaluating the activity of alpha or beta particles in the studied surfaces.

Another technique allowing the mapping of charged particles is the use of the Beaver™ device, a filmless digital autoradiographic system based on a Micro Patterned Gaseous Detector (MPGD) technology originally used in particle physics [19–21]. This device allows the real-time counting of charged particle emission from the analysed surface, performed with very high sensitivity and linearity. This tool has until now been exclusively used for mapping beta particles from biological preparations. As far as we know, digital autoradiography has not previously been tested for quantitative mapping of alpha emission from rock sections.

This paper therefore presents the first original application of the Beaver™ device for mapping alpha emission in geomaterials. As a first step, this contribution presents the main principle of the Beaver™ autoradiographic system, and a method for the selective quantization of alpha particles in a rock section. The method was first applied to the rock sections containing ^{238}U bearing minerals at secular equilibrium. After the observation of alpha emission at the scale of the thin section, the measurement of alpha activity using the Beaver™ was compared to the activity deduced from the uranium content of the uranium-bearing mineral. Secondly, the Beaver™'s alpha particle count was compared with the count obtained using an alpha spectrometer.

2. The Beaver™ autoradiographic method

2.1. The Beaver™ system

The Beaver™ is a real-time digital autoradiographic system based on the use of thin micromeshes in a gaseous detection medium. Samples are immersed in a gas mixture so that particles such as beta or alpha emerging from the surface release their energy by ionizing the gas. Thanks to appropriate electric fields, the electrons created during the interactions are multiplied, drifted and localized onto a 2D pixelated anode. Each disintegration triggering the acquisition electronics is reconstructed and creates an image entry. Acquisition software permits the real-time reconstruction and visualization of the radioactive distribution.

This device is capable of imaging beta or alpha particles with a high spatial resolution. As an example, the typical spatial resolution obtained with samples labeled with tritium reaches 20 μm. Thanks to its MPGD the Beaver's sensitivity is 5.10^{-4} cpm mm^{-2} and is not affected by X or gamma rays. Its linearity covering 5 orders of magnitude allows the visualization of highly contrasted emitting structures [19–21].

2.2. Mapping alpha particles with the Beaver™

The detection structure used in the Beaver™ proved its ability to map beta emission [19]. In geological sections, natural emitters such as ^{235}U, ^{238}U and ^{232}Th produce alpha and beta particles together with gamma and X radiations. Note that other natural long-life emitters found in minerals are extensively employed by geologists for rock dating, such as ^{40}K, ^{87}Rb (which are mainly beta emitters), and ^{147}Sm (which is an alpha emitter).

Due to the low density and small thickness of the detection media used in the Beaver™, gamma and X rays rarely interact and produce a very low background. Alpha and beta particles both ionize the gas atoms of the detection media by Coulomb interaction. However, their ionization tracks are separate because of the difference in ionization densities. Alpha particles create highly ionized straight tracks comparing to the erratic tracks of electrons. Because of this behavior, the energy converted into primary charges per unit path is intrinsically higher for alpha particles than for electrons. By adjusting the amplification gain of the Beaver™, the primary charges produced by electron ionization do not trigger the chamber. Hence only alpha particles are detected and produce an image.

Each position of the tracks' origins is reconstructed and stored in a list file. This gives access to the direct count of alpha emission per unit area. Each image pixel signifies the number of alpha particles emitted during the sample exposure, represented on a color scale. This image is called an alpha map.

3. Studied test samples and experiments

The sample holder of the Beaver™ used for this study was originally designed for biological sections on microscope slides 26 mm wide and 76 mm long. Because thin sections of rock are different in size and thickness from biological ones, the glass support of the section was resized. We used rock preparations of 30 μm thickness, which is the standard thickness for petrographic thin sections, classically used for optical microscope observations. Two main sets of samples have been studied, according to two main experiments.

3.1. Alpha emission and uranium content of uranium-rich minerals

The first examined samples were rocks containing naturally-occurring uranium-bearing minerals. They were selected in order to check if experimental alpha activities measured by the Beaver™ can be predicted by the average alpha emitter content of the material. For sake of simplicity, the study was restricted to uranium-bearing minerals having high uranium content; thorium-bearing minerals were not investigated here. Two sections from Shea Creek, Canada (rock age 1300 My (million years), [22,23]) and Le Bernardan (LB), France (rock age 150 My, [24]) were used to map alpha emission using the Beaver™. These sections were selected for two main reasons. Firstly, they present large U-rich zones: the Shea Creek (SC) section is cross cut by a 1–2 mm thick uraninite vein, and the Le Bernardan (LB) section contains millimeter-sized β-uranophane crystals. Secondly, because of their geological ages, the decay chain of ^{238}U for these samples was expected to be in secular equilibrium. Additionally, the secular equilibrium state of the ^{238}U chain of these two sections was confirmed by alpha spectrometry. The uranium content of each type of mineral was measured by WDS microprobe (Cameca SX100) at Camparis facilities, University Pierre and Marie Curie, Paris. The measured uranium content (wt%) of uraninite and β-