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Chemical analysis of fragments of glass and ceramic ware from Tycho Brahe's laboratory at Uraniborg on the island of Ven (Sweden)

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Abstract
In addition to his astronomical observations the famous Renaissance astronomer Tycho Brahe (1546–1601) was known also for his interest in alchemy. He equipped his castle Uraniborg on the island of Ven with a state-of-the-art alchemical laboratory when it was erected around 1580. After Brahe's death Uraniborg was demolished upon a royal decree from 1601, a process which was completed around 1650. In the present study we have analysed four glass shards and one ceramic shard most likely from the alchemical laboratory and retrieved during an archaeological excavation in 1988–90. Cross sections of the shards have been analysed for 31 trace elements by Laser Ablation Inductively Coupled Plasma with Mass Spectrometry with the aim of detecting any traces of the chemical substances on the inside or outside of the shards used in the laboratory. Four of the elements found in excess on the exterior surfaces of the shards, Cu, Sb, Au, and Hg, are in accordance with the reconstructed recipes of the three Paracelsian medicines for which Brahe was famous—*Medicamenta tria*. This is the first experimental data casting light on the alchemical experiments that took place at Uraniborg 1580–1599.

Keywords Tycho Brahe, Uraniborg, Alchemy, LA-ICP-MS, Trace elements, Paracelsian medicine, *Medicamenta tria*

Introduction

In recent years the study of alchemy has evolved into a transdisciplinary research field incorporating both history, archaeology, and chemistry, which now aims to focus on alchemists and their activities, rather than disembodied alchemical ideas, and to research the history and archaeology of alchemical practices [1, 2]. In the 1980's a sixteenth century alchemical laboratory was discovered and excavated in Oberstockstall, Austria [3, 4]. In the new world more than 2000 items was unearthed from an alchemical laboratory in Jamestown, Virginia, operated 1607–1610 [5], and recently crucibles from a seventeenth-eighteenth century chymistry laboratory, the Old Ashmolean laboratory in Oxford, have been analysed by modern analytical techniques [5, 6]. In all three cases exceptionally many pieces of laboratory equipment were uncovered. These examples set the stage for the present work, which reports the first results of analyses of the surficial layers of four glass shards and one ceramic crucible from the alchemical laboratory build by Tycho Brahe in c. 1580 on the island of Ven, present day Sweden. In the present case of Brahe's laboratory, we have a wealth of information from Brahe himself, and many of Brahe's thoughts about medicine making and alchemy. We also have precise dates of the operation of the laboratory, which lasted only approximately two decades until Brahe left Denmark in 1597. What is missing, though, is the precise nature of the medicinal recipes which we only know about from indirect and not so accurate sources [7, 8].

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Astronomy and alchemy under royal patronage

On February 11th 1576 the Danish King Frederik II (1534–1559–1588) summoned his subject, the nobleman and scientist Tyge Brahe (or Tycho Brahe as he called himself in Latin) and offered Brahe the island of Ven as a lifelong fief. Ven is a small and peaceful island centrally situated in the Sound (Øresund). In Brahe's time Scania and Ven were Danish land, but after 1658 both became parts of Sweden. The king promised to pay for the construction of a suitable palace on the island according to Brahe's wishes and to arrange the necessary funding ([9]: 25–29; [10]: 21–30).

Tycho Brahe accepted the royal offer and during the years 1576–1580 he erected Uraniborg as a unique combination of palace, observatory, and alchemical laboratory—in short, a scientific research centre (Figs. 1 and 2). It attracted students from all over Europe who lived and worked on Ven for months or years as members of Tycho Brahe's household, or as he expressed it in Latin his "*familia*" [11, 12]. As long as Brahe's patron King Frederik II lived and during the childhood and teenage years of his successor King Christian IV the activities of Tycho Brahe flourished. But when King Christian IV had come of age and had been crowned in Copenhagen in 1596, he and his counsellors looked at Tycho Brahe in a less favourable light ([13]: 201–215). In 1597 Tycho Brahe was deprived of his funding and had to leave Denmark for good, bringing all his instruments with him. After a lengthy journey over Rostock, Wandsbeck, Dresden, and Wittenberg he ended up in Prague at the court of the Roman-German Emperor Rudolph II. Here Brahe ended his career and died shortly after in 1601. After his death his palace and observatory on Ven was demolished and the stones reused for new, humbler buildings. The knowledge of the physical frames of his scientific work, as well as his astronomical instruments, would have been lost, if Tycho Brahe had not himself in time published prints of Uraniborg, the underground observatory Stjerneborg, and all the astronomical equipment.

In the history of science Tycho Brahe has been almost exclusively connected with his ground-breaking astronomical observations. This is quite understandable since his astronomical work turned out to be of vital importance for the scientific development of astronomy and the understanding of the universe. Yet in 1576 the king, according to Tycho Brahe, explicitly stated that he wanted to support Brahe's work with astronomy and alchemy; in Brahe's Latin: "*exercitijs tam Astronomicis quam destillatorijs*" ([9]: 27), so Tycho Brahe's royal patron considered both these two scientific activities important. This was in

Fig. 1 Uraniborg, partly the southern façade and partly a cross section of the basement. The alchemical laboratory can be seen in the lower left corner of the drawing. (Blaeu's Atlas Maior 1663, Wikipedia)

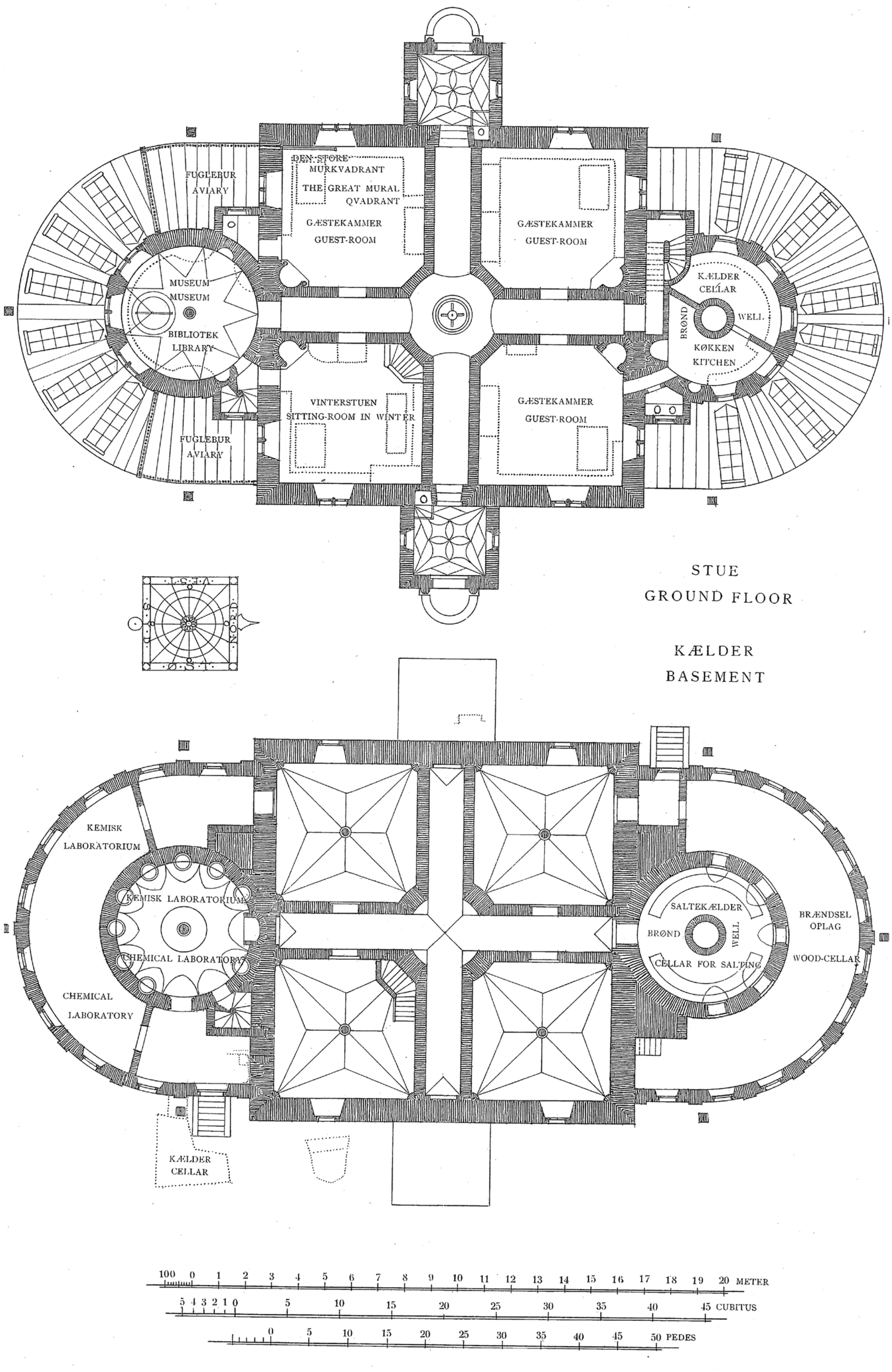

Fig. 2 Ground plan of Uraniborg, both ground floor (upper) and basement level (lower). North is to the right. The alchemical laboratory is seen in the left (southern rotunda) in the basement. The Winter Room is seen in the lower left quadrant of the main house on the ground floor (from Christensen and Beckett [16])

accordance with Brahe's own opinion that astronomy and alchemy were two sides of the same coin. He could refer to his alchemical research as earthly astronomy, since the heavenly regions and the Earth are part of the same all-encompassing system, united by a divine spirit that one can approach through one's studies.

In 1588 Tycho Brahe sent a letter to his colleague in Kassel, the mathematician Christopher Rothmann, listing some obvious analogies between celestial bodies, earthly elements, in this case metals, and parts of the human body: Sun/gold/heart; Moon/silver/brain; Jupiter/pewter/liver; Venus/copper/kidneys; Saturn/lead/spleen; Mars/iron/gall bladder; Mercury/mercury/lungs. He added that the analogies could be continued with *e.g.,* minerals and precious stones, so that emerald for example is connected to Mercury ([14]: 67). The latter analogy must be due to inspiration from the so-called Emerald tablet (*tabula smaragdina*), a text that Renaissance alchemists ascribed to the mythical god-like creature *Hermes Trismegistos* (Hermes being the Greek equivalent to the Roman god Mercury). One of the statements of the Emerald tablet reads: "That which is above is like to that which is below, and that which is below is like to that which is above, to accomplish the miracles of one thing." ([15]: 183). Based on the same line of thought Tycho Brahe had two stone-sculptures placed over the entrance doors to Uraniborg. One depicted astronomy as a reclining man holding a celestial globe and with the inscription in Latin: "By looking up, I'm looking down". The other depicted alchemy as a reclining man holding a cup, a snake, and some herbs, placed on top of the alchemical laboratory, with the accompanying text: "By looking down, I'm looking up" ([11]: 93).

The scope of Tycho Brahe's alchemy

Uraniborg was a highly original piece of architecture designed by the Danish court architect Hans van Steenwinckel (1587–1639) after ideas from Tycho Brahe himself. The core of the palace with living quarters had a symmetrical, square plan, with entrance towers to the east and west. Larger, circular extensions to the north and south were surmounted by wooden galleries and towers used as observation platforms. The extension to the north contained kitchen and storerooms while the south extension was totally reserved for scientific purposes. It had a library and "museum" on the ground floor and an alchemical laboratory in the basement, containing no less than 16 ovens that Brahe himself has specified: "Three bath-heaters, one digesting furnace with ashes, four large athanors, two small ones, two furnaces for distillation in sand or ashes, one for [use with] a large bellows that is connected with it by two pipes. Another special furnace was fitted with lamps. There were two reverberatory furnaces, one of which reflected the heat directly, the other reflected it along a spiral path, freely and yet indirectly." ([17]: 218) Depictions of the palace show that each oven stood in its own niche and that there was a circular working table around the central column of the vaulted room (Figs. 1 and 2). A spiral staircase led directly from the "Winter Room", which was the daily living room of Tycho Brahe and his family, down into the laboratory, and after some years a further five chemical ovens were even set up inside the Winter Room itself to avoid too much running up and down the staircase when an experiment had to be supervised. Tycho Brahe willingly demonstrated all the wonders of his palace to his guests, including the laboratory.

Johan David Wunderer visited the laboratory in 1589 and noted that the underground room devoted to the alchemical art contained "many unusual furnaces and apparatus, various large flasks of thirty measures, many amazing kettles, alembics, cucurbits, and similar strange utensils, which all had been procured at great cost" ([18]: 138). Another German guest, Martin Zeiller, gave this description: "Downstairs next to the wine cellar is a large vault in which Tycho's stills and furnaces are, upon which stand a large quantity of distillation flasks. A lot of them are curved at the top, made of copper, going out through some windows and in through others, in which unusual things are distilled" ([17]: 215). Some of the condensers fitted to the distillation vessels were apparently lead through the windows out into the cool open air and back.

Tycho Brahe was very secretive when it came to relating the actual experiments that he and his assistants carried out in the laboratory. In contrast to the astronomical work which Tycho Brahe discussed in letters and published in books, he never precisely described any results from his alchemical studies. In the book about his work and instruments from 1598 he cryptically states: "I have with much labour and at great expense made a great many findings with regard to the metals and minerals as well as the precious stones and plants, and other similar substances. I shall be willing to discuss these questions frankly with princes and noblemen, and other distinguished and learned people, who are interested in this subject and know something about it, and I shall occasionally give them information, as long as I feel sure of their good intentions and that they will keep it secret. For it serves no useful purpose, and is unreasonable, to make such things generally known" ([19]: 77–79).

He did, however, occasionally reveal his views on alchemy and its right use. In his major work on the new astronomy *Astronomiæ instauratæ progymnasmata*, published posthumously in Prague 1602, Brahe very explicitly distanced himself from alchemists trying to create gold: "Supposing that someone by means of the art they call "alchemy" can imitate nature, they are after all not able to make anything as good and lasting as nature itself." Alchemists might be able to process something that looks like gold in colour and weight, but the "gold" will reveal itself as fake when heated to a sufficiently high temperature or tested with antimony or acid. "Only very few have, as far as we know, succeeded while the great majority have been disappointed and have, far too late, regretted after wasting much time, labour and money" ([14]: 72–74). He never used the word "alchemy" to describe his own studies which he termed either *ars pyronomica* or *ars spagyrica*. In a letter from 1580 he mentioned that his studies in the laboratory primarily concerned "spagyrical medicine." His inspiration was the German medical writer and philosopher Theophrastus Paracelsus (1492–1541) who based his ideology on a hermetical concept of the universe as a totality imbued with Gods wisdom which could be approached and understood through careful observations and alchemical experiments [20]. Paracelsus' thoughts were developed and refined by a number of pupils and followers, among which the possibly most important was the Danish medical doctor Petrus Severinus (in Danish Peter Sørensen) who in 1571 published an influential book on the ideal of philosophical medicine. Severinus was well acquainted with Brahe, and Brahe clearly shared his interest in the study of iatrochemical, Paracelsian medicine ([19]: 51–94).

We know very little of Tycho Brahe's medical recipes. For instance, his most renowned medicine, *elixir Tychonis*, which consisted of three ingredients, *Medicamenta tria,* was widely known at the time, but Brahe never revealed the recipe for this. The earliest description of medicine from his hand is found in a letter from May 18th 1571 to his close friend Anders Sørensen Vedel. Vedel had asked for a recipe against fever, and Tycho answered that he with much effort had persuaded a good friend and doctor to reveal it to him in return for four secrets from Tycho Brahe's lips. The recipe is a quite traditional herbal medicine based on a water solution with knapweed (*centaurea*), blessed thistle (*cardus benedicta*), and chicory (*cichorium intybus*) as well as some powder prepared from the lower jaw of a stickleback. Many people, Brahe added, are also cured by wearing a necklace with a nutshell containing a large, living spider. Brahe continued that he knew several other secret and reliable medicines to cure fever, but, if necessary, he would prefer to reveal them to Vedel only orally ([10]: 20–21).

In 1599 Brahe sent a recipe for an elixir against plague to his new patron, emperor Rudolph II. The elixir was based on *theriac*, which was a substance mixed from several, mostly herbal ingredients including opium, to which a powder made from the flesh of vipers was added as an important ingredient. The result of a complicated process of extraction, filtering, and distillation was an *aqua vitae* to which one could add, among other things, a vitriol (sulphate) of copper or iron. Further oils and herbal ingredients were added during another process of filtering and distillation. The resulting elixir against plague could be further improved by adding a tincture of *e.g.,* corals, sapphire, hyacinth, and drinkable gold (*aurum potabile*) ([21]: 2–3). This recipe is only slightly influenced by Paracelsian medicine, in contrast to a laxative which Brahe mentions in the same letter, consisting of white antimony powder, prepared from pure antimony metal. A recipe sent by Brahe to the Danish governor in Schleswig–Holstein Heinrich Rantzau in 1597 against skin disease and blood infection was clearly based on Iatrochemical theories as revealed by its mercury and antimony ingredients ([21]: 4). Brahe's own writings do not allow to infer much more regarding the precise ingredients in his iatrochemical work.

Archaeology in Uraniborg

In light of Tycho Brahe's secrecy it is a tempting idea to attempt an investigation of possible traces of Tycho Brahe's alchemical activities, although such a project is severely inhibited by the almost total destruction of Uraniborg already in the early seventeenth century. The laboratory has disappeared, and its content has been destroyed and scattered over the area where Uraniborg used to stand. In 1824 the scarce remains of the buildings were unearthed. The chemical laboratory could be identified as a circular basement room about 11,3 m in diameter. Traces of two ovens were found under the floor. One was round, still with remains of ashes in it, the other rectangular, with remains of charcoal. Close to the laboratory area, but outside the palace proper, a separate vaulted cellar was excavated. Besides some architectural fragments of plaster and stone it contained broken retorts of glass and stone, as well as charcoal and sulphur, which was interpreted as waste from Tycho Brahe's alchemical experiments ([22]: 20–21). More recent excavations 1988–90 concentrated on the garden and the surrounding earth constructions and smaller buildings [23]. Among the sparse finds from these modern excavations were fragments of elaborate vessels of glass and ceramics that may have originated from the laboratory. The present work reports analyses on four glass samples and one ceramic sample found during the excavation in 1988–90 [23] with the aim of revealing any traces of the inorganic chemical substances worked with or treated in these particular vessels.

Sample material

Four glass samples and one pottery sample were selected for the present study from inventory number 30373, feature 22 in excavation shaft X, which is situated outside the garden wall's south gate facing southeast. The approximate position of shaft X is marked with a red arrow on Fig. 3 and shown on the detailed excavation plans Figs. 4 and 5. At a first glance the position of the finds can be thought to be too far removed from the laboratory to merit a connection. It is, however, difficult to propose an alternative provenance for the items which rather clearly look like funnels, flasks, and other laboratory equipment (Fig. 6), which was high-tech equipment at the time, on the solitary island of Ven. It can be speculated that because the items were deposited just outside the southern gate, it is likely that the deposition took place during the time when Uraniborg were in operation, as opposed to a deposition during the demolition phase. However, it was the field excavator's off hand interpretation that the finds from feature 22 were deposited as part of the demolition phase. In either case it is hard to imagine an alternative source other than the alchemical laboratory for the funnels and flasks, also in light of the findings reported below. The Lund Museum find number is 145 collectively for the shards shown on Fig. 6, whereas only 144 samples appear on the find list of the excavation filed at the museum. The explanation being, that several finds from shaft X were extracted and collectively termed 145 when these were put on loan to the Tycho Brahe Museum on Ven, according to curator Jenny Bergman at the Historical Museum at Lund University. The preservation state of the glass samples varies from well-preserved (KLR-13155) to some degree of degradation (KLR-13156, KLR-13157, and KLR-13158). The ceramic shard, KLR-13159, has a slip applied to both the external and the internal surfaces. Descriptions of the items are summarized in Table 1.

The parts of the samples marked by red squares on Fig. 6 were cut by the Historiska Museet at Lund University and shipped to University of Southern Denmark for analysis. The large shard (top left in Fig. 6) was deemed too fragile to survive cutting without being broken to pieces. Only one of the ceramic shards depicted in Fig. 6 were sampled, because the three

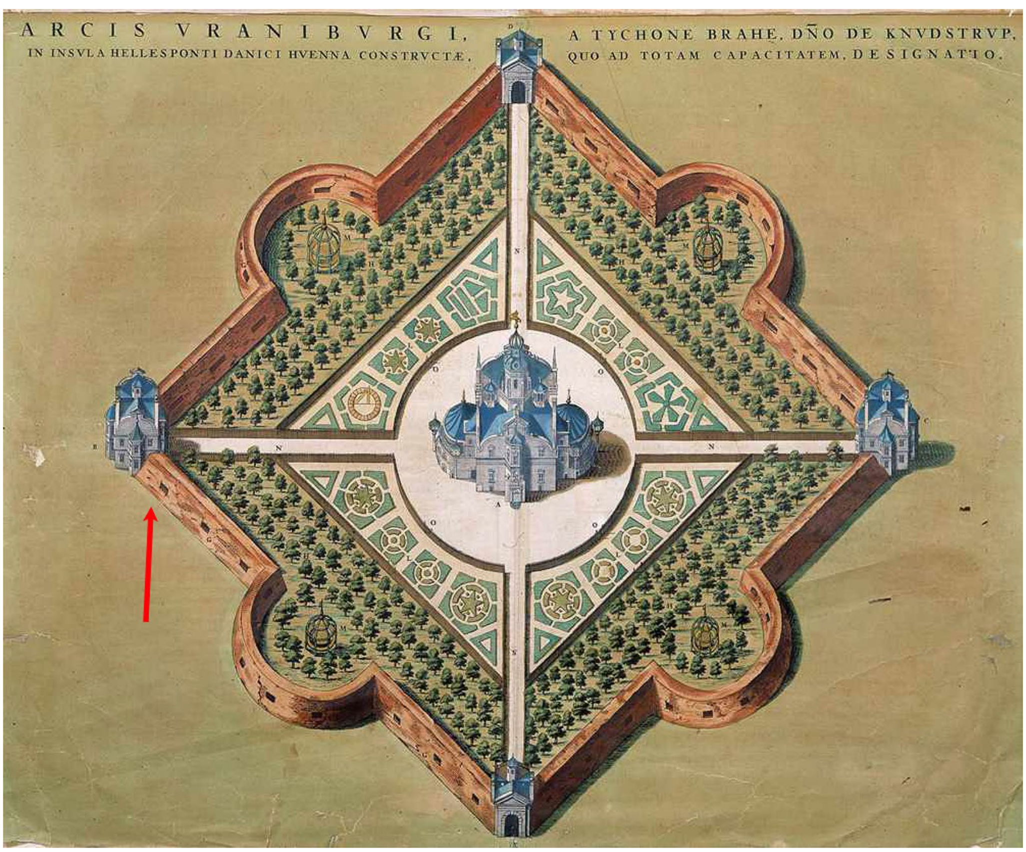

Fig. 3 View of the garden surrounding Uraniborg. North is to the right. The outer garden wall is oriented with corners pointing in the four corners of world, and thus rotated 90 degrees with respect to the orientation of Uraniborg itself. The approximate position of shaft X is marked with the red arrow, against the southeastern garden wall, near the southern gate. Adapted from Wikimedia Commons

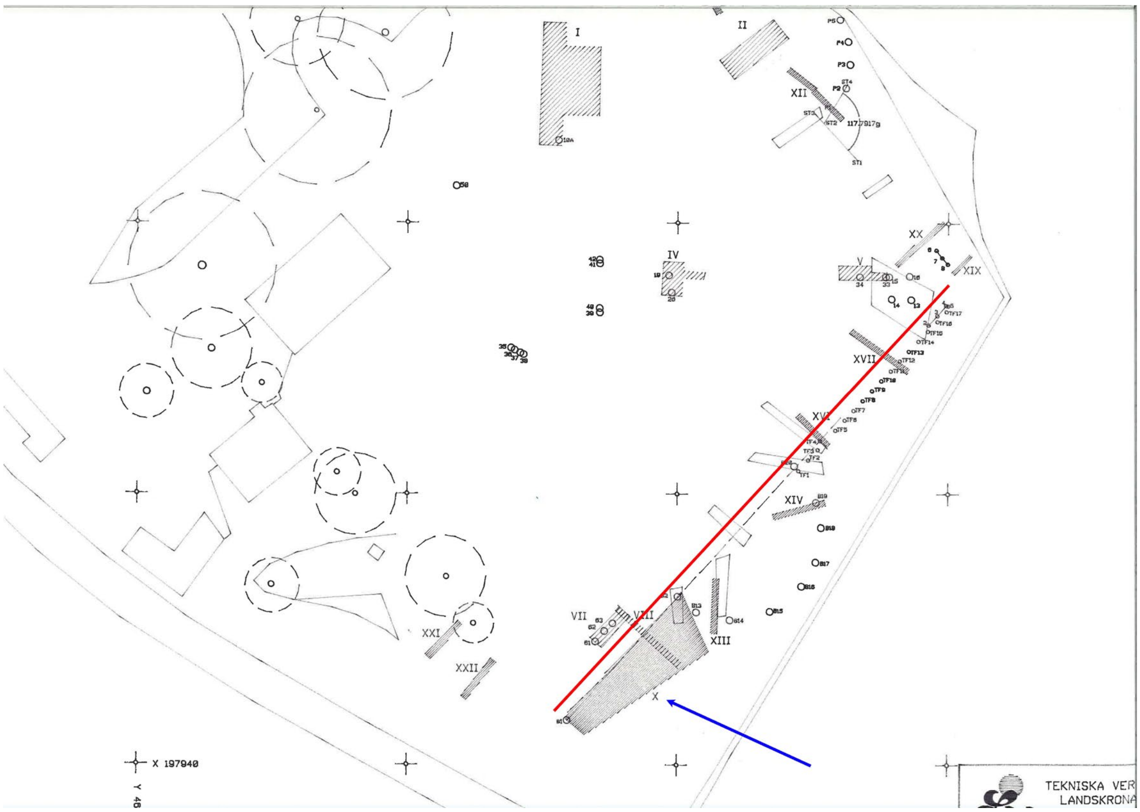

Fig. 4 Excavation plan from the 1988–1990 excavation. North is upwards. The samples from the present study are from inventory no. 30373, feature 22, shaft X. Shaft X is indicated by the blue arrow. The southeastern garden wall is indicated by the red line (Excavation plan, Landskrona Museum 1992)

ceramic shards were thought to be so similar in appearance and thickness, that they likely originated from the same vessel. Subsamples were cut at SDU and were embedded edge-on in epoxy resin from Struers. The embedded samples were polished to 1 µm diamond finish.

Methods

Micro-X-ray fluorescence (µ-XRF)

An ARTAX-800 µ-XRF manufactured by Bruker-Nano was used for the µ-XRF measurements. A high tension of 50 kV and a current of 600 µA were used. Absolute calibration of the concentrations has been performed by the DCCR-method (Direct Calibration from Count Rates) provided by the Bruker software using the standard reference material NIST-610 for the four glass samples, and the standard reference material NIST-2711 for the ceramic sample. The NIST2711 was formed into a pellet by applying a pressure of 120 kN. The results can be considered quantitative because of the similar matrix of the samples and the standard materials.

Laser ablation inductively coupled plasma mass spectrometry analysis (LA-ICP-MS)

Laser Ablation was performed with a CETAC LXS-213 G2 equipped with a NdYAG laser operating at the fifth harmonic at the wavelength of 213 nm. A circular aperture of 25 µm was used, and the shot frequency was 20 Hz. The line was scanned at 10 µm s^{-1}. The helium flow was 600 mL min^{-1}. Laser operations were controlled by DigiLaz G2 software provided by CETAC.

The ICP-MS analyses were undertaken with a Bruker Aurora M90. The radiofrequency power was 1.30 kW, plasma Ar gas flowrate was 16.5 L min^{-1}, auxiliary gas flowrate was 1.65 L min^{-1}, and sheath gas flowrate was 0.18 L min^{-1}. The following isotopes were measured without skimmer gas and are reported in this study: ^{23}Na, ^{29}Si, ^{31}P, ^{33}S, ^{35}Cl, ^{39} K, ^{44}Ca, ^{55}Mn, ^{57}Fe, ^{60}Ni, ^{65}Cu, ^{66}Zn, ^{75}As, ^{85}Rb, ^{88}Sr, ^{103}Rh, ^{105}Pd, ^{107}Ag, ^{111}Cd, ^{118}Sn, ^{121}Sb, ^{137}Ba, ^{182}W, ^{185}Re, ^{189}Os, ^{193}Ir, ^{195}Pt, ^{197}Au, ^{202}Hg, ^{205}Tl, and ^{208}Pb. The analysis mode used was peak hopping using 3 points per peak. Isotopic interferences were corrected for by the ICP-MS software. The dwell time on each peak was 2000 µs, expect for Na and Ca where

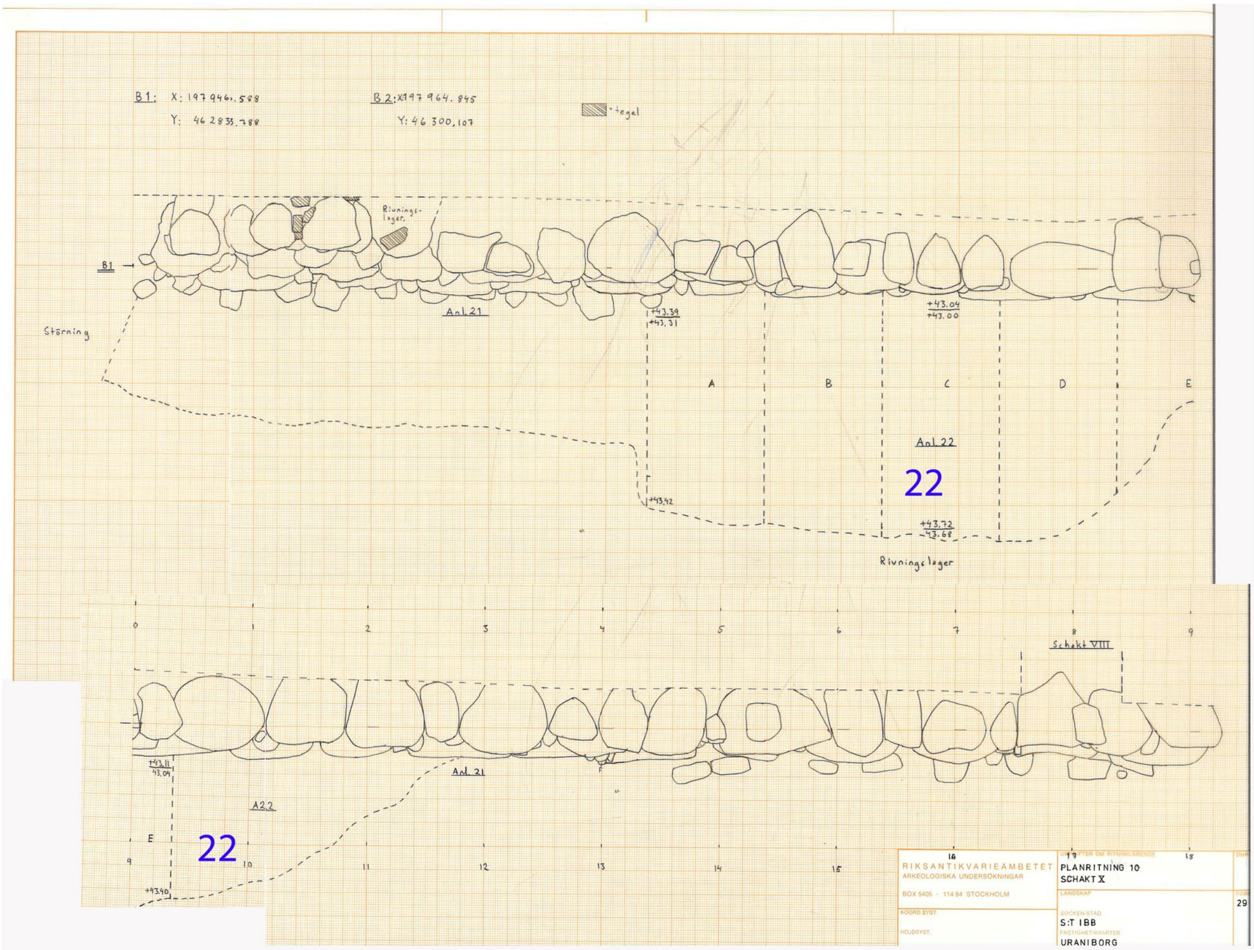

Fig. 5 Cross section and Plane view of section of shaft X. Feature 22 can be seen in both drawings (Field drawing, Landskrona Museum 1992)

the dwell time was reduced to 500 µs, and P, S, Cl, and K where the dwell time was set to 1000 µs to avoid overflow in the counter. In order to increase the count rates the dwell time for following elements were set to 10.000 µs: Rb, Sr, Rh, Pd, Ba, W, Re, Ir, and Pt. The measurements were preceded by a 10 s gas blank, the count rates of which were subtracted from the isotope count rates.

The quantification was performed by first adjusting the count rate for the isotopic abundance for the analysed isotopes. Following that the shape of the count rate as a function of atomic number was determined by analysing seven elements in a 5 ng g^{-1} multi-element standard solution (XXI for MS by Accustandard,). Further descriptions of the method can be seen elsewhere [24–27]. The isotopes used were: ^{9}Be, ^{25} Mg, ^{59}Co, ^{115}In, ^{140}Ce, ^{206}Pb, and ^{232}Th. The expected count rate in a 5 ng g^{-1} solution of the isotopes analysed for in this study was then calculated by interpolation between the measured count rates of these seven isotopes. Finally, the conversion from count rate to g g^{-1} was done by multiplying by a fixed ratio, determined by taking the average of the measurements in the interior parts of the samples and there fixing the average Ca concentration to the values obtained by µ-XRF.

All glass specimens were stained to some degree, and the laser ablation lines were placed at sites on the specimen surfaces with a low or no occurrence of stains and corrosion products.

Results

µ-XRF

Calcium concentrations were measured by µ-XRF in the interior of the shards to be 10.89 wt% (for KLR-13155), 15.8 wt% (KLR-13156), 15.1 wt% (KLR-13157), 15.3 wt% (KLR-1358), and 0.30 wt% (for KLR-13159). These concentrations were used for the quantification of the LA-ICP-MS results as described above.

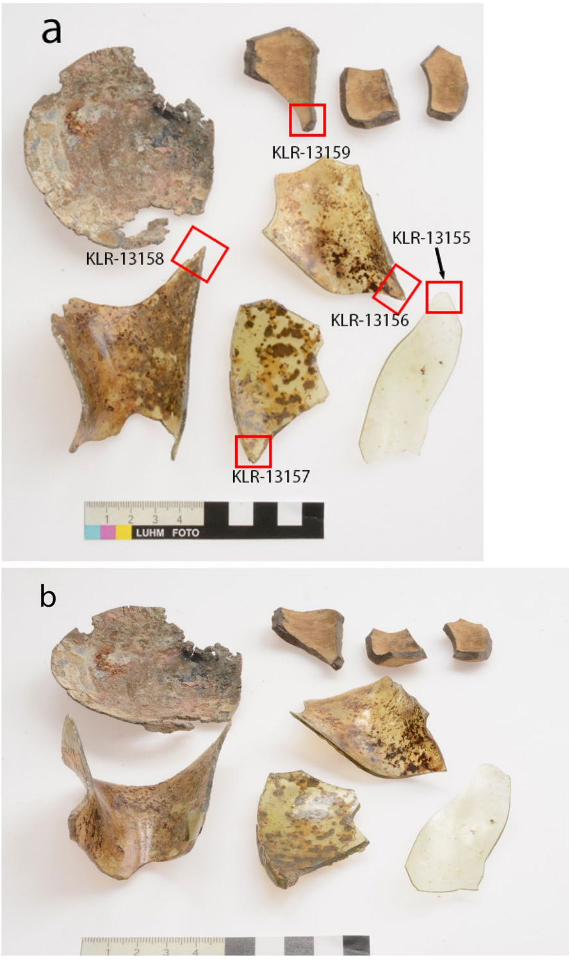

Fig. 6 **a** Four glass fragments and one ceramic fragment selected for the present study. The Lund Museum find number is 145 collectively for all the shards. **b** The same shards photographed at an angle to help perceive the 3 dimensionality of the shards (Photos by Lund Museum)

Table 1 Description of the items sampled for this study. See photographs in Fig. 6

Lab No	Type	Description
KLR-13155	Glass	Transparent, whiteish, well preserved
KLR-13156	Glass	Transparent, yellowish, some degradation
KLR-13157	Glass	Transparent, greenish, some degradation
KLR-13158	Glass	Transparent, yellowish, some degradation
KLR-13159	Ceramic	Dark brown interior, light brown to yellowish slip on both sides, no glaze

LA-ICP-MS

For the samples KLR-13155, KLR13156, and KLR-13157 the laser ablation line crossed the entire width of the glass samples (Fig. 7). For sample KLR-13158 and KLR-13159 traces entering from either side were made. No further averaging of the data was used, and the data shown in Fig. 8 originate from points along a single ablation line.

Discussion

Most elements exhibit rather constant concentrations across the interior and the ends of the samples. Examples can be seen in Fig. 8A, which shows the concentration profiles for Ca, Mn, Fe, and Zn for glass sample KLR-13155, where almost constant concentration values can be observed. However, some elements are enriched at the surfaces of the shards (Fig. 8 and Table 2). For example, the element W exhibits a sharp concentration peak at the outer surface of the vessel KLR-13155, and the three elements Sn, Au, and Pb have sharp concentration peaks at the interior surface of the same shard (Fig. 8B). Precisely how high the concentrations are in the surficial peaks is not possible to establish due to the width of the laser beam and the unknown thickness of the enriched layer. However, what is important to note here is that they are enriched without any doubt. For instance, the peak in W is 36 times higher than the background count rate in the glass matrix of KLR1-13155, and similarly 4 times for Pb.

The presence of W is most peculiar and unexpected. Tungsten in its pure form was not known to alchemists in Brahe´s times. It was discovered as a new element and isolated as a metal only by the end of eighteenth century. Important W ore minerals include scheelite ($CaWO_4$) and wolframite ((Fe, Mo) WO_4). Molybdenum was not measured in the present work, so a distinction between these two minerals cannot be made in the present project. It can be speculated that a W bearing mineral was included in a process conducted in the laboratory not knowing the nature of this mineral or by mistaken identification of the mineral. Still, the source of the elevated W levels remains a real puzzle.

Some elements are not significantly enriched at the surface or are exhibiting low count rates at the surface as well as in the interior of the shards. These 20 non-enriched elements are: Na, Si, P, S, Cl, K, Mn, Fe, Rb, Sr, Rh, Pd, Ag, Cd, Ba, Re, Os, Ir, Pt, and Tl. There can be various potential reason as to why these elements are not enriched. It could be because they were not used in the alchemical laboratory at Ven, or alternatively that shards containing these elements on their surfaces were not selected for the present study.

For some of the surfaces, the outer surface of KLR-13156, the inner surface of KLR-13157, and both the inner and outer surface of KLR-13158, no enrichments are observed for any of the elements analysed. One inference that can be drawn from this is that enrichments are seen on both inner and outer surfaces. It is hardly surprising that some elements are present on the inner surfaces of the vessels, but the occurrence

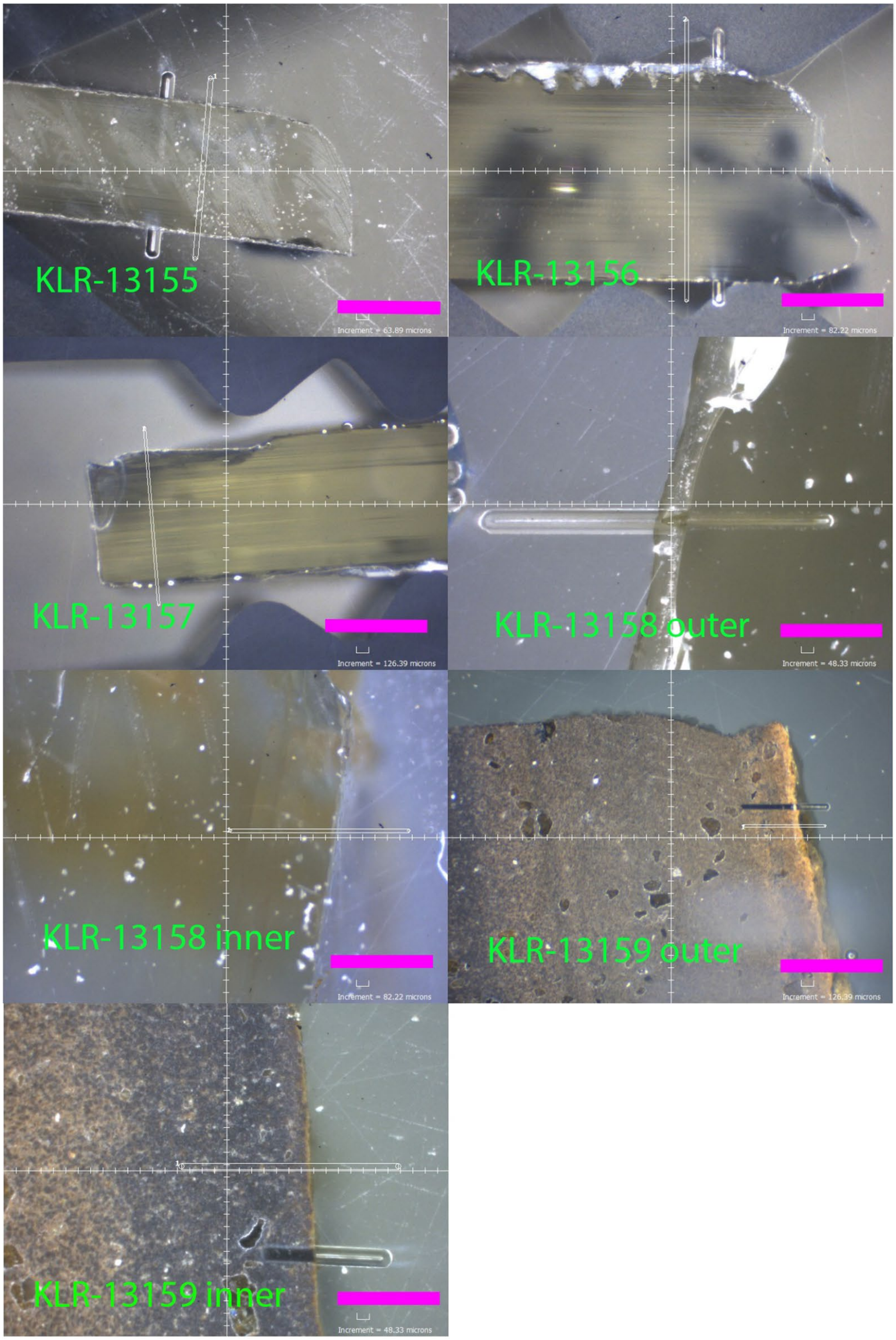

Fig. 7 The Laser Ablation lines as seen from the camera of the Cetac Laser Ablation machine. All lines are marked and prior to ablation. The ablation track seen are from test runs. The magenta scale bars are 500 µm long

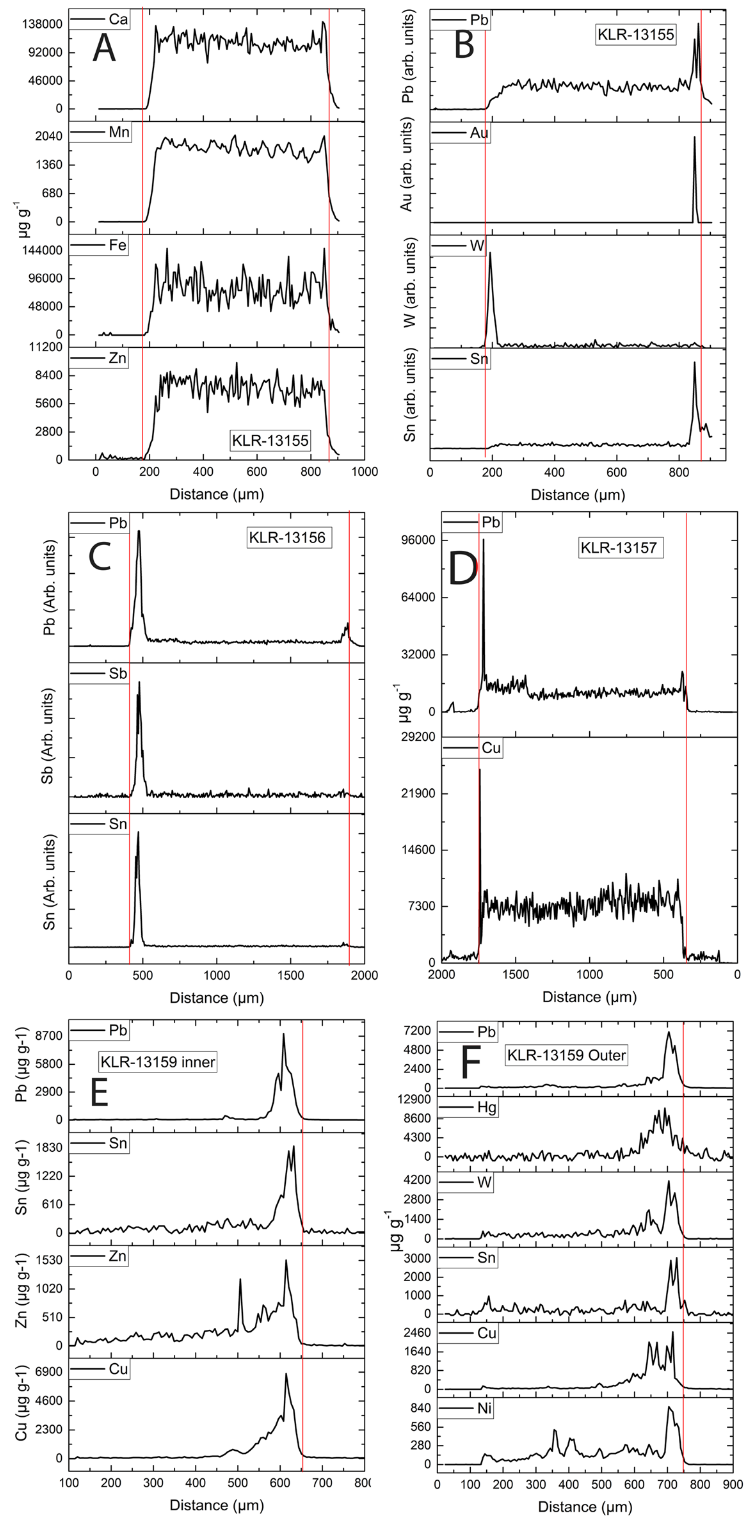

Fig. 8 Calibrated LA-ICP-MS elemental concentrations in a 25 µm wide and ca. 25 µm deep ablation line perpendicular to the surface of the samples are given in µg g^{-1} (**A**, **D**, **E**, and **F**). The peak heights for some of the elements in B and C exceeded the linearity of the calibration and are therefore shown in arbitrary units (**B** and **C**). The vertical red lines in the diagrams mark the border between the epoxy embedding material and the sample. For samples KLR-13155, KLR-13156, and KLR-13157 (**B**, **C**, and **D**) the outer part of the vessel is to the left in the diagrams, and the inside of the vessels to the right. For the ceramic shard KLR-13159, diagram E has the inner surface of the vessel to the right, while in F the outer surface is to the right in the diagram. With the exception of KLR-13155 (**A**), the elements not deviating from a flat line at the surface are not shown

Table 2 Summary of enriched elements at the surfaces of the shards.

Sample	Surface enrichment	Presence in recipe
KLR-13155 inner surface	W	
KLR-13155 outer surface	Sn, ***Au***, Pb	**Prep I**, *Prep II*
KLR-13156 inner surface	Sn, <u>**Sb**</u>, Pb	**Prep I**, <u>Prep III</u>
KLR-13156 outer surface	–	
KLR-13157 inner surface	–	
KLR-13157 outer surface	*Cu*, Pb	*Prep. II*
KLR-13158 inner surface	–	
KLR-13158 outer surface	–	
KLR-13159 inner surface	*Cu*, Sn, Zn, Pb	*Prep II*
KLR-13159 outer surface	Ni, *Cu*, Sn, W, <u>Hg</u>, Pb	*Prep II*, <u>Prep III</u>

Last column summarises the participation in the three preparations for the *Medicamenta tria*

The elements occurring in preparation 1 are marked with bold, preparation II with italic, and preparation III with underlining

of enriched elements potentially linked with alchemical activities on the outer surfaces (KLR-13155, KLR-13157, and ceramic vessel KLR-13159) is interesting. It could indicate that these vessels were dipped in other—larger—vessels, containing compounds of the elements listed in Table 2, or alternatively just haphazardly sprayed or sprinkled upon. A third possibility is that the enriched elements entered the exterior surfaces of vessels during heating in a sand-bath, as described by Brahe for two of the 16 ovens present in the basement. A fourth possibility is that the ceramic vessel served as a lid which was put on other vessels during operations taking place in the ovens. It must be emphasised that all four explanations should merely be regarded as hypotheses, we have no evidence for either one of them. The many elements present on the ceramic shard has some bearings on what type of experiments were conducted in the laboratory, although such details are hard to decipher with the present limited number of samples.

The ceramic vessel KLR-13159 attracts attention because it exhibits the largest number of enriched elements (seven). Copper, Zn, Sn, and Pb were present on the inner surface and Ni, Cu, Sn, W, Hg, and Pb on the outer surface of the vessel. There were, however, no signs of glazing or glass on either side of the vessel. One possible interpretation of this large number of enriched elements could perhaps be that the ceramic vessel was used as a garbage container for the experiments. An alternative interpretation is that it could have been used in metallurgical operations involving copper alloys or the dissolution of metals into strong acids.

Comparison with the trace element inventory in Brahe's bones

Following the Danish-Czech exhumation of Tycho Brahe in 2010 in the Tejn Church in Prague, Czech Republic, chemical analyses of Brahe's hair revealed that he had experienced an excess exposure to Co, As, Ag, and Au during the last two months of his life [28–31]. These analyses reflect only the time when Brahe was living in Prague and surroundings. The exposure recorded in the bone samples of Brahe, both cortical (5–10 years) and trabecular (3–5 years) will, however, partly reflect the time when Brahe still lived on the island of Ven, and therefore possibly worked in the alchemical laboratory in Uraniborg. Most of the attention has been directed at the element Hg, with which it was speculated that Brahe could have been poisoned. However, no excess Hg levels, lethal or non-lethal, were observed beyond what was normal for people of the time. The Brahe bone samples which have been analysed showed Hg concentrations between 0.0146 and 0.036 $\mu g\ g^{-1}$, and the non-exposed medieval and post-medieval populations have been shown to have concentrations less than 0.08 $\mu g\ g^{-1}$ for cortical tissue and 0.3 $\mu g\ g^{-1}$ for trabecular tissue [32–34]. In this respect, it is particularly interesting to note that enriched levels of Hg were found in the ceramic vessel KLR-13159. This means that there was indeed work taking place using Hg in the alchemical lab of Uraniborg, even though no traces of excess Hg were found in the skeletal remains of Tycho. It seems that either Brahe was very cautious when he practiced alchemical experiments involving Hg, or alternatively that he did not in his older days participate much in these practical laboratory endeavours himself but left it mainly to his pupils and co-workers.

Kucera et al. [28] reported trace element concentrations in five bone samples, measured partly by INAA and partly ICP-MS. The elements Ni, Cu, Sn, and W were not reported or were below detection in Kucera et al., but the other elements found in excess on the surfaces of shards in this study are reported [28]. For Zn, the bones samples of Tycho Brahe ranged between 120 and 550 $\mu g\ g^{-1}$, in accordance with the normal non-exposed medieval population, which ranged between 75 and 857 $\mu g\ g^{-1}$ [28]. For Sb the bones samples of Brahe ranged from 0.065 to 0.177 $\mu g\ g^{-1}$, while modern people have Sb concentrations between 0.01 and 0.3 $\mu g\ g^{-1}$, also values which indicate normal concentration levels for the time. For Au the five bones samples of Tycho Brahe exhibited concentrations between 0.008 and 0.038 $\mu g\ g^{-1}$, while the modern population have between 0.00003 and 0.00039 $\mu g\ g^{-1}$. Finally, for Pb, the Brahe bone samples were measured to range between 72 and 111 $\mu g\ g^{-1}$, while the medieval population normally ranged between 1.8 and 35.7 μg g^{-1}, but some individuals have been seen with a Pb concentration of up to a high of 1835 μg g^{-1}.

The comparison shows that only for Au there seems to be concentration levels in Brahe's bones that are exceptionally high. This was also the case for Brahe's hair [33]. The enhancement in Au has been ascribed to a luxurious lifestyle of the very wealthy Tycho Brahe, who may have dined with gold-plated cutlery or tableware [28, 30]. This assumption has, however, been corrected since the use of gold or silver utensils may have had a small impact but would not have contributed significantly to the overall levels of silver and gold in Tycho Brahe's remains [7]. Hair samples from Diane de Poitiers (1500–1566), a renowned favourite of King Henry II of France (1519–1559) was also found to have excess gold concentrations, which has been interpreted as a result of ingestion of potable gold [35, 36].

Gold was found to be present in one of the glass samples from Uraniborg (KLR-13155 outer surface), which goes to prove that gold was indeed a part of the experiments at Brahe's laboratory. Considering that Brahe himself mentions the use of potable gold in one of his few known medical elixirs, and that an enhanced level of gold is found in the sample from Uraniborg it seems more likely that the traces of gold in Brahe's hair and bones should be interpreted as a result of consumption of medical elixirs containing precious metals, made in Brahe's own laboratory.

Inferences regarding medicine-making

Tycho Brahe was known to have produced Paracelsian medicine. He produced a so-called *elixir Tychonis*, which consisted of three prescriptions—*Medicamenta tria*—and which contained inorganic substances. As mentioned in the introduction, the knowledge about the *Medicamenta tria* is not based on Tycho Brahe's own writings, but instead of various parts of the recipes published by Pierre Gassendi in 1654, Thomas Bartholin in 1662, and the readings of two handwritten documents published by Karin Figala in 1972 [8]. According to the translations and interpretations of Karpenko and Kucera [7] summarising the mentioned sources the Latin texts concerning the three elixir prescriptions can be read as follows:

Preparation I: "Pro morbo epidemico pestilentiali vel aliis contagiis venenatis tam quoad praeservationem quam curationem." (For an epidemic disease or other infectious contagion both as regards prevention and treatment.)

Preparation II: "Pro morbis epilepticis et huc pertinentibus." (For epileptic diseases and related diseases.)

Preparation III: "Pro morbis cutem et sanguinem infestantibus, quails est scabies, inveterata lues venerea, ac elephantiasis, et similes." (For diseases affecting the skin and blood, such as scabies, chronic venereal diseases, and elephantiasis, and the like.)

Preparation 1 seemingly involved the use of Sb, Ag, and Au according to Karpenko and Kucera [7]. Enhanced Ag was not detected in any of the shards of the present investigation. Antimony and Au were found on two vessels (KLR-13155 and KLR-13156), one on the inside of the vessel and the other on the outer surface.

The recipe of Preparation II contained Cu, Ag, and Au. As mentioned before, Ag was not detected in excess in this work. Copper was found in increased amounts in three instances, the outer surface of KLR-13157, and on both sides of the ceramic shard KLR-13159.

Preparation III contained Sb and Hg. Antimony was found in excess on the inner surface of KLR-13156, while Hg was found in excess on the outer surface of KLR-13159.

In the present study we thus find four out of the five known elements mentioned in the three preparations (see Table 3).

The need for no less than 16 ovens at first sight draws a picture of a large production of quite refractory compounds like inorganic Paracelsian compounds. But the use of some of the ovens may have been for moderate temperatures, because Brahe was known to have a herbal garden with plants, which according to his own accounts were used for medicinal purposes [35, 36]. The preserved recipes show that herbal components went through various distillation processes by use of the laboratory equipment, and that further non-organic elements of Paracelsian character were added.

Table 3 Summary of the alleged inorganic composition of the three reconstructed *Medicamenta tria*

Element	Prep 1	Prep 2	Prep 3	Enriched in samples
Ni				+
Cu		+		+
Zn				+
Ag	+	+		
Sn				+
Sb	+		+	+
W				+
Au	+	+		+
Hg			+	+
Pb				+

Given the relatively few samples investigated in the present work, it is not to be expected that all elements worked on in the laboratory are to be found. The only element known to occur in the *Medicamenta tria* and not found in the present work is Ag. But the fact that the four other elements used in *Medicamenta tria*, Cu, Sb, Au, and Hg, were indeed detected in the five shards investigated here does, however, lend support to the thesis that preparations of the same type as the three known prescriptions could have been made in the alchemical laboratory beneath Uraniborg. Sulphur was also used in making the *Medicamenta tria,* and S has not been found in the present study. The missing S can—like Ag—be due to the small number of samples analysed, or alternatively S could have been present in a form that could be geochemically mobilized, *e.g.,* by the action of passing groundwater and left the shards during the residence in the soil for c. 400 years.

The elements found in enhanced quantities and not reported to be used in either of the three *Medicamenta tria* preparations are Ni, Sn, W, and Pb. The experimental character of Tycho Brahe's own alchemy could well have brought him into fields that turned out to have no direct medical use. Tycho Brahe explicitly distanced himself from alchemical attempts to create gold. Tycho Brahe's sister Sophie Brahe apparently shared her brother's focus on chemical experiments to make medicine. But she was engaged and later married to the alchemist Erik Nielsen Munk Lange (1553–1613) who spent his entire fortune on gold-making ([14]: 23–25). He is known to have paid week-long visits to Uraniborg on several occasions alone or in the company of Sophie Brahe, and he may well as a beloved guest have borrowed Brahe's well-equipped laboratory trying to transform other less noble elements into gold. The evidence from the Uraniborg shards show that there may have been experiments with other medicinal elixirs than the ones we by chance know of. But they may also offer a glimpse into other, non-medical alchemical experiments at the laboratory at Uraniborg.

Conclusions

The investigation is based on trace element analyses of four glass shards and one ceramic shard excavated from the ruin of Tycho Brahe's castle Uraniborg on the island of Ven. A series of elements were found not to be enriched on the surfaces of the five shards: Na, Si, P, S, Cl, K, Mn, Fe, Rb, Sr, Rh, Pd, Ag, Cd, Ba, Re, Os, Ir, Pt, and Tl. These elements could have been undetected due to the low number of shards analysed in the present study, but it could also be so that minerals containing these elements were not present on the shelves of the Uraniborg laboratory. On the other hand, the present work documents the presence of excess amounts of nine elements: Ni, Cu, Zn, Sn, Sb, W, Au, Hg, and Pb, collectively on the inner and outer surfaces of the shards. Four of these elements, Cu, Sb, Au, and Hg, are known to have been used in the three reconstructed recipes of Brahe's Paracelsian medicine, and these preparations could thus have been made in the alchemical laboratory on Ven. It should, however, be stipulated that the presence of these elements is not a proof that they were indeed used for making Brahe's medicine. The other five elements Ni, Zn, Sn, W, and Pb, were not mentioned in the preserved recipes and are therefore likely residues from alchemical experiments of an unknown character. The results of the present work offer a first unique glimpse into the alchemical work of Tycho Brahe's laboratory on Ven.

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Conceptualisation, sample acquisition, chemical analyses, discussion, writing: KLR. Historical context, discussion, writing: PGH. Both authors have approved the final version.

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