# Boron content determination in ore samples using Geant4-simulated PGNAA and MCLLS algorithm

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# 01 OVERVIEW

Prompt Gamma Neutron Activation (PGNAA) based online elemental analyzers have gained widespread adoption in the cement industry as a non-destructive and rapid method for determining the elemental composition of bulk raw materials. Currently, more than 500 cement facilities worldwide rely on PGNAA-based online analyzers for precise measurements. The growing demand for such analyzers is driven by the need for efficient and accurate quality control processes in cement production. Traditional methods of measuring elemental composition often require time-consuming and expensive sample preparation, increasing the overall cost of production. PGNAA-based online analyzers provide a precise measurement without the need for extensive sample preparation, thereby saving valuable resources. As a result, these analyzers have become indispensable tools for cement manufacturers seeking to optimize their production processes and ensure consistent product quality

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# PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS

Neutron Activation Analysis (NAA) stands as the top analytical technique among more than 100 different methods. NAA can be classified into two categories based on the timing of measurements: Prompt Gamma Neutron Activation Analysis (PGNAA), where measurements occur during irradiation, and Delayed Gamma-Ray Neutron Activation Analysis (DGNAA), where measurements occur after radioactive decay. PGNAA offers a non-destructive method that provides overlapping-free spectra, minimizing interference effects. One of the most significant applications of PGNAA involves using 14 MeV neutrons to trigger the 16O(n,p)16N reaction. F, Mg, Al, Si, Cu, Fe, P, and Zn elements are frequently determined using this technique. PGNAA's ability to provide precise and accurate measurements has made it an indispensable tool in the field of analytical chemistry.

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# PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS

In PGNAA, the elemental composition of a sample is determined by measuring the prompt gamma radiation emitted from the sample following neutron capture. The emitted gamma rays provide information on the elemental composition of the sample. Furthermore, PGNAA has become a standard analytical technique for determining elemental composition in various industries, including cement production, mining, and environmental monitoring. The sensitivity and precision of PGNAA make it an essential tool for quality control and research purposes in different fields, including analytical chemistry, materials science, and nuclear engineering. With its non-destructive nature, high sensitivity, and minimal sample preparation requirements, PGNAA has become a powerful analytical technique in the field of neutron activation analysis.

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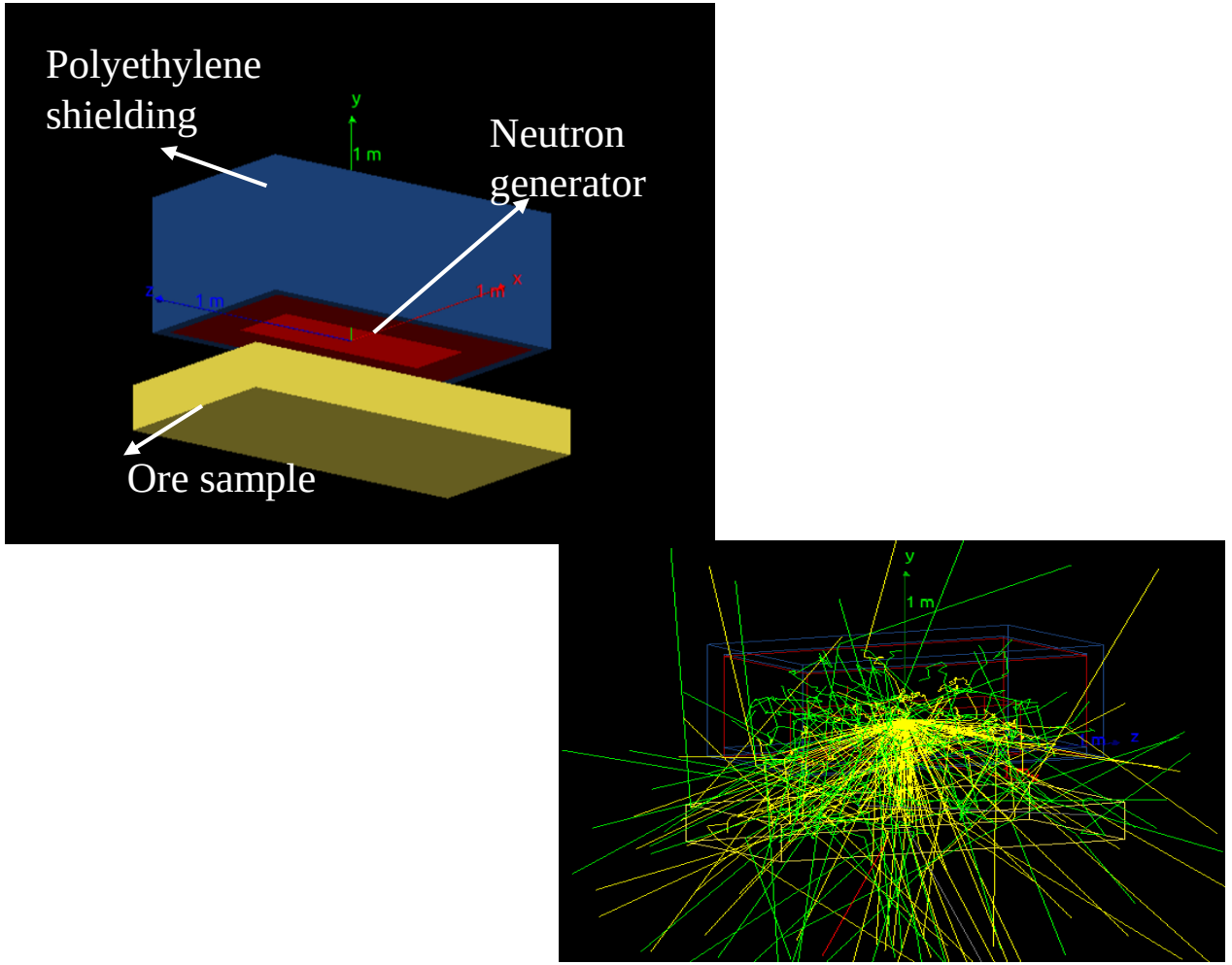
# PROMPT GAMMA NEUTRON ACTIVATION ANALYSIS

NAA is a nuclear analytical technique that involves bombarding a sample with a beam of neutrons generated from a neutron source. The neutrons undergo inelastic scattering and capture reactions, producing characteristic gamma rays emitted from the sample. These gamma rays are detected and analyzed using a spectrometer, which enables the identification of neutron-capturing elements based on the energy of the emitted gamma rays. The intensities of the peaks at these energies indicate the concentration of the elements in the sample, enabling the determination of elemental composition in various materials with high sensitivity and precision. NAA has found numerous applications in different fields, including materials science, environmental monitoring, and forensic science, where it has proven to be a powerful analytical tool for non-destructive and accurate elemental analysis.

# 03 GEANT4 SIMULATION

In the Geant4 software simulation, irradiations are done by four million neutrons shooting with an energy of 14.1 MeV. Emerging characteristic gamma-rays from both inelastic scattering of neutrons and neutron capture were collected from 0 to 10 MeV with 1024 channels. To speed up the simulation, some unneeded particle tracking is avoided. Also, empirical or semi-empirical parameters could be applied to adjust some detector properties.




Experimental setup of the Geant4 simulation toolkit

# 04 MONTE CARLO-LIBRARY LEAST SQUARE

This mathematical approach is grounded in the concept of evaluating the spectrum of an unknown sample, which is obtained by combining the library spectra of individual elements. The least-squares criterion is employed to determine the most probable values of the relative elemental compositions in the simulated ore samples. Equation (1) represents the summation of the library spectrum for a single element, while Equation (2) defines the least squares norm used to ascertain the most appropriate values of the related elemental contents in the measured Tinkal samples. The underlying assumption of the MCLLS method is that the spectra of an unknown sample can be divided into segments associated with monoenergetic components. With this approach, the library of single-element spectra constructed using LLS can be employed to perform a reverse matrix operation (Wang et al., 2012) for determining the compound compositions within the spectra of an unknown sample.

$$y_i = \sum_{k=1}^{9} a_{ik} x_k + e_i \,, \qquad i = 1,2,3, \dots, 1024 \tag{1}$$

$$M = \sum_{i=1}^{1024} \omega_i \left( y_i - \sum_{k=1}^{9} a_{ik} x_k \right)^2 \tag{2}$$

# 04 MONTE CARLO-LIBRARY LEAST SQUARE

In Equation (2), $\omega_i$ represents the statistical weight assigned to channel i, $y_i$ denotes the count rate in channel i for the spectra of the unknown sample, $a_{xk}$ stands for the count rate in channel i corresponding to the single-element spectrum of element k, and $x_k$ represents the relative content of element k. Within the MCLLS approach, it is assumed that the value of $a_{xk}$ remains constant regardless of the elemental contents. Hence, a partial derivative with respect to a specific relative content $x_k$ can be utilized to determine the minimum value of the parameter M. This is achieved by setting the derivative of M to zero, as shown in Equation (3):

$$\partial M/\partial x_k = \sum_{i=1}^{1024} \omega_i \left( y_i - \sum_{k=1}^{9} a_{ik} x_k \right) a_{ik} = 0 \quad \textbf{(3)}$$

Equation (3) can be reformulated into a matrix model as follows:

$$A^T \omega y - (A^T \omega A) X = 0 \quad \textbf{(4)}$$

By solving Equation (4) for X, we obtain:

$$X = (A^T \omega A)^{-1} A^T \omega Y \quad \textbf{(5)}$$

The elemental contents can be determined using Equation (5).

# 05 RESULTS

In this study, a comparison between the laboratory results and the results obtained utilizing the MCLLS method was done for the unknown sample. The MCLLS technique was used to create a single-element library from the spectra of ore samples. The table below demonstrates how the MCLLS method and Geant4 could precisely quantify the boron oxide level. To demonstrate the capabilities of this method, the contents of the other compounds also were given.

| Compound | Monte-Carlo LLs | LAB | Difference (ABS) | Percentage % |
|---|---|---|---|---|
| **$B_2O_3$** | 27.608 | 26.800 | **0.808** | 3.016 |
| **$Na_2O$** | 10.328 | 11.550 | **1.222** | 10.583 |
| **CaO** | 7.258 | 6.768 | **0.490** | 7.244 |
| **$SiO_2$** | 5.567 | 5.234 | **0.333** | 6.368 |
| **$SO_3$** | 0.138 | 0.054 | **0.015** | 9.629 |

# 06 CONCLUSION

The number of samples used in this study was relatively low compared to an industrial setup, which typically requires 50–100 samples. Despite this, the combination of the library least squares approach and prompt gamma neutron activation analysis with a neutron generator demonstrated accurate and consistent measurement results. However, increasing the number of samples would enhance result accuracy, and extending the simulation time by boosting neutrons would improve library least squares calculations. It's worth noting that the accuracy of the element library, constructed through experimental and Monte Carlo methods, remained satisfactory even with a low number of samples.

Differences observed between the Monte-Carlo-LLs, and lab results can be attributed to several factors, such as methodological differences, variances in structure between the real and simulated samples, fluctuations in detector gain, and variations in the sigma values of the real detector and the broadened histograms.

# THANKS