A neutron activation technique based on deferred gamma rays applied to the quantification of fluorine in fluorspar mining samples

M A Rey-Ronco¹, T Alonso-Sánchez² and M P Castro-García²

¹Departamento de Energía, Universidad de Oviedo, Oviedo 33004, Spain.
²Departamento de Explotación y Prospección de Minas, Universidad de Oviedo, Oviedo 33004, Spain.

E-mail: castromaria@uniovi.es

Abstract. In this work the technique of deferred neutron activation to determine the fluoride content in fluorspar mining samples has been used. The results of the analysis carried out by neutron activation of samples from different points of a fluorspar mining concentration plant are shown. These results are compared with the chemical analysis results normally carried out at the plant.

1. Introduction

Neutron activation analysis (NAA) is a useful analytical technique for quantitative and qualitative analysis to determine many elements in various scientific and technical fields. We highlight the speed and reliability of this method.

Neutron activation analysis consists of the bombardment of a sample with neutrons and recording the gamma radiation produced in the sample. The equipment required to carry out activation analysis consists of a neutron source and a radiation detector. Each element has a characteristic radiation decay with a particular intensity, energy and decay time, which are unique, thus allowing the detection, identification and quantification of a specific element in the sample.

There are several ways to perform this technique. This article presents the results of the analysis of gamma radiation detected after removing a fluorspar sample from a neutron field produced by an Americium Beryllium source. This type of neutron activation technique is called "delayed", i.e. measurements are taken during the time when the radioactive decay of nuclear reaction products occurs.

This work is composed of two parts: a theoretical one with the study of the reactions that occur in the activation process of neutron activation between neutrons from the Americium Beryllium isotope source, and the elements present in the mining samples, and an experimental one with the design of a prototype to carry out the irradiation and reading.

2. Theoretical study

The theoretical study previous to bombarding the fluorspar sample with neutrons consists of determining the constitutive elements in a mineral sample and analyzing the features of the reactions of each one of these element to neutrons [1]. The fluorine is only found in fluoride; therefore its
determination serves to analyze the amount of fluoride in the sample. There are 12 elements together with the fluorine that can be found in the mineral samples.

Because of the availability of an Americium Beryllium source with a known neutron energy spectrum, the theoretical study is focused on possible reactions of the isotopes of the 12 elements that are contained in a sample, to neutrons in this energy range (3-10 MeV). In total, 468 reactions from database EXFOR [2] have been analyzed.

Firstly, among the possible reactions, we rejected those without a significant cross section for the energy of the available neutrons (fast neutrons). Secondly, we selected those which lead to the emergence of a radioactive element from among the reactions not previously discarded. From this selection process, 29 radioactive products were obtained.

One of the reactions produced is $^{19}$F(n, α)$^{16}$N. The $^{16}$N originating in the process is radioactive and decays to produce $^{16}$O emitting gamma rays with an energy of 6128 MeV and a half life of 7.13 s [3]. The energy of this radiation is the greatest of the theoretical spectrum emitted by the sample so it is easily distinguishable, and valid as a means of analysis.

On the other hand, in a fluorspar sample, the $^{16}$N only originates from three types of reactions:

(a) $^{16}$O(n,p)$^{16}$N, (b) $^{17}$O(n,d)$^{16}$N and (c) $^{19}$F(n,α)$^{16}$N.

Neutrons which come from an Americium Beryllium source have less energy than that needed to produce $^{16}$N from the oxygen present in the sample. Therefore, the $^{16}$N only comes from fluorine. Hence the dependency between the radiation measurement of 6128 KeV and the fluorine concentration in the sample is established.

We need to pass to the experimental phase to determine whether radiation could be detected by a conventional gamma radiation detector of NaI, i.e, if neutron activation is high enough for this technique to be used in the grade control in this mining operation.

3. Prototype design
The previous study affords the knowledge of the basic parameters needed to design the neutron activation technique using a detector NaI (Tl) 2" in diameter and 2" in length, and a multichannel analyzer Canberra and Americium Beryllium neutron source.

The operation procedure is as follows: (1) introduction of the sample into a sample container, (2) displacement of the sample container, guided by the hollow guide to a position facing the source, (3) activation sample, (4) moving the sample to its place in front of the detector, (5) reading of the irradiated sample (figure 1). The duration of the whole process must be minimized without losing accuracy in all readings collected from the irradiated sample.

![Activation and reading positions of an irradiated sample.](image-url)
It was found that in order to activate the sample adequately (figure 1), the distance between the source and the sample should be as short as possible \cite{4}.

On the other hand, it is shown that between the detector and source there should be a lead wall to eliminate the gamma radiation from the source which produces a high background signal. Also, the most suitable times for irradiation, displacement and reading were established.

Using this prototype measurements were made on dry fluorspar samples of grades between 4.20 and 97.20\% in CaF\(_2\) and variable masses between 50 and 450 g. This design allowed:

- the existence of a proportionality between fluorspar grade and the sum of counts produced in the range between 4 and 6 MeV, including the escape peaks, in a scintillation detector with a gamma radiation of 6128MeV \cite{5},
- the number of counts in the range of energies considered was not large, and the reasons for this low reading were due to the equipment used and that,
- if a large number of spectrum samples with the same grade, mass and registration were added together, the spectrum obtained was clearly of \(^{16}\text{N}\). This was equivalent to having accomplished a cyclic activation.

![Figure 2. Comparison between the results of chemical and neutron activation analysis.](image)

Finally, the signal reaching the detector from the fluorspar sample previously activated by neutrons has been modeled. To do this, the two phenomena that occur in this procedure were considered. Both the flux of neutrons and gamma radiation produced have to cross the space through the sample, and thus the neutron flux is reduced and the gamma radiation influencing directly on the number of counts received at the detector is weakened. A regression between grades identified by chemical analysis and neutron activation has been carried out and its results are illustrated in figure 2.

4. Conclusions
It is possible to use delayed neutron activation analysis to determine fluoride grades of fluorspar samples from a concentration plant using the reaction \(^{19}\text{F}(\text{n},\alpha)^{16}\text{N}\) an Americium Beryllium source of 1Ci activity and a 2”x2” NaI(Tl) detector. The \(^{16}\text{N}\) originated in the process is radioactive and decays to produce \(^{16}\text{O}\) emitting gamma rays with an energy of 6128 MeV and a half life of 7.13 s. It has been
shown the $^{16}$N is from fluorine. It is thus demonstrated there is a dependency between the energy of gamma-rays emitted from 6128 KeV and the fluorspar concentration in the sample. A prototype that allows neutron activation analysis in fluorspar samples has been designed, whereby a manual sample displacement system between positions is used. The signal reaching the detector from an activated fluorspar sample has been modeled. Hence the conclusion was reached that neutron activation analysis is suitable.

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