

A Study on standardization of Neutron Activation Analysis for Geologically Based Artifacts : Evaluation of Applicability to Goryeo Celadon

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1. Introduction

Artifacts excavated from archaeological sites in Korea are composed of various materials. Among these, geologically based artifacts—such as pottery, ceramics, roof tiles, and glass—manufactured from raw materials of geological origin account for a significant portion of the excavated quantity. These inherently reflect the formation environment and geological background of the raw materials in their composition. Geochemical fingerprint can be used as a basis for estimating the origin and production site of the artifacts, and further provide important evidence for restoring social networks and trade relationships. In particular, trace elements such as rare earth elements are used as key indicators in archaeological research because they can reflect regional differences by sensitively varying depending on the geological formation environment. In this regard, neutron activation analysis has the advantages of high sensitivity, accuracy, and precision for elements present in trace amounts, as well as the capability to simultaneously determine multiple elements. Furthermore, from the perspective of conservation science, it is suitable for the analysis of artifacts because it is possible with a very small amount of sample.

In practice, neutron activation analysis has been employed in various studies to classify the characteristics of geologically based artifacts, revealing differences related to their production sites and periods. However, there has been insufficient systematic review of the analytical conditions and quantitative methods for neutron activation analysis suitable for archaeological relics. This results in differences in the range of detectable elements, measurement uncertainty, and quantitative uncertainty depending on the analyst, which may limit the reproducibility of results and the reliability of cross-study comparisons.

Accordingly, this study aims to examine the optimal analytical conditions and quantification methods for each element using geological standard reference materials, and to evaluate the applicability of this methodology by applying it to excavated Goryeo celadon, one of the geologically based artifacts.

2. Materials and Methods

2.1 Materials

The samples used for the study of optimal analytical conditions and quantitative method were 15 geological reference samples provided by the Geological Survey of Japan (GSJ). 8 pieces of celadon excavated from four kilns, For the celadon samples, 28 pieces excavated from four kilns, four kilns, No. 3-4, No. 20 in Sindeok-ri, and No. 17, No. 20 in Jinsan-ri, Haenam, Jeolla Province, Korea, were analyzed by removing the glaze and powdering the body. The Jinsan-ri kiln site is characterized by its chronological succession, as it emerged when the ceramic production groups from the Shindeok-ri kilns relocated to this area.

2.2 Methods

To examine the optimal analysis conditions for each element, two analysis conditions were applied for short-lived radionuclides and three conditions for medium- to long-lived radionuclides, as summarized in the table. The resulting spectra were used to examine the interfering radionuclides and the counting statistical errors under each condition. Using the adopted conditions, quantitative values were derived using absolute, relative, and internal mono standard methods. Accuracy was then verified through comparison with certified values and precision through repeated measurements. Also, regression analysis was conducted to evaluate the relationship between the certified and measured values about each reference sample.

Table I: Analytical conditions according to the half-lives of the radionuclides.

condition	Short – Lived		Medium · Long - Lived		
	1	2	1	2	3
Sample mass	20 mg		50 mg		
Irradiation	10 sec		2 hr		
Cooling	15 min	17 min	1 week	2 weeks	3 weeks
Measurement (s)	120	1200	4800	7200	8400

The analysis conditions and quantitative methods obtained through the previous process were applied to the celadon samples, and the applicability of the methodology to actual artifacts was evaluated using elemental bi-plot, rare earth element patterns, and various statistical analyses (PCA, LDA, and CA).

3. Results

The analyzed radionuclides exhibit stable counting rates under different analytical conditions, depending on factors such as half-life, thermal neutron capture cross-section, and gamma-ray emission probability. The certified values of the reference sample were plotted on the x-axis, and the quantitative values derived through the confirmed conditions for each element was plotted on the y-axis. The slope of the regression line indicating the correlation between the two variables and the coefficient of determination (R^2) indicating linearity were evaluated. As a result, most standard samples yielded values close to 1 for both metrics when using the internal mono standard method, indicating high accuracy in quantification, while the relative method also demonstrated a high level of quantitative performance. Both quantification methods avoid using the thermal neutron flux and thermal neutron cross-sections in their calculations, thereby overcoming the errors associated with these parameters.

Finally, in the process of quantification for celadon using the optimal analysis methodology, the value derived through the internal mono standard method is expressed as a ratio with the internal standard element. Therefore, by additionally performing the relative method for the standard elements, the values could be converted to actual content values. The celadons from Shindeok-ri and Jinsan-ri were well classified by several indicators. In particular, LDA analysis effectively distinguished the celadon from the two regions, with Cs, Sc, and Cr identified as the elements contributing most significantly to the discrimination. As these elements represent rare earth elements and some transition metals, which are known for their immobility and stability [1], the findings validate the use of trace elements in geologically based artifacts for provenance determination and confirm the utility of neutron activation analysis in studying these artifacts.

4. Conclusions

The analytical conditions established in this study provide a methodological basis for provenance research in that they ensure the reliable quantification of various trace elements. Among various quantification methods, the internal mono standard method demonstrates particularly high accuracy and precision for critical trace elements identified in previous studies [1, 2, 3]. This performance is likely attributable to a feature that eliminates uncertainties arising from both external standards and other contributing factors. Application of

the proposed analytical methodology to excavated celadon samples demonstrated that it enables stable classification of kiln-derived specimens exhibiting differences in operational periods and intra-regional locations. In particular, when combined with discriminant analysis, a multivariate analysis that generates discriminant functions by integrating compositional elements and predicts groups, the resulting clustering behavior is even more effective. Accordingly, if this analytical methodology is standardized and applied in subsequent celadon studies, it is expected to contribute to ensuring data consistency and comparability across research contexts.

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