ARCHAEOMETRIC ANALYSIS OF SOME SCYTHIAN AND CELTIC GLASS BEADS FROM HUNGARY•

NÉHÁNY MAGYARORSZÁGI SZKÍTA ÉS KELTA ÜVEGGYÖNGY ARCHEOMETRIAI VIZSGÁLATA

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Abstract

We have analysed six Iron Age glass beads from Hungary (three Scythian stratified eye beads with bosses from Mezőtúr, two Celtic bobbin beads and one Celtic simple eye bead from Vác-Kavicsbánya) with handheld X-ray fluorescence (hXRF), micro-X-ray diffraction (μ-XRD) and electron microprobe analysis (EMPA) methods. Our aim was to determine the provenance of the beads, including the sources of the raw materials, and/or the production centres, since archaeological data about their provenance are ambiguous. The base glass of the beads (soda-lime-silicate glass) as well as their colourants (calcium antimonate for white, cobalt and copper for blue, iron-bearing lead antimonate for yellow) are similar and have parallels in the Iron Age Europe and the Mediterranean region (e.g. LBA Egypt, Celtic glass bracelets spread in La Tène coine, Vicenice (Czech Republic) in Late Hallstatt period, and ancient Greek colony of Apollonia Pontica in the Black Sea region). In addition, we identified a "new" colouring technique for Iron Age yellow glass beads, namely the use of iron-bearing lead antimonate that had been determined only sporadically in ancient world, for example LBA Egypt, then in the glasses of the Roman Empire. Although the mentioned parallels do not make clear the provenance of beads, according to archaeological data, Scythian beads with bosses most probably came from Greek colonies near the Black Sea through secondary trade connections, whereas the Celtic simple eye bead has three possible origins (Italy, Rhodes and Pontus), and the Celtic bobbin beads have unambiguous provenance according to the present archaeometric data.

Kivonat

Hat vaskori üveggyöngy (három szkíta dudoros pávaszemes gyöngy Mezőtúrról, két kelta orsó alakú gyöngy és egy szintén kelta egyszerű szemes gyöngy Vác-Kavicsbányáról) archeometriai vizsgálatát végeztük el kézi röntgenfluoreszcens (hXRF), mikro-röntgendiffrakciós (μ-XRD) és elektron-mikroszondás (EMPA) analízissel. A vizsgálat célja a gyöngyök származásának – beleértve a nyersanyagok forrását és/vagy a gyártó központokat – megállapítása, mivel a régészeti adatok eltérőek vagy bizonytalanok. A gyöngyök alapüvege (natúr szóda alapú üveg) és színezői (fehér: kalcium-antimonát, kék: kobalt és réz, sárga: vastartalmú ólom-antimonát) hasonlóak, párhuzamaikat pedig megtaláljuk a vaskori Európa és a Mediterráneum vidékén (pl. a késő bronzkori Egyiptomban, a La Tène kori kelta karpereceken, a késő Hallstatt kori Vicenice-ben, Csehországban, és a Fekete-tenger melléki görög gyarmatvárosban, Apollonia Ponticában). Emellett „új" színező eljárást is azonosítottunk a vaskori sárga üvegeknél, a vastartalmú ólom-antimonát használatát, amelyet ezidáig csak szórványosan mutattak ki az ókori üvegekben, például a késő bronzkori Egyiptom, valamint a Római Birodalom üvegeiben. Annak ellenére, hogy az említett párhuzamok nem teszik teljesen egyértelművé a gyöngyök származását, a régészeti adatokkal egybevetve a szkíta dudoros gyöngyök valószínűleg a Fekete-tenger melléki görög gyarmatvárosokból érkezhettek, míg a kelta egyszerű szemes gyöngyöknél akár három lehetséges származási hely is felvethető (Itália, Rodosz és Pontus), azonban a kelta orsó alakú gyöngyök eredete bizonytalan a jelenlegi archeometriai adatok tükrében.

KEYWORDS: SCYTHIAN, CELTIC, BEAD, SODA-LIME-SILICATE GLASS, LEAD ANTIMONATE

KULCSSZAVAK: SZKÍTA, KELTA, GYÖNGY, NATÚR SZÓDA ALAPÚ ÜVEG, ÓLOM-ANTIMONÁT

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Introduction

The present study deals with the archaeological issues and comparative archaeometric analysis of three Scythian stratified eye beads with bosses belonging to the Vekerzug culture (i.e. to the Scythian-like ethnic group of the Great Hungarian Plain in the Middle Iron Age), two Celtic bobbin beads and one Celtic simple eye bead of the La Tène culture (or as it is usually identified the Celts in the Late Iron Age). The main aim of the study was to increase the archaeometric database of the Iron Age glass found in the territory of Hungary. Besides, our further purpose was to compare the chemical composition of these beads as they can be related to each other by similar external features and function. Additionally, involving archaeometric data into the research of origin can help to determine the provenance of beads, thus to explore variable connections of the Iron Age Carpathian Basin. Although the study focuses on just a few artefacts, we got closer to answer from where these beads were exported to the mentioned cultures and what kind of technology was used to create glass jewellery at that time.

Archaeological background

For archaeometric investigations we chose a few, but significant glass jewellery from La Tène culture, which were the following: two Celtic bobbin beads of inhumation grave 29 and one Celtic simple eye bead of inhumation grave 11 from Vác-Kavicsbánya, dated to the second half of the 3rd century BC (LT C1) and in the first case belonging to a woman (Hellebrandt 1994; Hellebrandt 1999; Wolf 2013). The beads actually belong to the La Tène culture, but for the sake of simplicity in this article the ethical designation will be used more frequently. Besides, to get comparative archaeometric data from the preceding Vekerzug culture, three Scythian stratified eye beads with bosses deriving probably from an inhumation grave in Mezőtúr-Újváros, Mészárostelep and dated to the end of the 5th century BC, were selected (Kisfaludi 1983). It is evident that we deal with three different types of glass beads belonging to two cultures and deriving from two sites within Hungary, or in geographical sense the eastern region of Carpathian Basin (**Fig. 1.**). Thus, each type may originate from different contacts (e.g. commercial or diplomatic, etc.) and/or workshops.

Simple eye beads occur in eighteen various sites in the Carpathian Basin dating to between the 4th century BC and the end of the 1st century BC (LT B–LT D), but most popular were during 3rd century BC (LT C phase). In some cases larger number of beads, but in other cases lonely occurrence of beads was noted. The accompanying circumstances also show great varieties and are doubtful. We can only state that these were rare parts of wear. In the case of Vác-Kavicsbánya, the bead in question may have come from a grave of a woman formerly significant in society according to the rich furnishing in the proportions which are characteristic to the Carpathian Basin (Hellebrandt 1994; Hellebrandt 1999; Wolf 2013). Celts certainly imported eye beads; however, there is no uniform opinion from where. Another problem is that the issue was not in the centre of archaeological interests. Recently Tibor Kemenczei assumed that beads could come to the Carpathian Basin from Middle Italy with the help of Hallstatt culture as mediator in trade in the 8th century BC, thus in the LT ages (Kemenczei 2009). Furthermore, Kemenczei supposed that the Scythians had their beads from Greek colonies moved next to shores of the Black Sea just after the 7th or rather the 6th centuries BC (Kemenczei 2009). In contrast, low amount of information is available about Italian workshops functioning after the 6th or 5th century BC. Therefore, Natalie Venclová offered other provenance, like Rhodes or Greek centres of the Pontic area, and connected their production to the renascent fashion of the so-called stratified eye beads similar to the studied beads, flourishing in the 3rd and 2nd centuries BC (Venclová 1990; Angelini et al. 2010).

The issue of bobbin beads provenance is a bit more complex. We actually know only two cemeteries in Hungary, where this type of beads was excavated, and usually richer accompanying artefacts characterized these graves (Hellebrandt 1994; Hellebrandt 1999; Kaposvári 1969; Karwowski 2005; Tankó 2006). Although the type is also represented in the Central European Celtic material (e.g. Domaniowice, Poland), the number of known beads is low, thus the beads can be considered as rare (Karwowski 2005; Venclová 1974; Venclová 1990). Celts may have wanted to imitate the mask beads which were produced in ancient Greek colonies in the first half of the 3rd century BC and were widespread in Central Europe (Haevernick 1977; Karwowski 2005; Seefried 1982; Szabó & Borhy 2015). Although, according to the latest research, bobbin beads were produced just after the time when mask beads production was ceased in the Pontic Greek colonies (in the second half of the 3rd century BC or LT C1) (Rustoiu 2011), Celts were acquainted with them. The mask and bobbin beads coming from the same grave are the best proofs of this statement, like in the case of grave 29 of Vác-Kavicsbánya (Hellebrandt 1994; Hellebrandt 1999; Wolf 2013). Therefore, the local production is hypothesized within the Celtic Koine. Maciej Karwowski, and earlier Natalie Venclová localized a glass manufacturing centre in southwestern Slovakia, which could be one of the early centres producing bobbin beads modelled on mask beads and early types of glass bracelets for the Celts inhabited the Central European region, as the similar and common ornaments demonstrate (Karwowski 2005; Tankó 2006; Venclová 1990; Szabó & Borhy 2015).

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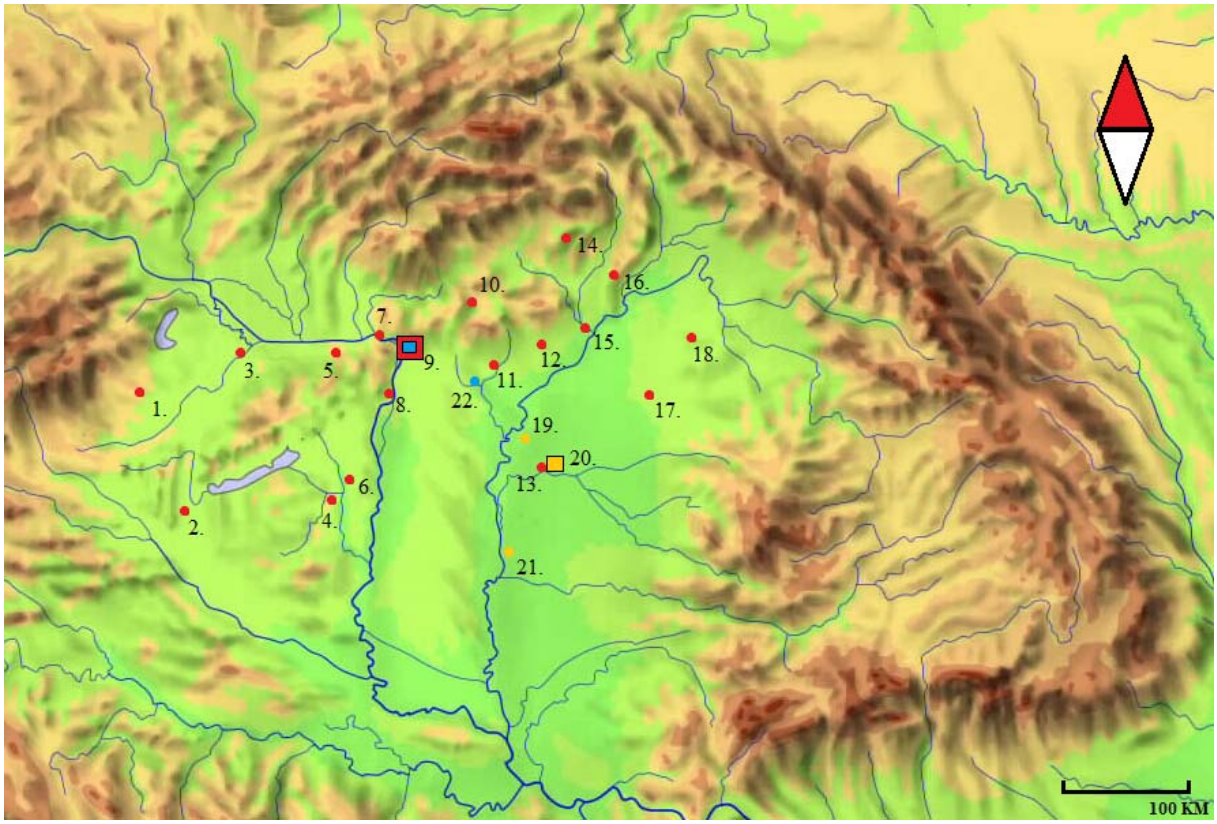

Fig. 1.: Map showing the sites, where Scythian eye beads with bosses (yellow circles), Celtic bobbin beads (blue circles) and Celtic simple eye beads (red circles) were found in Hungary. The sites of the studied beads are marked with squares with the artefact relating colour.

Key: 1: Velem-Szent-Vid; 2: Magyarszerdahely-Homoki-dűlő; 3: Győr-Ménfőcsanak; 4: Szárazd-Regöly; 5: Tarján; 6: Cece; 7: Szob-Kőzúzó; 8: Budapest, Gellérthegy-Tabán; 9: Vác-Kavicsbánya; 10: Szurdokpüspöki-Tsz major; 11: Jászberény-Cserőhalom; 12: Besenyőtelek-Szőrhát; 13: Szarvas; 14: Szendrő-Csengő barlang; 15: Sajópetri-Hosszú-dűlő; 16: Karcsa; 17: Álmosd-Homokbánya; 18: Nyírbátor; 19: Törökszentmiklós-Surján; 20: Mezőtúr-Újváros; 21: Szentes-Vekerzug; 22: Jászberény-Cserőhalom (based on the list of Karwowki 2005, Kemenczei 2009 and Wolf 2013, map from László Zentai).

1. ábra: A szkíta dudoros pávaszemes (sárga kör), kelta orsó alakú (kék kör) és kelta szemes gyöngyök (vörös kör) magyarországi elterjedése. A négyzettel jelöltek a vizsgált gyöngyök lelőhelyeit mutatják.

Jelkulcs: 1: Velem-Szent-Vid; 2: Magyarszerdahely-Homoki-dűlő; 3: Győr-Ménfőcsanak; 4: Szárazd-Regöly; 5: Tarján; 6: Cece; 7: Szob-Kőzúzó; 8: Budapest, Gellérthegy-Tabán; 9: Vác-Kavicsbánya; 10: Szurdokpüspöki-Tsz major; 11: Jászberény-Cserőhalom; 12: Besenyőtelek-Szőrhát; 13: Szarvas; 14: Szendrő-Csengő barlang; 15: Sajópetri-Hosszú-dűlő; 16: Karcsa; 17: Álmosd-Homokbánya; 18: Nyírbátor; 19: Törökszentmiklós-Surján; 20: Mezőtúr-Újváros; 21: Szentes-Vekerzug; 22: Jászberény-Cserőhalom (Karwowki 2005, Kemenczei 2009 és Wolf 2013 gyűjtése alapján, a térkép Zenta Lászlótól származik).

Besides similarities among mask and bobbin beads, the difference between their production technologies, the plasticity and colours of the used decorations, and the glass body shape is well visible, as well as another discrepancy is the time elapsed between their productions (Grose 1989; Karwowski 2005; Tankó 2006). This contradiction can be solved with the supposition of a migrating (maybe Greek) craftsman who knew the older fashion and was ready to serve the Celtic needs, or Celts reused the precedent Hallstatt traditions (Karwowski 2005; Rustoiu 2011; Szabó & Borhy 2015; Tankó 2006; Venclová 1974).

The eye beads with bosses of the Vekerzug culture were found in three sites of the Great Hungarian Plain, occurring in mainly richly furnished graves with various rites probably dated to the different phases of 6th–5th centuries BC (Alekseeva 1975; Csalog & Kisfaludi 1985; Kemenczei 2009; Kisfaludi 1983; Párducz 1954). This group has its parallels in the contemporaneous findings of Central Europe (e.g. Vicenice, Czech Republic) and Pontus as well, and at the same time in a large number in one grave, but still occurs rarely (Alekseeva 1975; Frána et al. 1987; Venclová 1974). In the case of the Mezőtúr beads, the finding circumstances were quite uncertain, due to the accidental recovering of human bones and artefacts.

In spite of this, the accompanying finds (like gold flitters, kauri, etc.) indicate an eastern provenance of the beads, mainly located to the Pontic region (Alekseeva 1975; Bottyán 1955; Kemenczei 2009; Kisfaludi 1983; Párducz 1954; Venclová 1974; Venclová 1990). On the other hand, southern origin (Aegean, Egyptian, Carthaginian) is also supposed by archaeologists (Csalog & Kisfaludi 1985; Dušek 1966). Besides, the third idea of local production inside the Carpathian Basin was also proposed (Dušek 1966; Venclová 1974), which can be ignored in the absence of any trace of glass manufacturing workshop at this time, so far. Obviously, eye beads with bosses can be considered as import goods, and depending on e.g. the accompanying finds, different origins can be determined, thus multiple provenances can be supposed regarding this bead type.

The comparison of Celtic and Scythian beads can be performed due to the fact that the beads have common features like their colour. Furthermore, they have common function, which can be an old tradition traced back to the Late Bronze Age Near East or Egypt, where simple eye beads appeared as apotropaic amulets (see more: Angelini 2011; Bottyán 1955; Chacheva 2015; Eremin et al. 2012; Fráná et al. 1987; Hunyadi 1942; Seefried 1982; Stolba 2009; Varberg et al. 2015).

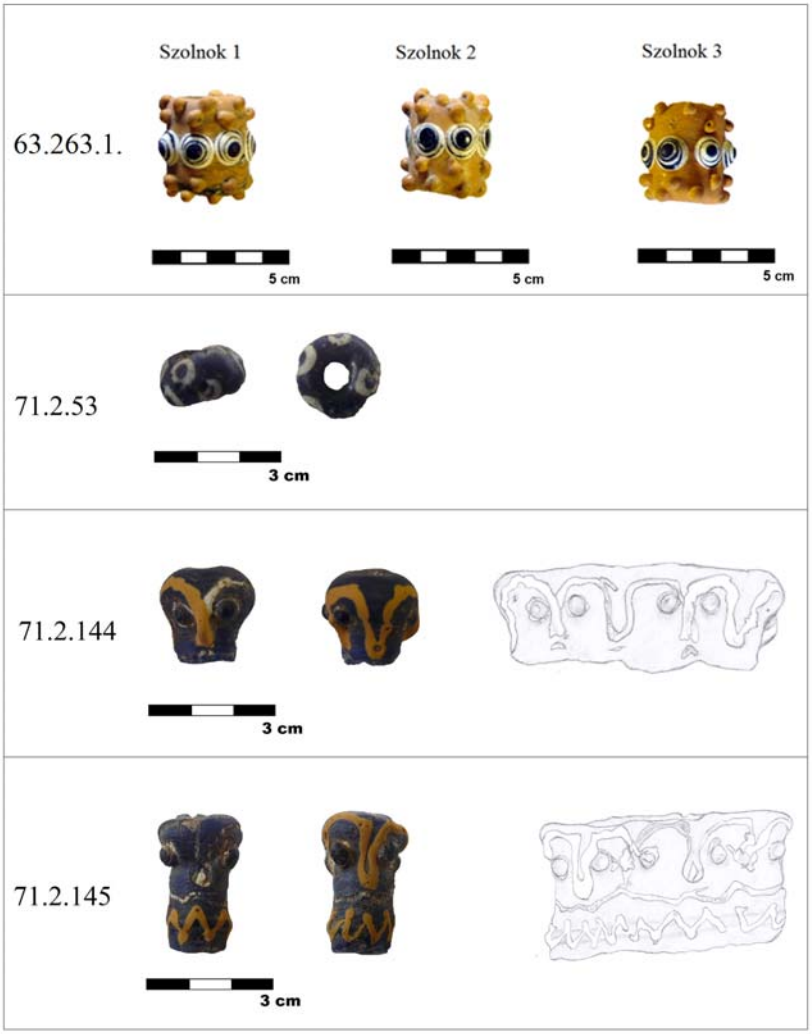

Fig. 2.: Three Scythian stratified eye beads with bosses (Szolnok 1–3) found at Mezőtúr. (Damjanich János Museum, Szolnok. Inventory nr.: 63.263.1), one Celtic simple eye bead from grave 11 and two Celtic bobbin beads from gave 29 excavated at Vác-Kavicsbánya (Tragor Ignác Museum, Vác, Inventory nr.: 71.2.53; 71.2.144; 71.2.145). (Photographs and drawings: Zsófia Osváth)

2. ábra: A mezőtúri szkíta dudoros pávaszemes gyöngyök (Szolnok 1–3) (Damjanich János Múzeum. Ltsz.: 63.263.1), a Vác-kavicsbányai kelta szemes gyöngy a 11. sírból és két orsó alakú gyöngy a 29. sírból (Tragor Ignác Múzeum, Ltsz.: 71.2.53; 71.2.144; 71.2.145). (Osváth Zsófia fotói és rajzai)

Studied artefacts

Simple eye bead (La Tène or Celtic) (Fig. 2.)

On the less transparent dark blue globular shaped glass bead nine flat glass eyes, made of opaque white combined with blue layers, are visible (from the Tragor Ignác Múzeum, Vác, Inventory n.: TIM 71.2.53). The bead surface is well preserved, but porous, and the white ornaments are not friable. Inside the bead eyelet a reddish yellow clayish layer (sediment) can be seen. Sizes: h.: 1.5 cm; w: 0.5–0.7 cm; diameter of eyelet: 0.8 cm.

Eye beads with bosses (Vekerzug culture or Scythian) (Fig. 2.)

Due to their common inventory number (Damjanich János Museum, Szolnok, Inventory n.: DJM 63.263.1), during analysis we used Szolnok 1, 2 and 3. All the three beads are opaque ochre yellow, and on the middle of cylindrical bead body seven circular and bulging stratified eyes of dark blue and opaque white layers can be seen, further seven bosses on each bead end are visible also in opaque ochre yellow shade. The eyes of Szolnok 3 are particularly bulging. Some bosses were broken off the beads. The surface of massive yellow bodies is a bit corroded, but glossy. On the surface of Szolnok 1 and 3 beads greyish marbly patterns can be observed, which cover almost the whole one end of bead Szolnok 1. Black spots occur on yellow bosses, maybe related to their seams. The opaque white layers of the eyes seemed porous, whereas inside the eyelet porous sediment with clayish particles, especially in Szolnok 3, are apparent, and the edges of the eyelets are fragmented. Sizes: Szolnok 1.: h.: 3.6 cm, diameter: 3.1 cm; Szolnok 2: h.: 3.5 cm, diameter: 2.8 cm; Szolnok 3: h.: 3.4 cm, diameter: 3.0 cm.

Bobbin beads (La Tène or Celtic) (Fig. 2.)

The four Janus-like faces formed by opaque ochre yellow, transparent blue and opaque white filiform glass ornaments can be found on the flared parts of both dark blue cylindrical beads. The blue bodies are visibly liny and chambered. The white glass contour marking the noses and around the eyes has already been fallen out. The yellow contours ran out of line probably due to the fluidic consistence of the glass. The other end of bead TIM 71.2.144 is fragmented and corroded, whereas around the thin end of bead TIM 71.2.145 there is yellow zigzag ornamentation, and the edge is eroded as well. Inside the eyelets reddish yellow sediment is visible. We used the inventory numbers as sample numbers (from Tragor Ignác Museum, Vác, Inventory n.: TIM 71.2.144, TIM 71.2.145, and the letter „v" in front of the inventory numbers signs Vác). Sizes: v 71.2.144: h.: 2.0 cm, w.: 1.6–2.1 cm, diameter of eyelet: 0.8 cm; v 71.2.145: h.: 3.2 cm, w.: 1.6–2.1 cm, diameter of eyelet: 0.8 cm.

Analytical methods

Handheld X-ray fluorescence analysis (hXRF)

Regarding the beads from Vác only non-destructive handheld XRF analysis was allowed to be performed. We applied this method on the beads from Mezőtúr as well, only to compare the chemical compositions.

The non-destructive chemical analysis of the artefacts was performed using a Spectro X-Sort Combi instrument, which is able to detect the elements from Mg to U, and light elements, like Na, are not detected. The measurement area was a circular spot of 0.3 cm in diameter. The ornaments and the bodies of the beads were measured separately for several times. Instrumental parameters: 15–50 keV, 21–50 μA (environmental calibration), Rh source, SDD detector with Peltier cooling, 1 minute count time.

Micro-X-ray diffraction analysis (μ-XRD)

For detecting the crystalline phases of the beads, like colourants, a Rigaku D/Max Rapid II instrument was used. Although sampling is not needed, the beads from Mezőtúr were sampled due to their large size. The digital camera equipped to the instrument helped to select the proper measurement site. Analytical parameters: Cu K_α, 50 kV, 0.6 mA, image plate detector, 1, 6 or 12 minutes measurement time, 300 to 800 μm collimators.

Electron microprobe analysis (EMPA)

The yellow glass of Szolnok 1–3 beads was sampled (cc. 0.1 cm) for microstructural and quantitative chemical analysis. The samples were embedded in resin, polished, and coated with carbon. The analysis of the samples was carried out using a JEOL Superprobe-733 instrument equipped with Oxford Instruments INCA Energy 200 type energy-dispersive X-ray spectrometer (EDS). Spot analyses were done for determination of the vitreous matrix composition by using an electron beam of 10 μm in diameter to avoid escape of alkali (40 seconds count time). In addition, area measurements were also carried out, analysed areas varied from 140 x 110 to 200 x 160 μm (15 minutes count time). Inclusions were analysed using focused electron beam 1 μm in diameter and 40 seconds count time. Analytical conditions: 20 keV accelerating voltage, 4-5 nA beam current. Typical components of the vitreous matrix were measured, like Na, K, Ca, Mg, Al, Si, Cl, Fe and Pb, in addition Sb for the colourants. The detection limit is circa 0.2% for most of the elements. Concentrations of elements are reported in oxides (except for Cl). Synthetic glasses of the Smithsonian Institution (USA, Vicenzi et al. 2002) and antimony telluride (Sb_2Te_2) were used as standards.

Results

hXRF analysis

The chemical composition of the beads from Mezőtúr are reported in oxide form in **Table 1a**, except for Mg, which has concentrations below or around the detection limit (0.7 wt%).

Table 1a: Chemical composition of the beads measured by hXRF (elements are expressed in oxide form in wt%). (Number of measurements: Szolnok 1 blue & white: n=3, yellow: n=2; Szolnok 2 blue & white: n=2, yellow: n=1; Szolnok 3 yellow: n=1; v 71.2.145 blue: n=2, white: n=2, yellow: n=2; v 71.2.144 blue: n=2, white: n=2, yellow: n=2; v 71.2.53 blue: n=2, white: n=2, eyelet: n=1).

1a táblázat: A gyöngyök hXRF analízissel kapott kémiai összetétele (az elemeket oxidos formában, tömegszázalékban adtuk meg). (A mérések száma: Szolnok 1 kék & fehér: n=3, sárga: n=2; Szolnok 2 kék & fehér: n=2, sárga: n=1; Szolnok 3 sárga: n=1; v 71.2.145 kék: n=2, fehér: n=2, sárga: n=2; v 71.2.144 kék: n=2, fehér: n=2, sárga: n=2; v 71.2.53 kék: n=2, fehér: n=2, fűzőlyuk: n=1).

Beads and measurements	SiO_2	Fe_2O_3	K_2O	CaO	Sb_2O_3	MnO	CuO	CoO	PbO	Total
Szolnok 1 blue & white	60.81	1.41	0.71	6.14	1.31	0.01	0.18	0.05	0.24	70.86
Szolnok 1 yellow	29.23	0.63	0.33	3.31	0.60	0.00	0.00	<0.01	6.61	40.71
Szolnok 2 blue & white	66.73	1.35	1.83	6.47	0.96	0.01	0.19	0.05	0.34	77.93
Szolnok 2 yellow	24.40	0.32	0.50	2.05	0.39	0.00	0.00	<0.01	6.53	34.19
Szolnok 3 yellow	23.11	0.78	0.31	2.26	0.52	0.00	0.00	<0.01	7.56	34.54
v 71.2.145 blue	74.24	1.80	0.51	7.64	0.06	0.01	0.10	0.03	0.01	84.40
v 71.2.145 white	57.38	1.41	0.16	5.43	0.36	0.05	0.06	<0.01	0.15	65.00
v 71.2.145 yellow	19.48	0.95	0.26	2.00	0.44	0.00	0.00	<0.02	7.23	30.36
v 71.2.144 blue	70.30	1.40	0.63	7.25	0.11	0.01	0.11	0.06	0.07	79.94
v 71.2.144 white	65.38	1.60	0.39	7.73	0.19	0.01	0.08	<0.02	0.08	75.46
v 71.2.144 yellow	27.62	1.81	0.36	3.04	1.15	0.01	0.00	<0.03	10.66	44.65
v 71.2.53 blue	78.12	1.52	1.05	7.40	0.14	0.24	0.13	0.10	0.04	88.74
v 71.2.53 white	57.96	0.72	1.08	5.54	0.53	0.10	0.05	<0.01	0.09	66.07
v 71.2.53 eyelet	76.21	1.18	1.50	6.01	0.00	0.24	0.27	0.10	0.06	85.57

Table 1b: Chemical composition of the beads measured by hXRF and normalised to 90 wt% oxide totals.

1b táblázat: A gyöngyök hXRF analízissel kapott kémiai összetétele 90% oxidösszegre normálva.

Beads and measurements	SiO_2	Fe_2O_3	K_2O	CaO	Sb_2O_3	MnO	CuO	CoO	PbO	Total
N Szolnok 1 blue & white	77.24	1.79	0.90	7.80	1.66	0.01	0.22	0.06	0.31	90.00
N Szolnok 1 yellow	64.62	1.39	0.72	7.32	1.33	0.00	0.00	<0.02	14.61	90.00
N Szolnok 2 blue & white	77.06	1.56	2.11	7.47	1.11	0.01	0.22	0.06	0.40	90.00
N Szolnok 2 yellow	64.23	0.84	1.32	5.39	1.03	0.00	0.00	<0.03	17.19	90.00
N Szolnok 3 yellow	60.22	2.03	0.81	5.89	1.35	0.00	0.00	<0.03	19.70	90.00
N v 71.2.145 blue	79.18	1.92	0.55	8.14	0.06	0.01	0.10	0.03	0.01	90.00
N v 71.2.145 white	79.45	1.95	0.22	7.52	0.50	0.07	0.08	<0.01	0.21	90.00
N v 71.2.145 yellow	57.74	2.82	0.77	5.93	1.30	0.01	0.00	<0.06	21.43	90.00
N v 71.2.144 blue	79.14	1.58	0.71	8.16	0.12	0.01	0.12	0.07	0.08	90.00
N v 71.2.144 white	77.99	1.91	0.47	9.22	0.22	0.01	0.09	<0.02	0.09	90.00
N v 71.2.144 yellow	44.94	2.95	0.58	4.95	1.87	0.01	0.00	<0.05	34.69	90.00
N v 71.2.53 blue	79.23	1.54	1.07	7.51	0.14	0.24	0.13	0.10	0.04	90.00
N v 71.2.53 white	78.96	0.98	1.47	7.55	0.72	0.13	0.07	<0.01	0.12	90.00
N v 71.2.53 eyelet	80.15	1.24	1.58	6.32	0.00	0.25	0.29	0.10	0.06	90.00

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In the yellow glass of Szolnok 1–3 beads SiO_2 concentrations are lower (23–29 wt%) than in the stratified eyes (cc. 60–66 wt%). K_2O amounts are different in the eyes and in the yellow glass, however, the average K_2O content is 0.5–1 wt%, except for one eye of Szolnok 2 (cc. 1.83 wt% K_2O). CaO values in the blue and white eyes are about 6 wt% or a bit higher, whereas in the yellow glass only cc. 2–3 wt%. The eyes have a cc. 1.5 wt% Fe_2O_3 content, in the yellow glass Fe_2O_3 content barely reaches 0.4–0.8 wt%. The CoO and CuO contents of the eyes are 0.05–0.06 wt% and cc. 0.2 wt%, whereas in the yellow glass the concentrations of these elements are under the detection limit (CoO: 0.005 wt%; CuO: 0.01 wt%). In the eyes Sb_2O_3 is about 1 wt%, in the yellow glass it is below 1 wt%. The highest PbO values are measured in the yellow glass (cc. 6–7 wt%), whereas in the eyes PbO content are only 0.2–0.3 wt%. In general, the oxide totals of the eyes are much higher (70–77 wt%) than that of the yellow glass (34–41 wt%).

The chemical composition of beads from Vác are given in oxide form in **Table 1a**. In blue and white glasses SiO_2 content varies between 57 and 78 wt%, whereas in yellow ornaments it is only 19–27 wt%. In all three coloured glasses K_2O is less than 1 wt% or almost reaches 1 wt%, except for a simple eye bead (71.2.53) with 1–1.5 wt% K_2O content. CaO amounts are usually higher than 5 wt%, except for the yellow decorations, which have 2–3 wt% CaO content. The 71.2.53 bead has cc. 0.2 wt% MnO content. Fe_2O_3 contents are usually about 1–1.5 wt%. However, both bobbin beads show higher iron content, blue glass of bead 71.2.145 has 1.80 wt% Fe_2O_3; and the yellow ornament of bead 71.2.144 has 1.81 wt% Fe_2O_3 content. CoO and CuO concentrations in blue glass reach 0.03–0.10 wt%, and 0.10–0.27 wt%, respectively. Highest antimony and lead concentrations are measured in the yellow ornaments (0.44–1.15 wt% Sb_2O_3 and 7.23–10.66 wt% PbO), although antimony content is also high in the white glass (0.19–0.53 wt% Sb_2O_3). The PbO and Sb_2O_3 contents are lower in blue glass (0.01–0.07 wt% Sb_2O_3 and 0.06–0.14 wt% PbO).

Electron microprobe analysis

Only the yellow glass of Szolnok 1–3 beads were analysed, the average chemical composition of the vitreous matrix and the inclusions are reported in **Tables 2.** and **3.**, respectively.

According to the backscattered electron images, the heterogeneous glassy matrix shows lighter and darker grey strips. The darker ones contain less bright inclusions (**Fig. 3.**) than lighter ones. Furthermore, within the darkest strips of Szolnok 1 bead inclusions are totally absent (**Fig. 3/1/a**). The average PbO content in the matrix is cc. 9 wt% (**Table 2.**).

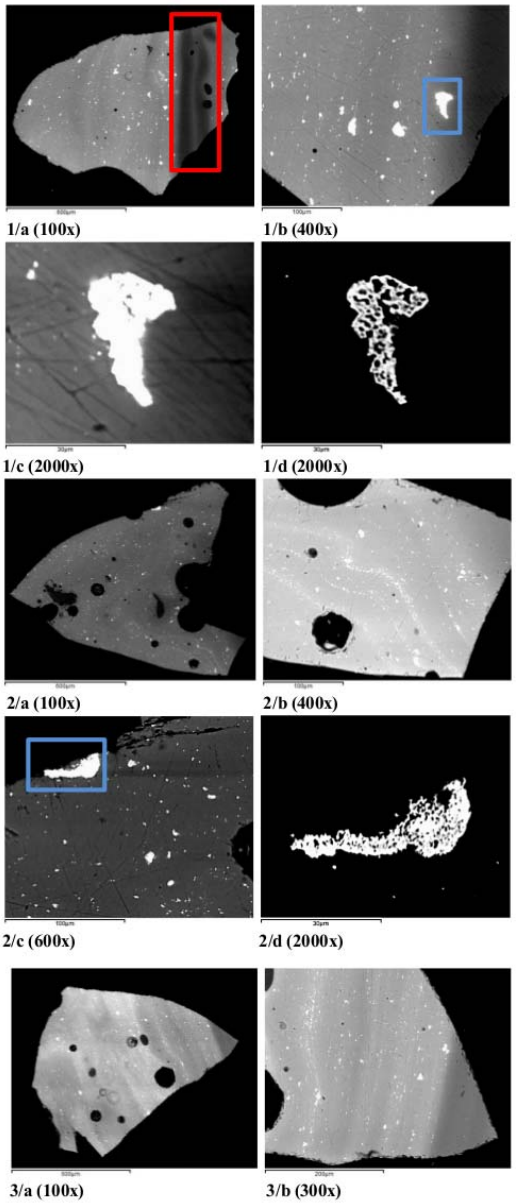


Fig. 3.: Backscattered electron images of Szolnok 1 (1/a–d), 2 (2/a–d) and 3 (3/a–b) yellow glass samples. Bright lead antimonate aggregates are present in the heterogenous glassy matrix (grey). The black spots mark the pores. In the darker grey strips less bright inclusion are present, whereas in the darkest parts of Szolnok 1 (red frame) inclusions are absent. In **Fig. 2/b** the pigments ordered in lines are well visible. The enlarged figures of inclusions in blue frames are in the following images. Magnifications are shown under the figures in parentheses. In **Figs. 1/c–d** a porous lead antimonate inclusion, and in **Figs. 2/c–d** a heterogenous and porous lead antimonate aggregate is visible.

3. ábra: A Szolnok 1 (1/a–d), 2 (2/a–d) and 3 (3/a–b) gyöngyök sárga üvegmintáinak visszaszórtelektron-képei. A szürke, heterogén üvegmátrixban világos (fehér) ólom-antimonát aggregátumok láthatók. A fekete foltok a pórusokat jelölik. A sötétszürke sávokban kevesebb zárvány van, míg a Szolnok 1 legsötétebb sávjában (vörös keret) egyáltalán nincs zárvány. A 2/b ábrán megfigyelhető a zárványok vonalak menti elrendeződése. A kék négyzettel keretezett zárványok kinagyított képei a következő ábrákon láthatók. A nagyítás mértékét az ábrák alatt adtuk meg zárójelben. Az 1/c–d és a 2/c–d ábrák egy-egy porózus, heterogén ólom-antimonát zárványt mutatnak.

Table 2.: Average chemical composition of glassy matrix of yellow glass of Szolnok 1–3 beads measured by EMPA (in wt%, st. deviation in parentheses, number of measurements: Szolnok 1: n=14; Szolnok 2: n=13; Szolnok 3: n=12).

2. táblázat: A Szolnok 1–3 sárga üvegminták mátrixának elektron-mikroszondás analízissel kapott átlagos kémiai összetétele (tömegszázalék, a szórás zárójelben, a mérések száma: Szolnok 1: n=14; Szolnok 2: n=13; Szolnok 3: n=12).

Samples	SiO_2	Al_2O	Fe_2O_3	Na_2O	K_2O	CaO	MgO	PbO	Cl	Total
Szolnok 1	67.4 (1.07)	2.08 (0.28)	1.02 (0.61)	11.45 (0.81)	0.43 (0.10)	6.81 (0.38)	0.51 (0.23)	10.10 (1.60)	0.64 (0.09)	100.39 (0.84)
Szolnok 2	63.64 (1.16)	2.21 (0.25)	1.15 (0.27)	16.03 (0.54)	0.68 (0.12)	6.45 (0.35)	0.48 (0.18)	8.74 (0.66)	0.84 (0.12)	100.21 (1.37)
Szolnok 3	63.48 (2.19)	2.24 (0.27)	0.95 (0.44)	15.65 (0.95)	0.67 (0.12)	6.63 (0.60)	0.47 (0.20)	9.08 (2.01)	0.93 (0.11)	100.1 (1.31)

Table 3.: Average chemical composition of inclusions in the Szolnok 1–3 yellow glass samples measured by EMPA (in wt%, st. deviation in parentheses, number of measurements: Szolnok 1: n=26; Szolnok 2: n=22; Szolnok 3: n=20).

3. táblázat: A Szolnok 1–3 sárga üvegminták zárványainak elektron-mikroszondás analízissel kapott átlagos kémiai összetétele (tömegszázalék, a szórás zárójelben, a mérések száma: Szolnok 1: n=26; Szolnok 2: n=22; Szolnok 3: n=20).

Samples	SiO_2	Al_2O_3	Fe_2O_3	Na_2O	CaO	Sb_2O_3	PbO	Cl	Total
Szolnok 1	25.85 (12.40)	1.10 (0,46)	3.51 (0.69)	6.11 (2.90)	4.47 (1.24)	22.46 (5.70)	36.09 (7.10)	0.36 (0.19)	99.99 (7.56)
Szolnok 2	17.84 (9.10)		3.83 (1.06)	6.69 (3.16)	3.12 (0.74)	21.01 (6.20)	34.08 (7.40)		86.57 (10.03)
Szolnok 3	14.70 (7.39)		4.68 (0.80)	4.79 (2.14)	3.43 (0.91)	27.03 (5.46)	42.40 (7.32)		97.04 (9.53)

The darker strips also differ from the lighter ones in chemical composition showing lower (0.6–3.08 wt%) PbO content. The SiO_2 content of the matrix is approx. 60–70 wt%. Na_2O content is higher than 10% (11.45–16.03 wt% Na_2O). K_2O concentration is cc. 0.4–0.6 wt%, but MgO content barely reaches 0.5 wt%. In all samples CaO amount is 6–7 wt%, whereas Al_2O_3 content is slightly higher than 2 wt%, and Fe_2O_3 content is approx. 1 wt%.

Bright inclusions of 1 to 40 µm, up to 50 µm in size (**Fig. 3/1/c, d; 2/c, d**) occur in the glassy matrix, furthermore they are organized to lines parallel to the greyish strips (**Fig. 3/2/b, 3/a, b**). Inclusions are heterogeneous (**Fig. 3/1/d; 2/d**), in their pores the components of glassy matrix are detected: beside the high amounts of PbO (30-40 wt%) and Sb_2O_3 (20-30 wt%), respectively, Na_2O (2–12 wt%), SiO_2 (5–40 wt%), CaO (3–4 wt%) and occasionally Al_2O_3 and Cl occur (**Table 3.**). In addition, the inclusions show elevated iron content (3–4 wt% Fe_2O_3) (**Table 3.**) compared to the matrix.

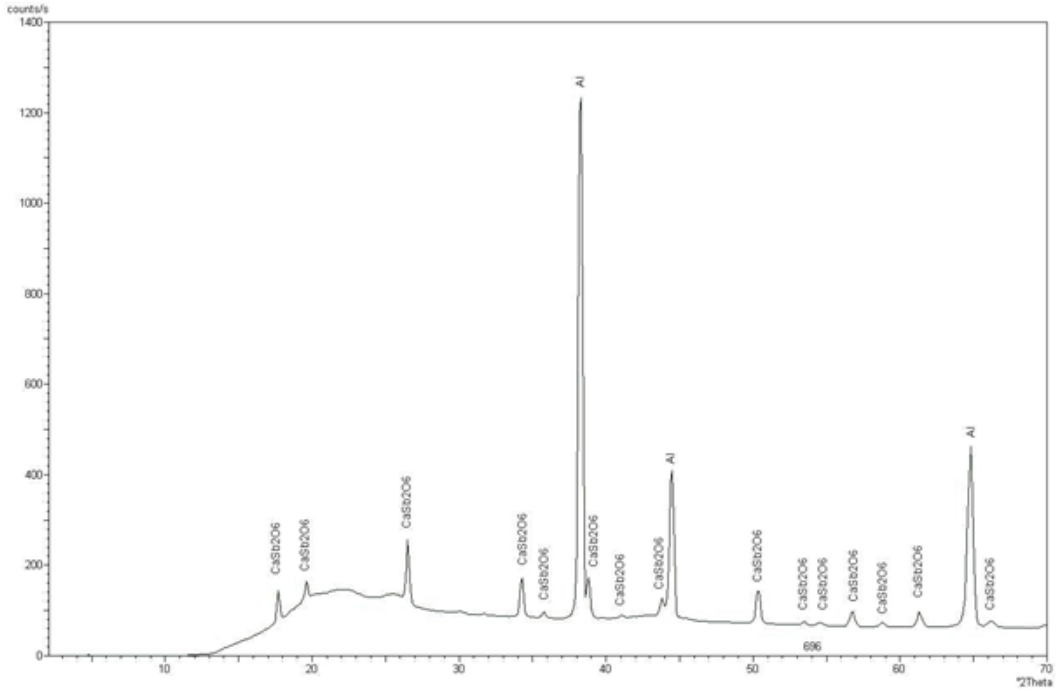
µ-XRD analysis

In the white glass samples of Szolnok 3 bead calcium antimonate ($Ca_2Sb_2O_6$) is detected (**Fig. 4a**), whereas in the yellow glass samples of Szolnok 1–3 beads lead antimonate ($Pb_2Sb_2O_7$) is identified (**Fig. 4b**).

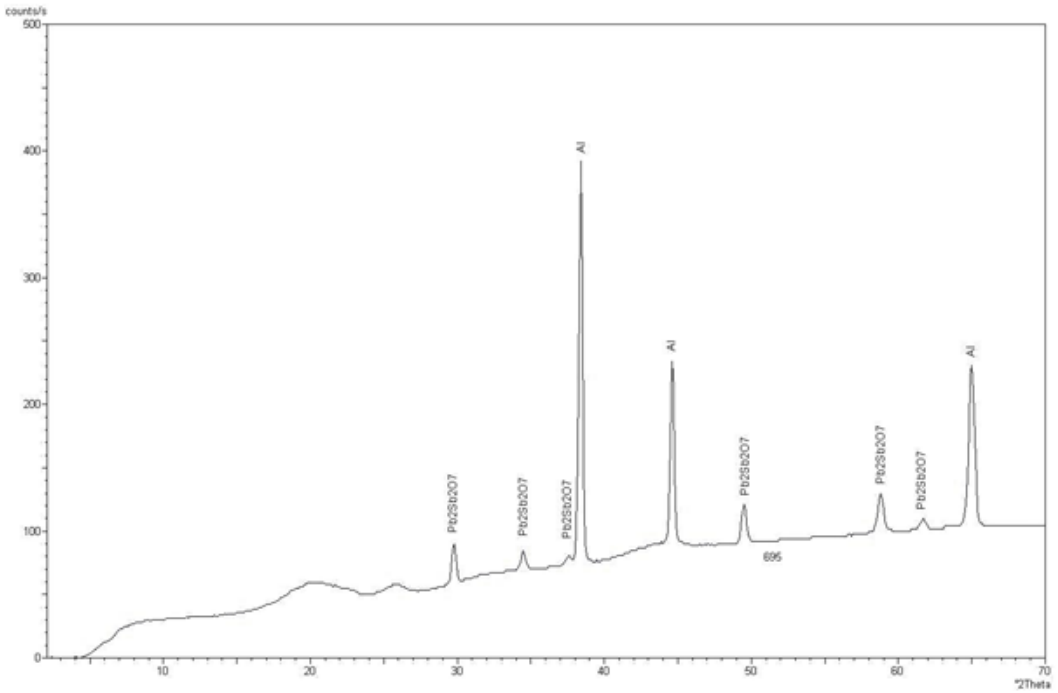
Discussion

Base glass

During Late Bronze Age two main types of glass occurred, which can be distinguished by the used alkali flux. At the beginning (15th century BC) plant ash-silicate glass was produced in both Mesopotamia and Egypt. At around the 10th century BC soda-lime-silicate glass appeared in ancient Egypt relating to the resources of natron or trona. This type of glass contains low magnesia and potash concentrations (MgO and K_2O lower than 1.5 wt%).



a



b

Fig. 4a: μ-XRD pattern of white glass sample scraped from a stratified eye of Szolnok 3 bead showing the peaks of calcium antimonate (Al: aluminum sample holder).

Fig. 4b: μ-XRD pattern of yellow glass sample from Szolnok 3 bead showing the peaks of lead antimonate (Al: aluminum sample holder).

4a ábra: A Szolnok 3 gyöngy egyik pávaszeméből vett fehér minta μ-XRD vizsgálatának eredménye. A diffraktogramon a kalcium-antimonát csúcsai láthatók (Al: alumínium mintatartó).

4b ábra: A Szolnok 3 sárga üvegminta μ-XRD vizsgálatának eredménye. A diffraktogramon az ólom-antimonát csúcsai láthatók (Al: alumínium mintatartó).

The other type of glass made of using plant ash has higher magnesia and potash content (MgO and K_2O higher than 1.5 wt%), besides the occurrence of phosphorous (P_2O_5 higher than 0.2 wt%). Both glass types belong to the sodic glasses, and differ not only in chemical composition, but in the region and ages they were spread. In spite of the abundant chemical data about the glasses from the Mediterranean region and the Near East, there are less glass compositional data from the the Iron Age Europe (Fórizs 2008; Henderson 1985; Henderson 2000; Rehren & Freestone 2015; Shortland et al. 2006).

In the yellow glass of the studied beads, the oxide totals and the concentrations of some elements (especially SiO_2) measured by hXRF are low. One reason for this phenomenon is that the measured surface is not flat, but curved, which is far from the ideal resulting in a significantly lowered total. Other explanation can be that much more colourant was added to the yellow glass than to the white or blue glasses, therefore the concentrations of other components seem to be much lower. The mentioned reasons could have a negative effect on the compositional data, thus all the hXRF results were normalized to 90 wt% oxide totals (**Table 1b**). The reason of normalizing to 90 wt% instead of 100 wt% total is that Na_2O is not measured by hXRF, however, its amount is at least 10 wt% based on the typical composition of sodic glasses.

According to the normalized hXRF values, the low K content (approx. 1 wt%, up to 2 wt% K_2O) and the Mg concentration below or around the detection limit in all beads indicate that glasses, including decorations, were made of soda-lime-silicate glass. It is supported by the much more reliable EMPA results of the yellow glass of Szolnok 1–3 beads showing K_2O and MgO contents below 1 wt% together with 11.45–16.03 wt% Na_2O.

Beside the main components of blue and white glasses the hXRF analysis showed low amounts of lead (0.01–0.3 wt% PbO), which might have been added to glass during production to facilitate forming (Nagy et al. 2010). In the case of Szolnok 1–3 eyes another explanation can be the wide measurement area, which included not only the blue-white eye, but the surrounding yellow glass as well.

Based on the above, it seems that soda-based glass has been certainly present in the Carpathian Basin (or in a smaller region, in present-day Hungary) since the 6/5th century BC, then occurred in La Tène ages as well (about the spread of soda-lime-silicate glass see Shortland et al. 2006; Fórizs 2008). Based on the compositional similarities we suppose the idea of continuous use of glass or raw materials of the same "workshop". We have only little information about Iron Age glass production centres using perhaps various recipes and about their typical glassware. Apart from the above mentioned, our results cannot be related to any of the centres with published compositional data, because specific impurities related to the raw materials or workshops were not detected (Caley 1962; Fórizs et al. 2012). Thus, the production centre of the six beads under study cannot be located unambiguously.

Colourants

Opaque white

The μ-XRD result indicates that in the white glass of a stratified eye of Szolnok 3 bead the colourant is calcium antimonate ($Ca_2Sb_2O_6$). In the case of beads from Vác only the hXRF results are available, which show elevated amount of Sb_2O_3 (cc. 0.2–0.7 wt%) compared to the blue glass. Accordingly, we suppose that the colourant of white decorations of all beads from Vác and Szolnok/Mezőtúr is calcium antimonate.

Colouring and opacifying with calcium antimonate was a well-known technology in the Near East region from about 15th century BC (LBA) (Brill 1970). Calcium antimonate is an artificial pigment, which can be produced in two ways. It is generally accepted that antimony was added to glass, and then antimony and calcium together formed calcium antimonate crystals in situ in the molten glass during cooling (Duckworth et al. 2012). According to a recent study, pigment could be prepared in advance, then added to the glass batch (Lahlil et al. 2010). It is particularly important to identify and differentiate the production technologies, which can help in determining workshops. Therefore, further studies concentrated on the inclusions may help us ascertain which method was applied to the studied beads.

Transparent blue

In the blue glass of all beads the blue (or rather ultramarine) colour is caused by the simultaneous presence of cobalt and copper in approximately the same amount according to the hXRF results. The combination of these two elements as colourants was used since the mid–2nd millennium BC (Smirniou & Rehren 2013), as it is supported by glass deriving from the time of the New Kingdom, Egypt, or Late Bronze Age Mycanean glass (Smirniou & Rehren 2013) and a few glass beads from Italy dated to the Early Iron Age (Arletti et al. 2010; Polla et al. 2011: cobalt and copper together with higher iron content; Olmeda et al. 2015). In addition, we know examples among the Celtic glass bracelets (Haevernick 1960; Girdwoyń 1986; Frána et al. 1987; Wobrauschek et al. 2000; Roymans et al. 2014). In the beads from Apollonia Pontica, an ancient Greek colony near the Black Sea (nowadays in Bulgaria, city of Sozopol), the blue colour is

supposed to be obtained by the combination of iron, cobalt and copper colourants (Lyubomirova et al. 2014).

In conjunction with the researchers of Celtic glass bracelets (Haevernick 1960; Henderson 1985; Roymans et al. 2014), we suppose the intentional use of both colourants. In our opinion, the reason of employing this colouring technique is related to the mineral resources, in which cobalt and copper occur within the same mineral association, and these well-known raw materials were consciously sought in Antiquity. Due to the lack of any detectable impurities, we cannot yet determine where the raw materials derived from, however, they have several deposits all over Eurasia (Bouladon 1989; Smirnov 1989; Zuffardi 1989; Hall & Yablonsky 1997; Gliozzo et al. 2012), and a common provenance with the raw materials of the blue glass of beads from Apollonia Pontica cannot be excluded either.

Opaque (ochre) yellow

According to the μ-XRD analysis the opaque yellow colour is due to the presence of lead antimonate (lead pyroantimonate, $Pb_2Sb_2O_7$) in the Szolnok 3 bead. In addition, the electron microprobe analysis detected significant amounts of iron beside lead and antimony in the pigment inclusions.

The glassy matrix is characterised by darker and lighter grey zones in the backscattered electron images with heterogeneous and porous inclusions arranged along lines. Lower amount of inclusions occurs in darker strips with lower lead content in the glassy matrix. Furthermore, the darkest parts of Szolnok 1 bead in the backscattered electron images (**Fig. 3/1/a**), looking grey macroscopically as well, have 0.6–3.1 wt% PbO content, that can be considered as the incipient PbO content of the original transparent glass. All the afore-mentioned results refer to a rapid production method, which caused unequal dispersion of pigments during molding (stretching) or colouring glass in haste, thus preventing lead to dissolve from its pigment (Tite et al. 2008; Duckworth et al. 2012; Molina et al. 2014) and ordering them in lines resulting in zoned appearance of matrix. In spite of rapid production, the Pb/Sb ratio (3:2=1.5) of the inclusions lower than stochiometric (Pb/Sb = 1.64) (Wainwright et al. 1987), as well as the cc. 10 wt% PbO content of glassy matrix indicate that PbO was partially released to the matrix.

The iron content (approx. 3–4 wt% Fe_2O_3) of the inclusions (**Table 3.**), higher than that of the glassy matrix (1 wt% Fe_2O_3, **Table 2.**), had a role in preventing the dissolution of lead, along with the control (or preservation) of the colour shade. However, antimony connected to lead, which is reactive with silica, serves for the stabilization of the pigment (Lőcsei & Tamás 1982; Wainwright et al. 1987; Dacapito et al. 2012; Molina et al. 2014), iron also serves as reducing and stabilizing agent as it was proven in the case of opaque red glasses (Brill & Cahill 1988; Fórizs et al. 1999; Fórizs 2008), as well as yellow glasses from Egypt dated to the 10th century BC and Roman age yellow glasses (Molina et al. 2014). Thus, using an additional component, production circumstances did not need as much care as without iron. On the other hand, iron causes changes in tone, therefore the ochre yellow shade of the beads can be related to this effect (Wainwright et al. 1987; Bultrini et al. 2006; Bajnóczi et al. 2009; Molina et al. 2014).

Based on the normalized hXRF data, the high PbO and Sb_2O_3 values (**Table 1b**) in yellow ornaments of bobbin beads refers to the use of lead antimonate. hXRF also measured higher iron (almost 3 wt% Fe_2O_3) concentrations in the yellow glass compared to the blue and white glasses of the same beads. Therefore, we assume that in the case of bobbin beads, the same as eye beads with bosses, the yellow glass was made of a special, iron-bearing lead antimonate supporting the idea of technological transfer amongst the two cultures in spite of the small number of analysed samples, and the lack of known compositional data of glasses from the period of 6th/5th century BC to 3rd century BC.

The hXRF and EMPA measurements did not detect specific trace elements in yellow glass, thus it is impossible to determine or localise the source of raw materials. If iron is considered as an impurity, the location of source(s) is still unknown (Rosi et al. 2008; Duckworth et al. 2012). Furthermore, it is not decided whether iron addition to the colourant was intentional (deliberate addition of iron to the lead and antimony compounds to produce lead antimonate was first described in the late Middle Ages in connection with production of the Italian maiolica, Wainwright et al. 1987; Bultrini et al. 2006; Bajnóczi et al. 2009), or iron was added accidentally as natural contaminant from the source (Wainwright et al. 1987; Molina et al. 2014). The issue becomes more complicated due to the absence of numerous analogies. Some of already published data might suggest the use of iron-bearing lead antimonate. Elevated iron content was detected besides lead antimonate in the yellow glass of similar, but bossless eye beads and other yellow glass artefacts from Italy dated to the 6th–4th century BC (Arletti et al. 2010: 0.9–1 wt%) and in the yellow ornaments of glass bracelets characteristic to the late La Tène phase and deriving from Poland, in the latter case the iron content of the yellow glass was twice as much as that of the blue glass (Girdwoyń 1986). Regarding to its correlations in archaeological sense, Apollonia Pontica deserves special attention. From compositional point of view, not only blue glass,

but yellow glass excavated here corresponds to the beads we have investigated, if we accept that the high iron content of the yellow glass mainly belongs to lead antimonate colourant (Lyubomirova et al. 2014). It is important to emphasize that all the mentioned data is ambiguous and indefinite, because beads were analysed non-destructively, that is investigations focused on pigments are lacking. For instance, Apollonia Pontica beads were measured by PIXE and PIGE, well-known methods to analyse the whole artefact without sample preparation (Lyubomirova et al. 2014). On the other hand, there is a great difference between the used analytical methods (and results, of course), which affects the comparison negatively. Only one certain study is to be mentioned, which was a unique investigation concentrating on yellow colouring pigments in Egyptian and Roman glass and interpreted the occurrence of iron in lead antimonate as impurity from the raw materials of the colourant (Molina et al. 2014).

It is not clear either whether iron served only to reach another shade of yellow (Girdwoyń 1986) or to stabilize the pigment and keep the yellow colour, since long-lasting heating causes that glass turns to white instead of yellow (Fórizs 2008), ergo facilitating the production technology. After all the remaining questions, it can be concluded that use of iron-bearing lead antimonate supposes a level of consciousness at least in selecting the raw materials. Furthermore, it represents the third yellow colouring technique beside simple lead antimonate and lead antimonate with glass anime related to Antiquity (Molina et al. 2014). Despite that the number of production centres can be restricted based on the used colouring technique, the above-mentioned reasons as well as the sporadic data about yellow glass do not allow us to determine any workshop. It seems that this sort of colouring technique could be first employed (maybe invented as well) in 10th century BC Egypt, and we can presume its continuous usage in the following times.

Conclusions

The studied Scythian eye beads with bosses, the Celtic bobbin beads and the Celtic simple eye bead were made of soda-lime-silicate glass. The colourants are cobalt and copper together in blue glass, calcium antimonate in opaque white glass and iron-bearing lead antimonate in opaque ochre yellow glass. The similarity of these beads belonging to two distinct cultures in aspects of chemical composition, especially the glass type and the colourants of blue and yellow glasses, cannot be the result of accidental coincidence, but technological continuity. A circle of workshops (i.e. the ideology), probably functioning for a few centuries, can be supposed to produce these beads. This idea is best demonstrated by the use of iron-bearing lead antimonate, which can be a good explanation to the common provenance as well. Besides, the latter fits well to the similar design and function of the beads already described.

Comparing with the previously cited archaeological and archaeometric analogues, it can be supposed that the Mezőtúr beads were imported by Scythians most probably from the Black Sea (or Pontic) region, whereas local production (in the Carpathian Basin) of bobbin beads cannot be proved unambiguously so far due to the similarities with Celtic glass bracelets and Scythian eye beads with bosses at the same time. However, in the case of bobbin beads the existence or the role of a migrating craftsman is also probable. Apollonia Pontica, which is unique amongst the excavated ancient Greek colonies because of available archaeometric data of the glass beads, has a special role in the derivation of these two types of beads. The compositional data of Apollonia Pontica glass beads compared to the Mezőtúr beads and the bobbin beads indicate that they were produced using the same recipe and/or from the same sources, thus the common origin can be assumed, or at least a technological continuity can be supposed. Presently it is supposed that Apollonia Pontica imported several goods including glass jewellery possibly from the Near East or Rhodes, which means that the colony could even play mediating role (Lyubomirova et al. 2014; Chacheva 2015; Boțan & Chiriac 2016). On the contrary, other compositional parallels are known from the Iron Age Europe and the Mediterranean, like Egypt, Italy, and the LT bracelets. It stands on the simple eye bead as well, thus its probable provenance could be localized to Pontus or perhaps Rhodes (Lyubomirova et al. 2014), and further the parallels found in middle Italian region cannot be denied. Besides, the Bohemian analogues complicate the question of relationships among the cultures lived in the Carpathian Basin in the Middle and Late Iron Age.

Although these are not the final results of a complex and long-term study, and more work is needed, we got closer to answer the most interesting archaeological questions and succeeded in creating the basis of the Iron Age glass database of Hungary. After all, it is obvious that a focused analysis (e.g. using Raman microspectrometry on yellow glasses, and EMPA on white ones) involving a wider range of artefacts (primarily eye beads with bosses, other bobbin beads, eye beads and mask beads and finally glass bracelets) is necessary, and sampling cannot be avoided.

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