

## New models for carrying out cyclic neutron activation. Discussion of the theoretical response.

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**Abstract.** This paper studies two specific procedures for analyzing mining samples through a neutron activation technique called DGNAA (Delayed Gamma Neutron Activation Analysis). This particular study is part of a broader line of research, whose overall objective is to find the optimal procedure for analyzing the fluorite content of samples taken from different parts of a fluorite concentration plant, using the DGNAA method [1-2]. The mining sample is fluor spar, which contains other minerals in addition to fluorite, such as silica, barite, iron oxides and silicates. The main contribution of the article is the development of a new method for determining the fluorite content in minerals and the increase of sensitivity in respect to the symmetrical method and single-cycle activation.

### 1. Introduction

Neutron activation consists of bombarding a sample with neutrons, and recording the spectrum of radiation that is produced. In previous articles [3-5], this research team demonstrated that the fluorite grade in a sample is directly proportional to its fluorine grade, which is, in turn, directly proportional to the quantity of  $^{16}\text{N}$ , produced in the reaction  $^{19}\text{F}(n,\alpha)^{16}\text{N}$ . The team found that there was no interference in either of the following two important aspects: there is no other reaction between the elements in the sample and the neutrons that could produce  $^{16}\text{N}$ , and there is no other radioactive product that emits gamma rays at around 6128 keV, which is the energy of the gamma rays emitted by  $^{16}\text{N}$ . The procedure followed in prior studies consisted of a single activation process called "monocycle activation," which was made up of two phases: a neutron bombardment phase, and a phase for counting the gamma rays emitted by the sample upon irradiation. However, this procedure yielded a low level of fluorine activation, which in turn led to a poor count of the gamma rays emitted (one count represents a 0.5% of grade mineral) [6].

To overcome this drawback, two theoretical models of cyclic activation have been designed [7], with  $n$  activation and counting cycles called "sym cyclic neutron activation" and "asym cyclic neutron activation," the characteristics and results of which are discussed in this article.

### 2. Theoretical models of cyclic activation

Figures 1 and 2 are graphic representations of the different models: the horizontal axis shows experimental time  $t$  in seconds, while the vertical axis shows the evolution of the concentration of the



radioactive product, in this case  $^{16}\text{N}$ . While the sample is being bombarded with neutrons, the concentration of  $^{16}\text{N}$ ,  $g(t)$ , increases according to a known law which is given by the expression:

$$g(t) = k \cdot (1 - e^{-\lambda \cdot t}).$$

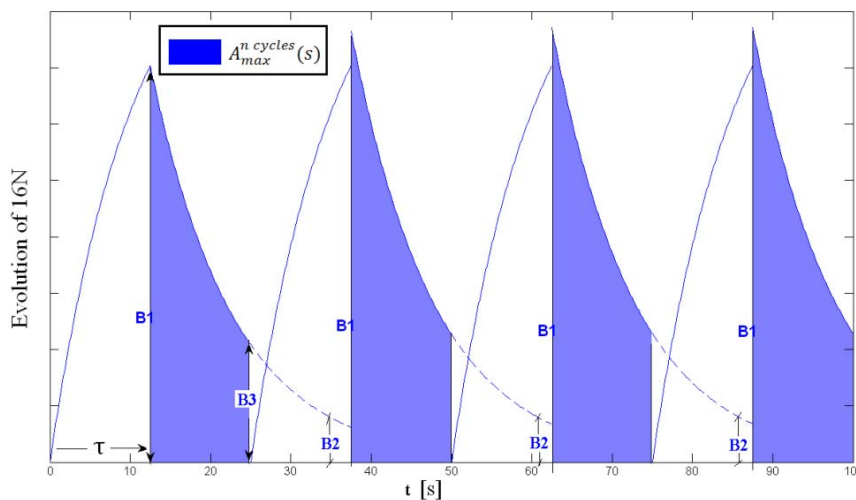
Where  $\lambda = \frac{\ln(2)}{T_{1/2}}$ , in which  $T_{1/2} = 7.13\text{s}$  is the half-time of the  $^{16}\text{N}$  and  $k$  is a parameter that depends on several factors, among them, the concentration in the sample of the element under study ( $F$  in this case), the type and intensity of the neutron source, the cross-section of the reaction, or the spatial arrangement of the device's components. While the mineral sample is in front of the neutron source, the concentration increases according to this exponential expression. This time is called "activation time," and it is represented by  $\tau$ . Once the sample is removed from the radiation source and placed in front of the radiation detector, the  $^{16}\text{N}$  concentration diminishes according to a function that depends on the initial concentration, which is  $k \cdot (1 - e^{-\lambda \cdot \tau})$ . This decay function is:

$$h(t) = k \cdot (1 - e^{-\lambda \cdot \tau}) \cdot e^{-\lambda \cdot (t - \tau)}$$

The sample is left in front of the gamma ray detector during the so-called "counting time." For the energy interval emitted by  $^{16}\text{N}$ , namely around 6000 keV, the reading for a given instant is proportional to the concentration of  $^{16}\text{N}$  at that instant. The gamma ray detector logs the spectrum accumulated during this time, and the log for the energy interval mentioned above is proportional to the shaded area in Figure 1, i.e. to the integral of the function  $h(t)$  over the period corresponding to the counting time. In this one-cycle study,  $A = \int_0^T h(t) \cdot dt$ , for each value of  $T$  the function  $A$  has a maximum that it is produced in an activation time  $\tau$ , which mathematically it is found when we derive the previous equation and equal it to zero, being  $T = \text{cte}$ , it was concluded that  $\tau_{\text{optimo}} = T/2$ , the maximum area occurred when the activation and counting times were equal.

The characteristic trait of cyclic activation is that after the first cycle the sample is again irradiated with the neutrons. At the end of the second cycle of irradiation, the total concentration of  $^{16}\text{N}$  is the sum of the residual concentration from the first cycle (B2), plus the concentration produced during the second cycle (B1 is kept continuous). In sym cyclic activation, the activation time and counting times are equal for each of the  $n$  cycles. Under these circumstances, the area for the total experimental time  $T$  and  $n$  cycles is proportional to  $A_{\text{max}}^{*n \text{ cycles}}(s)$  which is defined by:

$$A_{\text{max}}^{n, \text{cycles}} = \frac{n \cdot k}{\lambda} \cdot (1 - e^{-\lambda \cdot T/2 \cdot n})^2$$



**Figure 1.** The sym cyclic process.

The gain  $G$  achieved by sym cyclic activation over single-cycle activation, for the same total experimental time  $T$ , depends on both  $T$  and  $n$ , and is defined by the expression:

$$G = n \cdot \frac{(1 - e^{-\lambda T/2n})^2}{(1 - e^{-\lambda T/2})^2}$$

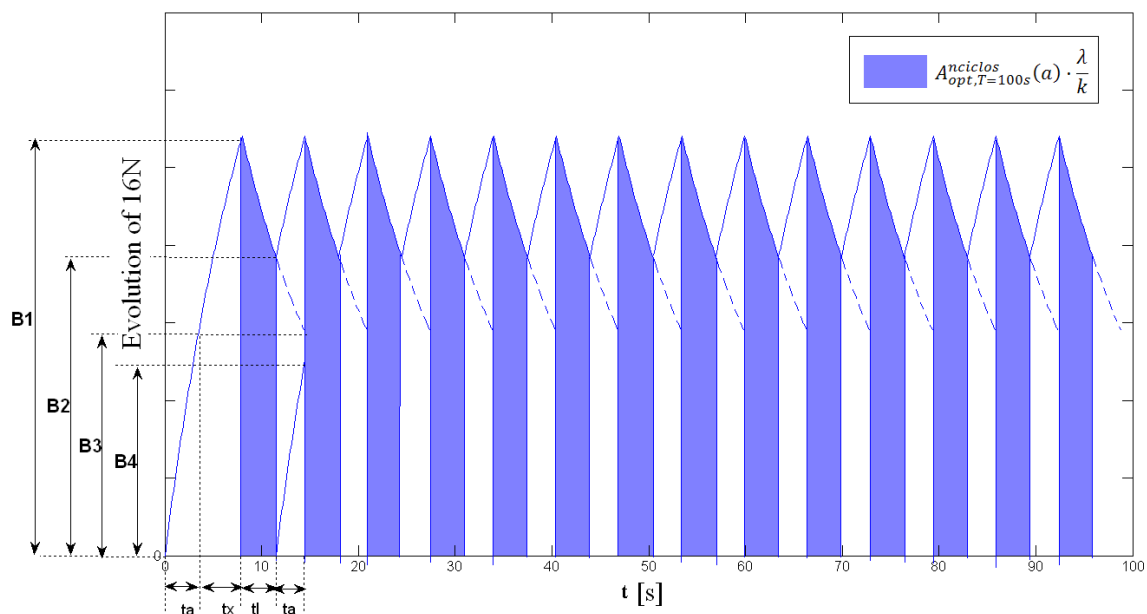
As such, for each experimental time  $T$  there is an  $n$  at which  $G$  is maximized,

which it is obtained when  $\frac{dG(n)}{dn} = 0$ . As such, for each experimental time  $T$  there is an  $n$  at which  $G$  is maximized. For example, for  $T=100s$ , cyclic activation's gain is maximized at  $n=4$  cycles, and the value of this gain is 2.010. Table 1 shows the theoretical gains for different values of  $T$  and  $n$ , which are obtained by replacement in the previous equation.

n	<i>Theoretical Vs n=1</i>			
	<i>Time of process, T</i>			
	300	200	160	100
2	1.997	1.969	1.921	1.690
3	2.954	2.770	2.570	1.961
4	3.794	3.327	2.940	2.010
5	4.473	3.672	3.114	1.963

**Table 1.** Theoretical gain of cyclic activation over single-cycle activation.

The process called asym cyclic activation is characterized by having an initial activation cycle with a higher activation time than subsequent cycles. It could be said that the first activation cycle is made up of two parts: a "normal" activation time  $t_a$ , where the  $^{16}N$  concentration is  $B_3$ , and it is equal to the reading time  $t_l$ . In this moment, the concentration is reduced from  $B_1$  to  $B_2$ , and it is repeated in all the cycles, plus an "extra" activation time  $t_x$ , which pushes the concentrations of  $^{16}N$  up to a higher range from  $B_3$  to  $B_1$ , than in sym cyclic activation, as can be observed in Figure 2.



**Figure 2.** The asym cyclic process

The area in the asym cyclic activation process is given by the expression:

$$A_T^{*n \text{ cycles}}(a) = n \cdot (1 - e^{-\lambda \cdot (t_a + t_x)}) \cdot [1 - e^{-\lambda \cdot t_l}]$$

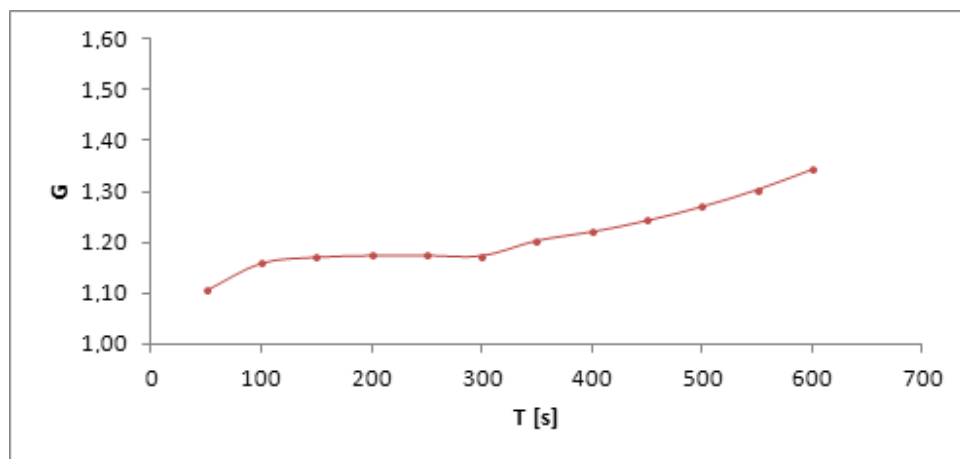
where:  $T = n \cdot (t_l + t_a) + t_x$

The comparison between the two processes is carried out under their respective optimal conditions for each one. In order to determine exactly what these optimum conditions are, it was necessary to first determine the number of cycles, activation and counting times, and the extra time for the first asym cycle that, for a determined experimental time  $T$ , will maximize the expression that defines the area mentioned above. Mathematical methods were used, the results of which are shown in Table 2. The areas for each type of activation have been called  $A_{T,max}^{*n\text{ cycles}}(s)$  and  $A_{T,max}^{*n\text{ cycles}}(a)$ , and for each experimental time we have indicated the theoretical value of the area, the times, and the number of cycles at which the area is maximized. In the last column we can see the improvement that asym cyclic activation achieves over sym cyclic activation.

$T [s]$	<i>sym cycle</i>			<i>asym cycle</i>					$G$ <i>asym/sym</i>
	$A_{T,max}^{*n\text{ cycles}}(s)$	$\tau [s]$	$n$	$A_{T,max}^{*n\text{ cycles}}(a)$	$t_l [s]$	$t_a [s]$	$t_x [s]$	$n$	
50	0.9729	12.5	2	1.0750	12.47	11.3	2.558	2	1.10
100	1.9679	12.5	4	2.2800	16.72	16.04	1.82	3	1.16
200	3.9579	12.5	8	4.6460	16.61	16.43	1.859	6	1.17
300	5.9479	12.5	12	6.9800	16.71	16.43	1.838	9	1.17
400	7.9379	13.33	15	9.6900	15.5	15.12	2.039	13	1.22
500	9.9279	13.16	19	12.6100	16.61	16.6	1.866	15	1.27
600	11.9179	13.04	23	16.0100	15.16	14.74	2.098	20	1.34

**Table 2.** Theoretical gain of *asym* cyclic activation over *sym* cyclic activation

The theoretical gain values for each experimental time  $T$  are graphed in Figure 3, where it can be seen how they increase with time  $T$ .



**Figure 3.** Graph showing the theoretical gain of *asym* cyclic activation over *sym* cyclic activation.

### 3. Conclusions

In conclusion, cyclic neutron activation provides a significant improvement over single-cycle neutron activation. Within the cyclic category, asym cyclic activation produces a greater concentration of  $^{16}\text{N}$  than sym cyclic activation, meaning that the fluoride grade in the sample can be better determined, as the number of counts increases.

### 4. Acknowledgments

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