A multidisciplinary study unveils the nature of a Roman ink of the I century AD

C. Stani[1*], L. Gigli[2], S. Pollastri[2], M. Sibilia[4], A. Migliori[4], F. D'Amico[2], C. Schmid[4], S. Licen[5], M. Crosera[5], G. Adami[5], P. Barbieri[5], J. R. Plaisier[2], G. Aquilanti[2], L. Vaccari[2], S. Buson[6] & F. Gonzato[6]

[1]CERIC-ERIC, Basovizza, Trieste, Italy. [2]Elettra-Sincrotrone Trieste S.C.P.A, Basovizza, Trieste, Italy. [3]Nuclear Science and Instrumentation Laboratory, Physics Section, IAEA, Seibersdorf, Austria. [4]Department of Engineering and Architecture, University of Trieste, Trieste, Italy. [5]Department of Chemical and Pharmaceutical Sciences, University of Trieste, Trieste, Italy. [6]Museo Nazionale Atestino, Este, PD, Italy.

chiaramaria.stani@ceric-eric.eu

The purpose of this work was to uncover the real nature and composition of a dry black ink powder found in a bronze inkwell (Fig.1) of the 1st century A.D. It was discovered during the excavation of a cemetery in the locality of Morlungo, Palazzina-Capodaglio, in the municipality of Este in 1878 [1]. Since 2500 BC and up to the thirteenth century AD [2] the *carbon-based inks* were the most common used. They were mainly composed of amorphous carbon obtained from soot charcoal, or bone black [3,4] usually dissolved in a binder, mixed with a small amount of water. During the IV century AD, a new kind of ink, called *iron-gall ink*, emerged. It was obtained by mixing gall-nuts, iron or copper metal sulphates, water and Arabic gum. From the early Middle Ages onwards, it became the most common ink in the history of the western world [2]. However, some recent studies on the chemical composition of inks, already spread on their ancient writing supports (papyri, parchment, or paper) [5–7], have changed this perspective. The importance and the novelty of this work resides principally in the opportunity of directly studying the ink powder, avoiding the interference from the writing support, as well as analysing its container that was fundamental for a correct interpretation of the experimental results. The investigation was conducted through a multi-technical approach, combining several and complementary synchrotron radiation (SR)-based techniques allowing us to confirm the ink nature of the sample and to distinguish its original formulation from the contaminants.

In particular, XRPD, XAS and FTIR measurements showed a substantial presence of silicates and common clay minerals in the ink along with cerussite and malachite, Pb and Cu bearing-carbonates, respectively. These evidence support the hypothesis of an important contamination of the ink by the burial environment (soil) and the presence of degradation products of the bronze inkpot. Moreover, the combined use of IR, Raman, and GC-MS evidenced that the black ink was mainly composed of amorphous carbon deriving from the combustion of organic material mixed with a natural binding agent, Arabic gum. This work also wants to underline how the intrinsic multidisciplinary approach based on SR experimental techniques represents the most efficient way to obtain a complete overview of complex materials such as archaeological artefacts.

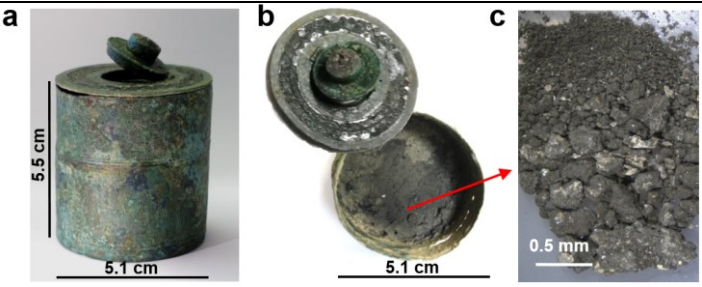

Figure 1. a. The bronze inkwell (front view); b. **i**nternal view of the inkwell with the black powder on the bottom and top view of the lid; c. the ink black powder residue collected from the bottom of the inkwell

[1] Presdocimi, A. Guida sommaria

[2] Aceto, M., Agostino, A., Fenoglio, G., Gulminie, M., Bianco, V., Pellizzi, E. (2012). *Spectrochim Acta Part A Mol. Biomol. Spectrosc*.**91**, 352.

[3] Christiansen, T., Buti, D., Dalby, K. N., Lindelof, P. E., Ryholt, K., & Vila, A. (2017). *J. Archaeol. Sci. Reports* **14**, 208.

[4] Lucas, A. & Harris, J. R. Ancient Egyptian Materials and Industries. (1962)

[5]. Ferrer, N. & Vila, (2006). *Anal. Chim. Acta* **555**, 161.

[6] Tack, P., Cotte, M., Bauters, S., Brun, E., Banerjee, D., Bras, W., Ferrero, C., Delattre, D., Mocella, V. & Vincze, L., (2016). *Sci. Rep*.**6**, 1.

[7] Brun, E., Cotte, M., Wright, J., Ruat, M., Tack, P., Vincze, L., Ferrero, C., Delattre, D., and Mocella, V., (2016). *Proc. Natl. Acad. Sci. U. S. A*. **113**, 3751

Keywords: multidisciplinary study; synchrotron radiation; roman ink

Acta Cryst. (2021), A**77**, C506