Provenence Analysis of Archeological Stone Fragments using Neutron Activation Analysis

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When French archeologists first discovered a Roman settlement near Metz (Lorraine) during the enlargement of the Metz airport, they assumed the used limestone had been taken from one of the nearby quarries [1]. These quarries – due to their excellent stone quality – had already been exploited by the Ancient Romans on a large scale and stone blocks had been shipped to larger settlements like Augusta Treverorum (Treves) or Mogontiacum (Mainz) on Moselle and Rhine [2].

For a provenance study limestone from the excavated buildings (fragments 1 - 4) were compared to stones from two nearby ancient quarries. Therefore, four stones of the archeological site, five stones of the first quarry and nine of the second one were chosen for neutron activation analysis (NAA). The irradiations of the samples were performed at the TRIGA Mainz.

Since the inhomogeneity of the limestone was taken into account, about 200 mg of stone powder were gained by drilling at three different areas. From these 200 mg an aliquot of 30 – 50 mg was taken for short time irradiation (one minute) in the pneumatic transport system. 130-150 mg was reserved for long term irradiation (one hour). To prevent contamination of the sample during drilling, a high purity wolfram electrode, which had been sharpened, was used (figure 1).

Figure 1: Sampling

Short time irradiation let to the following short lived isotopes: Mg-27, Al-28, V-52, Mg-56 and Sr-78m. Most isotopes were detected after long time irradiation. 24 hours after irradiation Na-24, K-42, Ca-47, As-76, La-140 and Sm-153 were detected. Two weeks later the activity of isotopes with half-lives of several days were plotted (Cr-51, Fe-59, Ce-141, Nd-147, Yb-169, Lu-177, Pa-233, Np-239). At this time the Compton background had ceased to some extent. After additional three to four weeks the last six isotopes, Sc-46, Co-60, Zn-65, Cs-134. Eu-152 and Tb-160, were registered.

The following equation was used to calculate the element concentration based on the isotope's activity a_{standard}. Therefore, single- and multi-element standards with known concentrations c_{standard} were irradiated according to the stone samples.

$$c_{\text{sample}} / c_{\text{standard}} = a_{\text{sample}} / a_{\text{standard}}$$

This way the element concentrations in the stones could be calculated and used for further contemplations. At first the results of the three different areas of each stone were compared to each other. Fortunately, they proofed to be sufficiently homogenous for a further approach. Secondly, correlation diagrams (figure 2) were made. This simple method already illustrated how the samples are related.

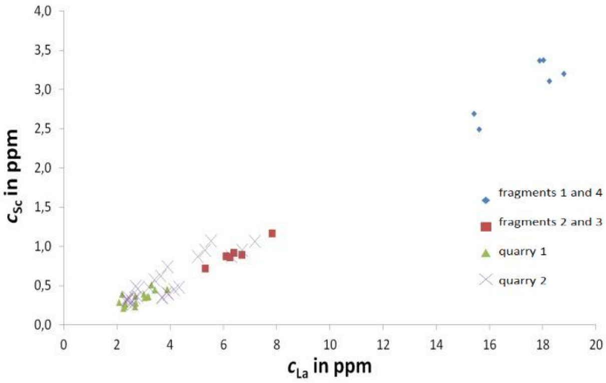

Figure 2: Correlation diagram of La and Sc

It showed that none of the fragments could be assigned to either quarry. At the same time the fragments split up into two different groups, suggesting two diverse origins. The two sampled quarries can hardly be separated. Here, more analyses may be helpful. The results have been unexpected and the fragment's origin is still unknown. Eventually the sampling of other quarries will lead to an answer. Archeologists assume that the precious limestone was ordered by the Roman emperor to more prestigious projects in close provinces and was not to sell at the local market [3].

References

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