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The Provenance of Raw Materials of the Second Complex (2018) of Stone Casting Moulds from the Archaeological Monument of Tokivske-1 (Ukraine)

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ABSTRACT

Petrographic research was undertaken on the second casting moulds complex discovered in 2018 at the archaeological monument Tokivske-1 in Ukraine. Metalworking of the Late Bronze Age in the Northern Black Sea region is known for its utilisation of stone moulds made from meta-ultrabasites ("talc schists") by representatives of the Sabatynivka culture. Field investigations in the vicinity of Tokivske indicated the probable place of stone mining, from where a considerable part of the casting moulds under study were made. The discovery of the place of stone mining near Tokivske-1 and finds of similar moulds in other regions give grounds for considering the monument to be not only a centre of metalworking but also stone-processing. The obtained data are also the first geologically proved evidence that the Kryvyi Rih area, which was previously considered the main supplier of "talc schists", was not the only place of mining of raw materials for the manufacturing of casting moulds in the Middle Dnipro area. Both complexes, the second and the previous one, found in 2017, contain matrices made of identical raw materials, namely, the samples made are untypical for the Middle Dnipro area species. This fact indicates the synchronicity of both complexes and a significant volume of casting production in Tokivske-1.

1. Introduction

The archaeological monument of Tokivske-1, the study of which began in 2012 by the expedition of the Dmytro Yavornytsky National Historical Museum of Dnipro, appeared to be one of the most interesting archaeological sites of the Middle Dnipro area recently included in the archaeological map. The monument was discovered thanks to the active work of local historian Y. I. Serhiichuk, who informed the museum archaeologists about the existence of an interesting feature, located near the village of Tokivske in the Dnipropetrovsk Region (Figure 1). The village is known for its deposit of pink granites of the Tokivskyi complex as well as for geological landmarks such as a canyon and waterfalls in the River Kamianka valley. The discovery of a stone megalithic construction and a cult pit during the archaeological excavations was the first result of the work. Further archaeological excavations allowed the establishment

of three main cultural horizons on the Tokivske-1 site. The first, the lowest horizon, dates back to the Middle Bronze Age (Babyne Cultural Circle). The second horizon belongs to the Late Bronze Age (Sabatynivka Culture). The upper horizon belongs to the Early Iron Age and is represented by Scythian monuments (Starik, 2017).

In the first stage of the monument's study, it was interpreted only as a "megalithic place of worship". Megalithic constructions, similar to the one in Tokivske, are known in the Dnipro Rapids area (the rapids existed on the river Dnipro in its flow between the cities of Dnipro and Zaporizhzhia). These constructions are usually "located over the water, on islands or the second terrace, usually on a little cape, formed by a gully in the place of its flowing into the river Dnipro, less often on the third terrace" (Lahodovska, 1949). In addition to the megalithic construction, which existed for a long time and was visited by representatives of various tribes, an altar stone, lingam-like and anthropomorphic stelae, several cult pits, several thousands of pottery fragments, nearly 80 flint implements and 53 stone implements, and

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Figure 1. The map of the Middle Dnipro and the Azov Sea areas with the Tokivske-1 archaeological site location.

numerous osteological materials were found in Tokivske-1. Archaeological excavation was accompanied by petrographic research of the stone materials, which allowed the sources of origin of the stone implements and raw materials for their manufacture to be determined (Nikitenko et. al., 2018; Nikitenko et. al., 2019). Additionally, based on the analysis of the raw material of a stone pestle-sceptre, a connection between the Tokivske-1 site and the Donetsk Mining and Metallurgical Centre of the Late Bronze Age was recognised (Nikitenko et. al., 2019), which was the most developed on the territory of Ukraine thanks to the richest reserves of copper ore in the Bakhmut lobe area (Brovender and Okalelov, 2019).

The need to re-estimate the role of the archaeological monument of Tokivske-1 became apparent after the first hoard of stone casting moulds of the Late Bronze Age (consisting of three valves of moulds) was found on the site in 2017. Furthermore, minor fragments of two ceramic moulds and one stone mould were found nearby. It was suggested that there is a monument of ancient metalworking on the site. Metalworking centres were very common and flourished in the Northern Black Sea region during the Late Bronze Age, and played an essential role in its economic development. The key to this was the links with the mining and metallurgical centres of the Balkans, the Urals and the Caucasus (Sava and Boroffka, 2013). We should note that the conclusions about the nature and typology of the Tokivske-1 monument could only be made after the entire territory of the cape, on which it is located, had been studied.

The following year, the expedition found one more hoard of casting moulds, which was discovered at a distance of 26 m to the southwest of the previous one. The hoard comprised seven items: a single-sided mould with a cover, used for a rounded ingot or a mirror casting; a preformed block or a mould cover for a rounded product casting; the

central element of a four-sided mould for casting a flat adze and three chisels; a valve of a single-sided mould for a celt casting; two double-valved double-sided moulds for dagger casting. Not far from the hoard, a preformed block for a valve and three little fragments of casting moulds were also found.

A trade in preformed blocks and readymade stone moulds was evidenced on the territory of Ukraine in the Late Bronze Age. The main raw material for their production was metaultrabasite, mainly with a composition of talc-chlorite-amphibole. In the Northern Black Sea region, the extraction of meta-ultrabasite and the exchange trade in casting moulds and preformed blocks are associated with the tribes of the Sabatynivka Culture (16th – 13th centuries BCE) (Leskov, 1967). A petrographic study of the first complex showed the unusual provenance of the meta-ultrabasites used for the matrices manufacturing, an issue which requires further research (Nikitenko *et. al.*, 2020).

This paper is devoted to the petrographic research of the second complex of casting moulds, including the determination of the provenance of its stone raw materials and a comparison with the materials of the first complex matrices. The main goals of the research were to find the most probable place of ancient mining, as well as to elaborate new data for the concept of mining the meta-ultrabasites for the casting moulds production in the Late Bronze Age.

2. Petroarchaeological background

Petrographic analysis is widely used in the investigation of Bronze Age stone implements. Its application allows the provenance of different raw materials to be determined. I can be also a key to understanding the system of ancient stone mining and its delivery. Examples of recent use of such methods are studies of stone tools from the archaeological



monument of Wroclaw-Vidava 17 in Poland (Borowski, 2014), Bronze Age basalt artefacts from Israel (Gluhak and Rosenberg, 2018), polished Middle Bronze Age stone tools and weapons from Central Hungary (Farkas-Pető et. al., 2014), and stone artefacts of the Sinsongri Site in Korea (Chan Hee Lee et. al., 2014), as well as many others.

The raw materials of the Bronze Age stone moulds, widespread in the Northern Black Sea region, have been repeatedly studied by a number of authors. The most profound research of the casting moulds was that conducted by V. F. Petrougne in the late 1960s. The researcher, applying petrographic analysis of thin sections and the immersion method, studied most of the stone casting moulds in the museum collections of the European part of the former USSR. V. F. Petrougne supposed that the mining of raw materials for the casting moulds made of so-called "talc schists" was carried out on the territory of the Kryvyi Rih Iron Ore Basin (Petrougne, 1967).

Petrographic research of two hordes of casting moulds was presented in two publications by I. M. Sharafutdinova, who studied the economy of the Sabatynivka Culture (Sharafutdinova, 1973; Sharafutdinova, 1985). In both these works it was established that the rocks used as a raw material for the casting moulds production were composed of talc, chlorite and tremolite in different percentages with a prevalence of the amphibole. Except for the Kryvyi Rih Basin, I. M. Sharafutdinova supposed one other possible provenance of the moulds, namely the Dnipro Rapids area.

The conclusions of the research of V. M. Gorbov and O. M. Smirnova, who studied the hoard of the casting moulds found on the settlement of Shyroka Balka 2 in the North-East Azov Sea area, rely on the petrographic analyses performed by V. F. Petrougne. The authors determined that the raw material for most of the casting moulds originated from the Middle Dnipro area (Gorbov and Smirnova, 2005).

One of the authors of the current work established Kryvyi Rih as the provenance for the casting moulds found during excavations of the Kartamysh archaeological micro-district of the Late Bronze Age Donets Mining and Metallurgical Centre. All the studied casting moulds, found in different places, were produced from identical talc-chlorite schists that probably originated from the same deposit (Nikitenko, 2010).

In his monograph, dedicated to the ancient metallurgical production of Eastern Europe, V. S. Bochkaryov also emphasised the Kryvyi Rih origin of raw materials for the stone moulds (Bochkaryov, 2010). Thus, the opinion among researchers today is for the predominantly Kryvyi Rih provenance of raw materials of all matrices made of "talc schists" (meta-ultrabasites) found on the territory of Ukraine, with the probable use of similar rocks from other occurrences of the Ukrainian Shield, especially in the Dnipro Rapids area.

After discovering the first complex of moulds in Tokivske, we made a petrographic study of them. The research of raw material samples was carried out conducting petrographic analysis on transparent thin sections, as well as XRD analysis. As a result of the study, it was established that the moulds have a similar mineral composition. Their difference from the other moulds, described in the above-mentioned studies, is that a larger volume of amphibole is attributed to anthophyllite, rather than to tremolite or actinolite. One fact of much consequence is that talc-chlorite-anthophyllite meta-ultrabasites are not typical for the Kryvyi Rih structure, and are quite rare in the Middle Dnipro area, except for some occurrences of rocks with petrographic features and a different mineral composition than the raw materials of the studied casting moulds. All this give grounds to make assumptions concerning the possible non-local origin of the raw materials of casting moulds of the first complex, especially the anthophyllite-chlorite variety, in particular, from the Azov Sea area (Nikitenko et. al., 2020). Thus, petrographic research of the new complex discovered in Tokivske becomes essential for solving the issue of metaultrabasite mining in the Northern Black Sea region.

3. Methodology

To conduct the research, we were provided with nine out of the eleven samples of the complex found in 2018. The samples were represented by moulds, their fragments, and preformed blocks for casting moulds production (Figure 2; Table 1).

According to the results of the petrographic analysis, it was determined that all the studied samples belong to meta-

 Table 1. Studied samples.

Inventory number	Description
A-14025	Central element of a four-sided mould
A-14027	Double-sided mould for a dagger casting
A-10428	Valve of a mould for a celt casting
A-14029	Valve of a mould for a rounded product casting (a mirror or an ingot)
A-14030	Valve (cover) of a mould for a rounded product casting
A-14031	Valve preformed block for a rounded product casting
HB-41178	Preformed block or a casting mould cover
HB-41180	Fragments of a casting mould valve
HB-41181	Fragments of a casting mould valve



Figure 2. The studied collection of stone moulds, their fragments and preformed blocks.



ultrabasites. The raw materials of some casting moulds were practically identical, which allowed a division of the studied items into four groups. At least one sample from each group was examined by the XRD analysis. Samples of two groups were also studied using the XRF method. The XRD and XRF methods were also used to analyse specimens from the outcrops and compare them with the materials of moulds.

Samples for the research were taken from the artefacts, whose size and integrity allowed taking suitable pieces of rock to produce thin sections. The petrographic research was performed microscopically in transparent thin sections using a polarising microscope POLAM R-312. Provenance of the rocks was determined by comparing them with materials from geological survey reports, literary data and thin sections of similar rocks from the outcrops and the raw materials of casting moulds from other archaeological collections (Kryvyi Rih, Donbas, Dnipro Rapids area, Kyiv Region).

The XRD method helps to define the mineral composition of rocks more accurately. It is applied when some minerals are difficult to distinguish in thin section due to the similarity of the optical characteristics or a crystal's small size. The XRF analysis is used to determine the chemical composition of rocks. In the case of meta-ultrabasites, which originate from ultramafic igneous rocks, similar chemical features may indicate that they are from the same magma in origin.

There were five samples in the collection whose condition allowed the taking of some extra material for the XRD analysis. In addition, analyses of specimens from natural exposures were performed. The research was carried out in the Laboratory of Crystal Chemistry and Structural Analysis of the M. P. Semenenko Institute of Geochemistry, Mineralogy and Ore Formation of the National Academy of Sciences of Ukraine by the analyst E. E. Grechanovskaya. The samples were ground into powder specimens using a laboratory mill. The XRD analysis was carried out using a diffractometer DRON-2 on copper radiation ($Cu_{K\alpha} = 1.54178 \text{ Å}$) and an automatic diffractometer DRON-3M on cobalt radiation $(Co_{\kappa_0} = 1.78892 \text{ Å})$. The acquisition was performed in the interval of angles $4-65^{\circ} 2\theta$, with a scan step of 0.5 deg/sec. To identify the minerals, a standard file catalogue of the PDF-2 database of the International Centre for Diffraction Data (ICDD) 2003 was applied using PCPDFWIN software. The positions of the diffraction maxima on the XRD pattern were compared with the given reference values of the minerals of this database.

Three samples of moulds and a sample from an outcrop were compared using the XRF analysis. Two available specimens were analysed in powder. The analyses of a mould and a rock from the outcrop were performed on polished surfaces because of there being no possibility of making a powder specimen from the artefact material. The analyses were carried out by the analyst Y. S. Perkov in the Analytical Research Laboratory of the Dnipro University of Technology. The applied ElvaX Plus stationary spectrometer used is a desktop energy-dispersive X-ray fluorescence (EDXRF) analyser with 60 kV X-ray tube voltage and a helium purge feature that allows analysing the element

range from Na (Z=11) to U (Z=92). The device contains a helium purge system and automatic primary filter changer combined with an SDD detector that allows light elements spectra registration.

4. Results

Group 1 is the biggest in the studied collection. It consists of four samples: A-14025, HB-41180, A-14029, and A-14030. As a result of the XRD analyses of samples HB-41180 (Figure 3a) and A-14030 (Figure 3b), the following phases were obtained: chlorite-clinochlore, talc, anthophyllite and actinolite. In thin sections, the rocks had similar petrographic features and were defined as talc-amphibole-chlorite schists (Figure 4a). The mineral composition of the rocks of this group, determined under a microscope, is as follows: chlorite, tremolite, anthophyllite, talc, magnetite and goethite. The studied samples differ in the content of chlorite - from 60 vol.% in the sample A-14025 to 75 vol.% - in the sample HB-41180. We have to mention the presence of two amphiboles in the rocks: tremolite and anthophyllite, which make up about one-third of the rock volume. Since most of the amphibole crystals are colourless under parallel nicols and have an inclined extinction at an angle of up to 23°, they most probably belong to tremolite. The crystals present with parallel extinction, on the assumption of the XRD analysis data, most likely belong to anthophyllite. Chlorite in thin sections is often represented by aggregates, which morphologically resemble elongated crystals of amphibole. In some cases, crystals with low birefringence, as in chlorite, have inclined extinction and are most likely represented by homoaxial pseudomorphoses. Talc is represented by scales with excellent cleavage, as well as scaly aggregates, colourless under plane-polarised light, which have high colours of interference under crossed nicols. Magnetite is represented by opaque grains, intensively changed into iron hydroxides. The texture of the rocks is glomeronematolepidoblastic.

Group 2 includes samples of HB-41178 and HB-41181. The rocks differ from other groups by their micrograin structure, as well as mineral composition. According to the results of XRD analysis of the HB-41181 sample, three phases were found: talc, clinochlore (chlorite) and, in small quantities, actinolite (Figure 3c). As a result of the examination of samples in thin sections, it was determined that the amphibole of the isomorphic series actinolite-tremolite is almost colourless and rather belongs to tremolite. The mineral composition of rocks determined microscopically is as follows: chlorite, talc, tremolite, ore mineral and goethite. Thus, both samples were identified as tremolite-talc-chlorite schists. Chlorite makes up the bulk of the rock, has a greenish colour, low interference colours, and parallel extinction. The mineral is represented by microscales that form aggregates and replace the crystals of amphibole. Talc is represented by microscales, morphologically similar to chlorite. Against the background of other crystals, it has higher birefringence. Tremolite is represented by tabular and

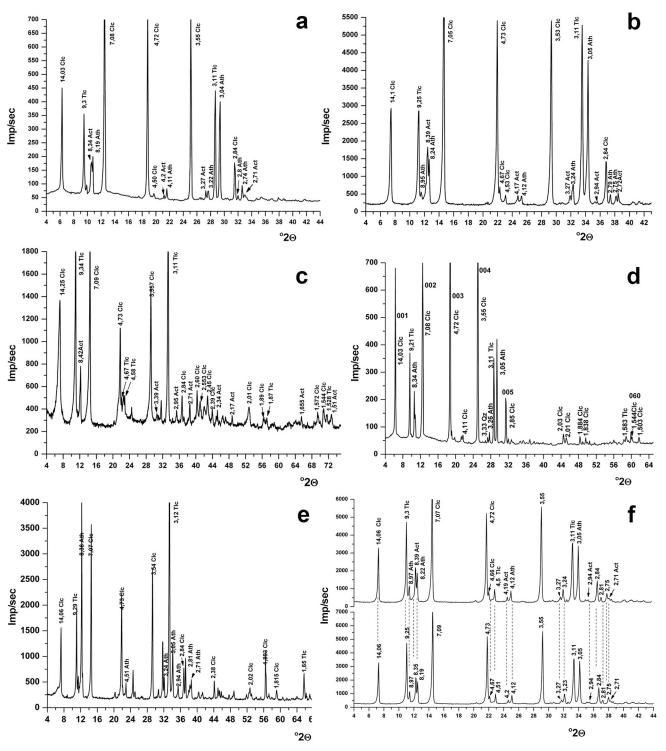


Figure 3. XRD patterns: a – HB-41180 (Group 1); b – A-14030 (Group 1); c – HB-41181 (Group 2); d – A-14027 (Group 3); e – A-14031 (Group 4); f – 6/3 (Bazavluk River, outcrop). Act – actinolite; Clc – clinochlore (chlorite); Ath – anthophyllite; Tlc – talc.

needle-like crystals with cleavage, located mainly crosswise or at an angle to the schistosity, often forming aggregates (Figure 4b). The mineral is colourless under plane-polarised light. The extinction is inclined. Much of the crystals are almost completely chloritised. The ore mineral is represented by opaque grains of irregular shape. Goethite develops as

a secondary mineral in chlorite as well as forming dispersed impurity in the rock.

Group 3 has two identical samples, A-10428 and A-14027. The XRD analysis of A-14027 sample revealed such phases, as chlorite-clinochlore, talc and anthophyllite (Figure 3d). XRF analyses of both samples showed their similarity (Table



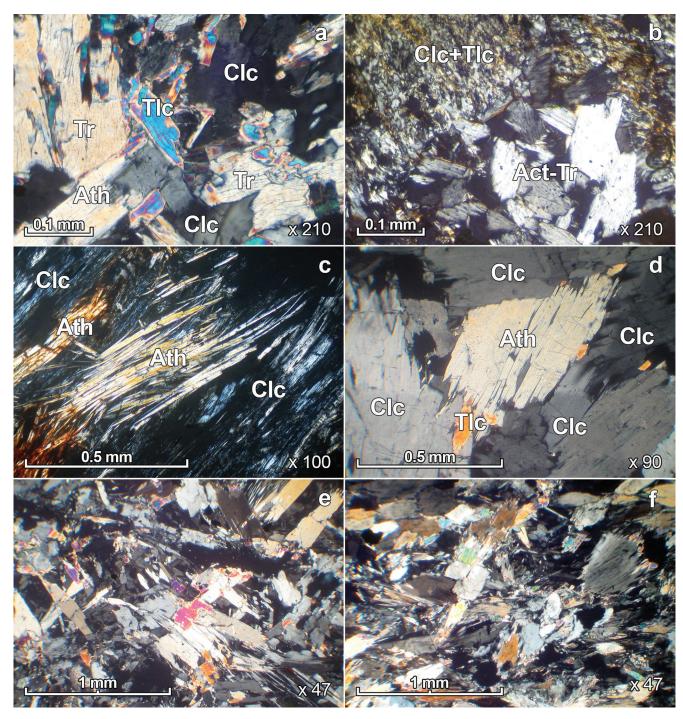


Figure 4. Thin sections of studied artefacts (transmitted light, nicols +): a – A-14030 (Group 1); b – HB-41178 (Group 2); c – A-14027 (Group 3); d – A-14031 (Group 4); e – A-14025 (Group 1); f – 6/3 (Bazavluk River, outcrop). Ath – anthophyllite; Clc – clinochlore (chlorite); Tlc – talc; Act – actinolite; Tr – tremolite.

2; Figure 5). The specimens were identified as anthophyllite-chlorite, talc-containing schists. The mineral composition of rocks is as follows: chlorite, anthophyllite, talc, ore mineral (magnetite), and goethite (Figure 4c). Most of the rock volume (over 70 vol.%) is composed of chlorite, the samples differing in their volume content of talc, which is higher in the A-14027 sample; however, in both rocks it accounts for a few per cent. The texture of both samples is glomeronematolepidoblastic.

Chlorite is represented by scaly aggregates oriented in one direction. Anthophyllite forms bundle-like aggregates of needle-like crystals oriented parallel to the schistosity. Crystals are transparent under parallel nicols, on edges they are replaced by goethite, having parallel extinction. Talc forms scales and scaly aggregates, colourless in transmitted light, which have high birefringence and excellent cleavage. It develops as a secondary mineral that alter the amphibole



Table 2. Chemical composition of the Group 3 samples obtained by XRF.

Element	A-14027	A-10428
SiO ₂	$50.504 \pm 0.095\%$	$47.894 \pm 0.095\%$
MgO	$26.316 \pm 0.133\%$	$26.060 \pm 0.132\%$
Al_2O_3	$12.697 \pm 0.064\%$	$15.954 \pm 0.070\%$
Fe_2O_3	$6.593 \pm 0.013\%$	$6.092 \pm 0.012\%$
CaO	$1.618 \pm 0.010\%$	$2.537 \pm 0.011\%$
TiO_2	$0.628 \pm 0.004\%$	$0.482 \pm 0.004\%$
K_2O	$1.024 \pm 0.017\%$	$0.358 \pm 0.019\%$
Cr_2O_3	$0.205 \pm 0.002\%$	$0.259 \pm 0.002\%$
P_2O_5	$0.054 \pm 0.005\%$	$0.144 \pm 0.006\%$
V_2O_5	$0.040 \pm 0.003\%$	$0.065 \pm 0.003\%$
BaO	$0.025 \pm 0.008\%$	$0.055 \pm 0.008\%$
S	$0.021 \pm 0.001\%$	$0.048 \pm 0.001\%$
MnO	$0.056 \pm 0.001\%$	$0.033 \pm 0.001\%$
NiO	$0.017 \pm 0.001\%$	$0.020 \pm 0.001\%$

crystals. The ore mineral is represented by opaque grains of an elongated shape, which form inclusions in the bulk of the rock, composed of chlorite, and are oriented according to the schistosity.

Group 4 of the studied collection is represented by the only sample A-14031, which differs in its petrographic properties

from the other rocks. According to the XRD analysis of sample A-14031, there were three phases established: chloriteclinochlore, talc and anthophyllite (Figure 3e). As a result of petrographic research, the specimen was identified as a talc-anthophyllite-chlorite rock with tremolite (Figure 4d). The chlorite is present in the form of pseudomorphs, which by their shape resemble tabular grains of primary minerals. Amphiboles are predominantly represented by anthophyllite, most likely developed in primary pyroxenes or hornblende and usually having a tabular shape. Sometimes the needle-like crystals are also present, which, in thin section, are predominantly represented by cross-sections with the amphibole cleavage. Tremolite differs from the anthophyllite by its inclined extinction. A small number of scaly talc crystals alter the amphibole. The rock retained the palimpsestic texture of ultrabasite, in the great bulk of which consisted of tabular grains.

5. Discussion

5.1 Provenance of the raw materials

In order to establish the provenance of raw materials of all four groups of moulds, we analysed the material of geological surveys conducted in the area of the excavations: A. G. Vinogorodskiy (1960), A. A. Zaitsev (1968), V. F. Kiktenko (1968) and V. V. Sukach (2006); as well as data

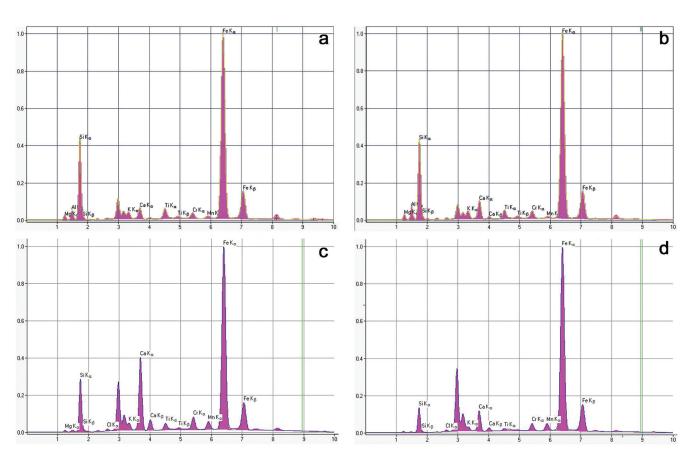


Figure 5. XRF patterns of the samples: a – A-14027 (Group 3); b – A-10428 (Group 3); c – A-14025 (Group 1); d – 6/3 (Bazavluk River, outcrop).



from related literature (Bairakov, 1975; Lazarenko *et. al.*, 1977; Lazarenko *et. al.*, 1981; Sukach, 2015; Usenko, 1953; Usenko, 1975). Based on their findings, a field study of outcrops was organised in the area of the excavations to find rocks similar to the raw materials of the studied moulds. The authors also had at their disposal a collection of thin sections of rock samples from Kryvyi Rih and the Azov Sea areas, as well as "talc schist" implements from previous studies (Kryvyi Rih, Donbas, Dnipro Rapids area, Kyiv Region).

The raw materials of all the studied casting moulds are represented by meta-ultrabasites, which are quite common in the territory of the Serednioprydniprovskyi (Middle Dnipro), Pryazovskyi (Azov) and Rosynsko-Tikytskyi cratons of the Ukrainian Shield. The area of the excavations belongs to the territory of the Serednioprydniprovskyi craton, which is a typical granite-greenstone structure. Meta-ultrabasites are found as bodies in greenstone structures, in the form of vein formations that intersect Archean rocks, including pre-greenstone granitoids, as well as among the rocks of the Skeliuvatska suite of the Kryvorizka iron ore series of the Proterozoic (Yesypchuk *et. al.*, 2004; Shcherbakov, 2005).

Similar to the Serednioprydniprovskyi craton is the structure of the western part of the Pryazovskyi craton of the Ukrainian Shield that also contains greenstone structures of Archaean age with meta-ultrabasites. Tremolite-chlorite-talc schists of the Rosynsko-Tikytskyi craton are found among the serpentinites in the area of Skvyra in the Kyiv Region (Usenko, 1975; Shcherbakov, 2005).

In the Middle Dnipro area, meta-ultrabasites are found on the rivers Bazavluk, Bazavluchok (right tributary of the river Bazavluk), Mokra Sura, Chortomlyk, Mokra Moskovka (on the territory of Zaporizhzhia City), Saksahan and Inhulets (Kryvyi Rih), as well as the Dnipro River. In the area of the archaeological monument Tokivske-1, in the south-west of the Dnipropetrovsk Region, meta-ultrabasites are exposed along the rivers Bazavluk, Bazavluchok, Chortomlyk, Inhulets and Saksahan. Their outcrops are well described in the material of the initial geological surveys and scientific works of the middle of the twentieth century, which were carried out before the building of many mining enterprises and reservoirs (Usenko, 1953).

We conducted field observations of most of the mentioned outcrops that remained intact. The exposures of similar rocks on the Chortomlyk river have not lasted to date. The varieties detected on the Bazavluchok River turned out to be actinolitites with different petrographic features. The meta-ultrabasites of the Kryvyi Rih area (Inhulets and Saksahan rivers) that crop out today are predominantly represented by essentially talc varieties that also differ from the samples under study.

Quite similar in mineral composition and petrographic features to the raw materials of the casting matrices of the first group are the rocks that crop out on the river Bazavluk in the village of Sholokhove, in the gully that flows into the river on the right side, 1 km downstream from the dam of the Sholokhove reservoir. The outcrop is located 3.5 km to the northeast from the monument Tokivske-1. The metaultrabasites occur in the form of a dyke with a sublatitudinal strike that crops out in the left board of the gully and can be traced from the mouth to its middle stream (Figure 6). The rock composition includes chlorite, amphibole and talc (Figure 4f). The crystals of amphibole have an elongated shape, are inclined and of parallel extinction. Chlorite is represented by scaly aggregates; it was formed as a result of amphibole crystal alteration, repeating the morphology of their crystals. Talc is present in the form of scales with parallel extinction and a high rate of birefringence. The content of

Figure 6. Outcrop of meta-ultrabasites in the valley of the Bazavluk River.





Table 3. Chemical composition of samples 6/3 (Bazavluk River, outcrop) and A-14025 (Group 1) obtained by XRF.

Element	Sample 6/3	A-14025
SiO ₂	$46,463 \pm 0,108\%$	$44{,}508 \pm 0{,}094\%$
Fe_2O_3	$21{,}776 \pm 0{,}045\%$	$15{,}324 \pm 0{,}030\%$
MgO	$12{,}982 \pm 0{,}295\%$	$12{,}166 \pm 0{,}245\%$
CaO	$11,\!841 \pm 0,\!035\%$	$17{,}769 \pm 0{,}041\%$
Al_2O_3	$2,\!284 \pm 0,\!076\%$	$3,\!974 \pm 0,\!070\%$
K_2O	$1{,}993 \pm 0{,}036\%$	$2,\!960 \pm 0,\!033\%$
MnO	$0,\!642 \pm 0,\!005\%$	$1,\!001 \pm 0,\!007\%$
Cr_2O_3	$0,\!551 \pm 0,\!006\%$	$0.205 \pm 0.002\%$
Cl	$0,\!545 \pm 0,\!013\%$	$0{,}542 \pm 0{,}012\%$
TiO_2	$0,\!340 \pm 0,\!009\%$	$0.813 \pm 0.009\%$
BaO	$0,\!215 \pm 0,\!016\%$	$0,\!215 \pm 0,\!019\%$
V_2O_5	$0,\!162 \pm 0,\!009\%$	$0,\!121 \pm 0,\!009\%$
NiO	$0,\!144 \pm 0,\!005\%$	$0,\!094 \pm 0,\!005\%$
ZnO	$0,\!045 \pm 0,\!007\%$	$0,\!003 \pm 0,\!006\%$
S	$0,\!015 \pm 0,\!002\%$	< 0,002%
Co ₃ O ₄	$0,003 \pm 0,013\%$	$0,013 \pm 0,010\%$

chlorite in various samples was from 50 to 75 %, amphibole from 15 to 35 %, and talc from 3 to 10 %. According to the results of the XRD analysis of two samples from the exposure, four phases were detected: chlorite-clinochlore, talc, anthophyllite and actinolite (Figure 3f). The latter, based on the lack of colour in parallel nicols, belongs to the tremolite. Thus, the studied rock contains two amphiboles: anthophyllite and tremolite.

Among the meta-ultrabasites of this occurrence, a sample was found that is very similar by its petrographic features to the raw materials of the matrices of the first group (Figure 4e-f). The comparison of the specimens was carried out using the XRF analysis (Figure 5c-d; Table 3). The rocks have a very similar composition and only minor differences in their content of Ca and Fe, which can be explained by varying degrees of weathering.

According to the survey data of A. G. Vinogorodskiy, who performed geological research in the area of the archaeological excavations (sheets L-36-8 B and G, L-36-20-A, B, C and D), the veins of meta-ultrabasites on the river Bazavluk have a regular zonal structure. Mainly amphibole rocks are developed in the central part, being replaced by chlorite-amphibole rocks along the periphery, and a layer of monomineral chlorite rock is present at the joint of dykes with the local rocks. It is more than likely that the ancient miners used rocks occurring closer to the contact chloritised zone, which were easier to process. Thus, the proximity of the location of the occurrence, the similarity of the petrographic features, as well as the XRD and XRF analyses suggest that the raw materials of the matrices of the first group may have originated from here.

It is possible that the raw materials of the casting moulds of Group 2, which differ in the absence of anthophyllite and fine-grained structure, also originate from this area. These tremolite-talc-chlorite schists can originate from any area of distribution of meta-ultrabasites, including the occurrence in the village of Sholokhove near Tokivske-1, where anthophylliteless meta-ultrabasites were also described in the survey reports. Sample A-14031 (Group 4) is a chloritised amphibole rock with a primary granoblastic texture, which may well be of local origin, and also being taken from the edge of the dyke.

Regarding the provenance of raw materials of Group 3, it should be noted that we have not found analogues for these rocks in the Middle Dnipro area either during field research or in the literature or materials of the primary geological survey. Anthophyllite meta-ultrabasites are typical for the Surska greenstone structure of the Serednioprydniprovskyi craton (Bairakov, 1975), but they have a significant depth of bedding. According to the survey materials of A. A. Zaitsev (1968), only talc-chlorite and talc-chlorite-actinolite varieties are exposed at the surface in the basin of the river Morka Sura. In the Kryvyi Rih area, where anthophyllite metaultrabasites occur, anthophyllite is a very rare mineral and it is associated with hornblende, pyroxene, biotite, cummingtonite, plagioclase, epidote, garnet, and on rare occasions cordierite (Lazarenko et. al., 1977). Though significantly anthophyllite meta-ultrabasites sometimes occur in the Middle Dnipro area, an example of which is the dyke found in the Peredatochne granite quarry in Zaporizhzhia, recorded in the geological report of V. F. Kiktenko (1968), we have to mention that anthophyllite meta-ultrabasites are more common in the Azov Sea area, where they are exposed on the rivers of Obitochna, Berda and others (Kravchenko and Rusakov, 2017; Lazarenko et. al., 1981; Usenko, 1953; Usenko, 1975).

Thus, we can state that the raw materials of the casting moulds of Group 3 may have originated from the territory of the Middle Dnipro area only if a body of the anthophyllite meta-ultrabasite ever occurred on this territory and was completely worked out by ancient miners. In another case, the Azov Sea area is the most possible for the provenance of the material of these casting moulds, as well as the raw material of casting moulds from the first hoard found in Tokivske-1 that were made from the same rock (Nikitenko *et. al.*, 2020). The similarity of the raw materials of the first complex matrices and the casting moulds of Group 3 of the second complex testifies in favour of their synchronous use, and hence the importance of Tokivske-1 as an industrial centre.

5.2 Mining localities and manufacturing of casting moulds

The results of the study of both complexes of casting moulds and preformed blocks provide an opportunity to reconsider the established ideas about the mining of "talc schists" for the manufacture of casting moulds during the Late Bronze Age in the Northern Black Sea region. As noted above, V. F. Petrougne concluded that the raw materials of casting moulds originate exclusively from the territory of the Kryvyi Rih Basin. The researcher explained this by the weak exposure of meta-ultrabasites elsewhere, the presence of quartz in the raw materials of some forms (talc schists



of the Skeliuvatska suite of the Kryvorizka series occur among meta-sandstones and meta-gravelites), as well as the insufficient study of other structures (Petrougne, 1967). In subsequent years, the study of other greenstone structures intensified, which gave grounds to I. M. Sharafutdinova, who actively collaborated with geologists, to suggest that "talc schists" could also be mined in other places of the Middle Dnipro area, particularly in the Area of Dnipro Rapids (Sharafutdinova, 1985). According to the results obtained during the study of the hoards from Tokivske-1, it is possible to expand the area of former raw materials mining for casting moulds production.

As a result of the study of the second complex of casting moulds from Tokivske-1, we have identified a possible place of extraction for the raw material of most of the matrices, one which was close to the site. Further, several moulds from both hoards were made of anthophyllite-chlorite rocks, which are not typical for the Kryvyi Rih Basin. Moreover, they may have originated from the territory outside the Middle Dnipro area, namely the Azov Sea area. All the above proves that, in addition to the Kryvyi Rih area, there were yet other mining centres for the raw materials for casting moulds. In the Middle Dnipro area, in addition to the studied outcrops along the Bazavluk River, meta-ultrabasites of the Chortomlyk and Sura greenstone structures, as well as dyke bodies of meta-ultrabasites along the Dnipro and Bazavluchok Rivers, could well have been exploited. In addition, anthophyllite species of rocks could be extracted in the Azov Sea area. Meta-ultrabasites of these localities are currently sufficiently studied and are not much inferior in their number of occurrences and volume of reserves compared to the Kryvyi Rih Basin, which at the time of the first studies of V. F. Petrougne had been much better studied due to the development of its numerous iron ore deposits.

According to the results of the current research, we may suppose that a mining site existed on the territory of the modern Sholokhove village, which supplied Tokivske-1 with raw materials for casting moulds manufacturing. We cannot yet assert that Tokivske-1 was a stone-processing centre, which produced matrices for export. However, the evidence of its broad trade connections is the analogues of the matrices of both complexes among the materials of numerous archaeological monuments of the Middle and the Lower Dnipro areas and the Northwest Black Sea region. Similar casting moulds are also known from the monument of Pobit Kamyk in Bulgaria. Bronze tools, jewellery and weapons, similar to those cast in moulds from Tokivske-1, are recorded in large areas of Eurasia. Among them, we should mention the hoards and monuments of the Middle Dnipro area, the Carpathians, Volga-Urals region, Central Asia and other regions (Starik and Kushan, 2020).

6. Conclusion

The results of the mineralogical and petrographic study of the second complex of casting moulds and preformed blocks indicate that the materials of most of the implements found at the archaeological monument of Tokivske-1 could have been mined in the immediate vicinity of the site – in the valley of the Bazavluk River, in the area of the village of Sholokhove. It is possible that the megalithic cult site was used by ancient craftsmen, and the monument itself could be both a metalworking and, to some extent, a stone-processing centre of the Sabatynivka Culture of the Late Bronze Age.

The materials of a portion of the matrices of the studied second hoard are identical to the raw materials of the complex discovered in 2017. Both complexes contain products from anthophyllite-chlorite rock, which has never been described as a raw material of foundry matrices of the Bronze Age. All this testifies to the synchronicity of both complexes and to large volumes of metallurgical production in Tokivske-1.

The obtained data prove that the development of metaultrabasites during the period of the Sabatynivka culture was carried out not only in the Kryvyi Rih area, but also in other places of occurrence of these rocks in the Middle Dnipro area and, probably, in the Azov Sea area. The existence of a network of stone-mining works could have had a positive effect on the development of the Northern Black Sea metalworking centre of the Late Bronze Age, as well as to provide a sufficient volume of exports of casting moulds and preformed blocks to other regions. To support this proposed theory, further studies of the raw materials of casting moulds from other collections and more field observations are needed using more precise research methods.

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