Contribution ID: **20** Type: **POSTER**

Contribution to the Roman glass study: analyses performed by PIXE/PIGE, INAA and XRF

Tuesday, 14 June 2022 15:20 (2 minutes)

As with ceramics and metallurgy, glass production is a high-temperature technology invented by ancient civilizations. The material quality and manner of manufacture of glass artifacts provide insight into the technological and social progress of the societies who developed them.
Despite the abundance of Roman glass discoveries in Bulgaria, only three archaeometric studies have been undertaken to date. They were conducted between 1999 and 2021 and mostly involve samples from southern Bulgaria and the country's Black Sea coast. Different instrumental methods were employed to determine the elemental composition: INAA for Roman glass from Odessos (modern-day Varna, Northeastern Bulgaria)(1), PIXE/PIGE for Southeastern Bulgaria samples(2), and XRF for Southwestern Bulgaria samples examined in 2021. The IAEA financed the examination of the largest collection of specimens from Southeast Bulgaria in 2010, which was conducted using PIXE/PIGE technique at "Jožef Stefan"Institute in Ljubljana (Slovenia).
Glass is a supercooled liquid (amorphous solid solution) of alkaline silicate containing sodium or potassium ions, with a divalent alkaline earth metal serving as the curing agent. Throughout the Roman Empire, the primary raw materials for glass manufacture came from natural sources of quartz sand, soda (trona), and limestone. Later, natural soda and limestone were phased out in favor of plant ash as a flux since it contains potassium and magnesium ions. Numerous mineral additions containing iron, manganese, antimony, and copper have been added to the product to color or clear it. Thus, the results of the elemental composition analyses of Bulgarian Roman glass can be summarized as follows:
1. During the Roman Empire, glass production was based on the sodium-calcium-(aluminum)-silicon type of glass.
2. The majority (almost 77 percent) of glass samples analyzed exhibit traits common to European Roman glass, such as the use of antimony and manganese as decolorizing agents and an increased level of impurities later in production. The increased concentrations of oxides of certain basic elements (Al, Mg) in some samples, as well as the observed scattering, indicate a change in raw material sources around the end of the third and beginning of the fourth centuries AD, and/or the expansion of glass production with new production centers, which in practice means less controlled production.
3. The majority of the glass objects discovered at Southwestern Bulgaria were made according to the so-called Middle Eastern formula, but with different raw materials and in a different location than the glass artifacts from Southeastern and Northeastern Bulgaria from the same period.
4. To gain a better understanding of the origins of the Bulgarian Roman glass, it is important to undertake future analyses on a larger number of glass finds from Bulgaria and to compare the results to data from likely raw material sources.

(1) Kuleff, I., R. Djingova (1999) Archaeometric investigation of Roman glass finds from Bulgaria. Berliner Beiträge zur Archäometrie 16, 183-198
(2) Lesigyarski, D., Ž. Šmit, B. Zlateva-Rangelova, K. Koseva, I. Kuleff (2013) Characterization of the chemical composition of archaeological glass finds from South-Eastern Bulgaria using PIXE, PIGE and ICP-AES. Journal of Radioanalytical and Nuclear Chemistry 295, 1605-1619

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Session Classification: Poster session

Track Classification: Track 2: Interpretation, presentation and dissemination of the scientific results