# Bronze age pottery from Turopolje and Podravina region - archaeometric analysis

Kudelić, Andreja; Mileusnić, Marta; Grzunov, Adriana; Wriessnig, Karin; Ottner, Franz

Source / Izvornik: Opvscvla archaeologica, 2018, 39/40, 37 - 52

Journal article, Published version Rad u časopisu, Objavljena verzija rada (izdavačev PDF)

https://doi.org/10.17234/OA.39.3

Permanent link / Trajna poveznica: https://urn.nsk.hr/urn:nbn:hr:169:166223

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Download date / Datum preuzimanja: 2024-07-23



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> RAČUNALNI PRIJELOM / COMPUTER LAYOUT Ivanka COKOL for FF-press

PRIJEVOD NA ENGLESKI / TRANSLATION TO ENGLISH Ana ĐUKIĆ & Vinita RAMLJAK

GODIŠNJAK / ANNUAL

Izdavanje časopisa novčano podupire ODSJEK ZA ARHEOLOGIJU FILOZOFSKOGA FAKULTETA SVEUČILIŠTA U ZAGREBU Publishing of the journal financially supported by DEPARTMENT OF ARCHAEOLOGY, FACULTY OF HUMANITIES AND SOCIAL SCIENCES UNIVERSITY OF ZAGREB

Službena kratica ovoga časopisa je *Opusc.archaeol. (Zagreb)* / Official abbreviation of this journal's title is *Opusc.archaeol. (Zagreb)* URL: www.ffzg.hr/arheo/opuscula

Dostupno na / Available at Ebsco Publishing (www.ebscohost.com)

Tiskano 2018. / Printed in 2018

OPVSC. ARCHÆOL. VOL. 39/40 STR. / PAGES 1-297 ZAGREB 2018.

# OPVSCVLA ARCHÆOLOGICA

# 2018

OPVSC. ARCHÆOL. VOL. 39/40 STR. / PAGES 1-297 ZAGREB 2018.

# FILOZOFSKI FAKULTET SVEUČILIŠTA U ZAGREBU FACULTY OF HUMANITIES AND SOCIAL SCIENCES, UNIVERSITY OF ZAGREB

RADOVI ARHEOLOŠKOG ZAVODA PAPERS OF THE DEPARTMENT OF A R C H A E O L O G Y

ISSN 0473-0992

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## Andreja KUDELIĆ, Marta MILEUSNIĆ, Adriana GRZUNOV, Karin WRIESSNIG & Franz OTTNER

### BRONZE AGE POTTERY FROM TUROPOLJE AND PODRAVINA REGION – Archaeometric analysis

doi: 10.17234/OA.39.3 Izvorni znanstveni rad / Original scientific paper

UDK / UDC 543:903-033.64(497.521.6+497.525)"637" Primljeno/Received: 05.10.2015. Prihvaćeno/Accepted: 16.05.2016. Andreja Kudelić Institute of archaeology Ljudevita Gaja 32 10000 Zagreb, Croatia andreja.kudelic@iarh.hr

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of the settlements and that the grog is the most commonly used temper in the analysed samples from both regions. According to the results of performed analyses, there is an indication of a difference in technological choices, i.e. different practice related to pottery firing techniques, between two micro-regions.

Keywords: archaeometry, pottery, Bronze Age, cultural group Virovitica, Turopolje, Podravina

#### INTRODUCTION

Analysis of material culture has been outgrown the questions of typo-chronological nature long time ago. Therefore, the questions are more focused to relate the material with society and social relations.

The paper presents results of the archaeometric analyses carried out on the Bronze Age pottery, found within the remains of settlements, located in the north-western part of Croatia (Turopolje and Podravina region). The main goal of this study is to determine the availability, types and characteristics of raw materials, as well as to reconstruct part of the technological process (preparation of raw material and firing technique) of pottery production. Potsherds are analysed using optical microscopy and X-ray powder diffraction (XRPD). Detailed mineralogical analysis (XRPD) with emphasis on clay minerals was performed on samples of potential raw material using oriented films of separated clay fraction and different treatments. Comparison between the data obtained from the analyses of potential raw materials and those obtained from the analyses of the pottery suggests that local clay was used for pottery making in the Bronze Age. Results of the study showed that raw material was probably collected in the vicinity

Pottery is the most common artefact in almost all periods of the past. It gives us many information not only about the form and chronology but also about production technology and use. In its structure, pottery contain a lot of information, therefore, macroscopic study combined with the microscopic analysis of pottery are the best approach to that will result a high quality set of data. Hence, study of pottery should be based on multidisciplinary approach, combining knowledge of archaeology and that of natural sciences.

Petrographic and mineralogical analysis are two basic archaeometric methods of pottery and clay analysis (Shepard 1954: 138; Rice 1987: 375). Petrography analysis permits permits us to observe: nature and characteristics of non-plastic inclusions (mineralogical composition and size, distribution and orientation of different particles), textural and optical characteristics of the clay matrix (birefringence and colour), shape, quantity and orientation of voids (Quinn 2013). One key objective of petrography is, therefore, the determination of the origin of clays and tempers used in the pottery production (Rice 1987; Quinn 2013). Application of this method on prehistoric pottery samples is extremely useful because it can provide a greater amount of information with regard to their abundant inclusions and temper. (Quinn 2013: 7). The mineralogical composition of ceramics and clays is usually studied by X-ray powder diffraction (XRPD). This method allows the identification of the main minerals that exist in the ceramic and their quantity. It also allows to determine crystalline phases such as clay minerals that are impossible to observe under petrographic microscope (Rice 1987: 382; Albero Santacreu 2014: 20).

This paper examines Bronze Age (cultural group Virovitica) pottery from the lowland settlements in north Croatia (Turopolje and Podravina regions). In the last decade, several extensive studies on the Bronze Age pottery technology have been performed resulting in a range of new information obtained by using not only chronological and cultural approach (Michelaki 2006; Kreiter et al. 2007; Cannavo et al. 2012; Sofaer & Budden 2012). There were no such comprehensive analyses done in Croatia. Reference can be made only to work of S. Karavanić who has done small scale analysis of the Late Bronze Age pottery fragments (X-ray diffraction, petrographic-mineralogical analysis, thermal analysis) from the Kalnik-Igrišče site (Vrdoljak 1995). Virovitica group pottery examined in this paper belongs to a slight older period (Middle and beginning of the Late Bronze Age) and has not yet been subjected to this type of analysis. The cultural

group Virovitica retain a very long period of time and at a relatively broad area. Therefore, the main goal of this study is to determine whether there are indicators of different technological traditions of pottery production between regions.

Consequently, research objective is to establish: (1) which raw materials were used; (2) how raw materials were prepared and, how the pottery vessels were formed (technique), surface-treated, and fired (temperature and type of atmosphere). To answer these questions, mineralogical and petrographic analysis (X-ray powder diffraction - XRPD and optical microscopy) were performed.<sup>1</sup> Mineralogical analysis is carried out on clay samples of potential raw material that was collected in the vicinity of the location of Bronze Age settlements and on pottery samples from these settlements. Thin section petrography is performed on pottery samples in order to study the clayey material (matrix), natural inclusions and temper that compose the ceramic body.

#### MATERIALS AND METHODS

#### POTTERY

Pottery was found within settlement remains which were documented and collected during the archaeological excavation at Kurilovec-Belinščica (Kudelić 2016) and Selnica Ščitarjevska sites in region of Turopolje (Sava valley), as well as during field survey of Podvratnec, Vratnec, Podgorica, Podpanje, Močvar and Jablanec sites in the region of Podravina (Fig. 1). It is part of a material remains of cultural group Virovitica that is by relative chronology placed at the end of the Middle and the beginning of Late Bronze Age (15 – 12 century BC).

Pottery production was abundant resulting in a huge amount of ceramic fragments found. According to macroscopic observation pottery is hand-made and of law quality (simple forms, high level porosity, low fired) which resulted in a state of poor preservation. After statistical evaluation (14245 pottery shard) and typological classification (21 vessel type), 182 samples of pottery were picked out for further selection using image analysis by a digital microscope Dino-Lite Pro (magnification of 20 – 60). Therefore,

<sup>&</sup>lt;sup>4</sup> Archaeometric analyses were performed as part of a Croatian – Austrian bilateral project: Pottery production in prehistoric cultures, especially Hallstatt culture, of Croatian and Austrian Danube regions (Institutions involved: Faculty of Mining, Geology and Petroleum Engineering, Faculty of Humanities and Social Sciences, Institute of Archaeology in Zagreb, and University of Natural Resources and Life Sciences in Vienna).



Figure 1. Location of the Bronze Age sites: Kurilovec-Belinščica (1), Selnica Ščitarjevska (2), Podvratnec, Vratnec, Podgorica, Podpanje, Močvar and Jablanec (3-8); regions: Turopolje (B), Podravina (C); broadly framed area of the cultural group Virovitica (A) (by: A. Kudelić).

samples were divided into three groups based on pottery texture: fine, semi-fine and coarse. Pottery textures were determined based on the type, size and frequency of the temper using image analysis of pottery cross-section. It was established that 99% of the analysed pottery samples contain grog grains. According to image analysis six main groups of pottery texture were established, and the main difference between them was the amount and size of grog grains. When it was possible, potsherds were selected based on the type of the vessel. Due to the high fragmentation of pottery, it was not always possible to single out a specific type of vessel for analysis. Selection of samples for mineralogical/ petrographic analyses was based on similar criteria: (1) the texture of pottery; and (2) vessel type. Differences in the pottery texture may indicate a diversity or consistency in the selection of technology while characteristic of clay paste used for making certain vessel type may indicate a connection between technology and function of the particular vessel.

Based on the macro analysis pottery of cultural group Virovitica was hand-made using several

techniques: modelling from a lump of clay by drawing and building up from coils or slabs. On just several fragments in the rim and the base zone of the vessel, horizontal traces of using a rotating device in the final stage of forming were recorded. In the final processing of the vessels, surface was smoothed (over 60% of the total pottery number) but rarely burnished (1-4%). Traces of using a slip have been recorded also. Outer and inner walls are mainly brown, brown-yellow and brown gray in colour while the core of pottery is almost always dark gray (over 95%).

Altogether 49 samples of pottery from Turopolje (32 samples) and Podravina (17 samples) region were selected and analysed. Pottery samples from Turopolje include: 23 samples of potsherds from Kurilovec-Belinščica site, as well as 9 samples of potsherds from Selnica Ščitarjevska site (tab. 1). Pottery samples from Podravina region include 17 samples from several sites: Podgorica (7), Podvratnec and Vratnec I (6), Podpanje (1), Močvar (2) and Jablanec (1) (tab. 2).

Site	Sample	Lab. number	Ceramic texture	Type of vessel	Remarks	XRD	ОМ
	A-1	14128	coarse	pot		+	+
	A-3	14130	fine	bowl	vessel type 5, 17 - small bowl with wart	+	-
	A-4	14131	semi-fine	amphora		+	+
	A-5	14132	semi-fine	pot		+	+
	A-6	14133	fine	-	thin walls and traces of red pigment	+	+
	A-7	14134	semi-fine / fine	small jug	extremely porous	?	-
	A-8	14135	semi-fine / fine	-	complete oxidation atmosphere	+	+
	A-9	14136	semi-fine	-	white slip	+	+
	A-10	14137	semi-fine	-	T rim pot	++	-
cica	A-11	14138	semi-fine /fine	-	undefined vessel type – T rim bowl	+	-
inšč	A-12	14139	coarse	ceramic tripod		+	+
Bel	A-13	14140	fine	bowl	from the kiln pit	+	-
vec-	A-14	14141	fine	cup	vessel type 18 - from the kiln pit	+	-
rilo	A-15	14142	semi-fine	bowl	from the kiln pit	+	+
Ku	A-17	14144	fine	Bowl	vessel type 5, 17 - small bowl with wart	+	+
	A-18	14145	semi-fine	Bowl	vessel type 19	+	+
	A-19	14146	fine	Bowl	vessel type 5, 17 - small bowl with wart	+	+
	A-20	14147	fine	Bowl	vessel type 5, 17 - small bowl with wart	+	+
	A-21	14148	semi-fine	Bowl	vessel type 19	+	+
	A-22	14149	semi-fine	bowl	vessel type 19	+	+
	A-23	14150	semi-fine	bowl	vessel type 19	+	+
	A-24	14151	semi-fine	pot	big fragmet of pot	+	+
	A-25	14152	semi-fine /coarse	pot		+	+
	A-26	14223	semi-fine	-	litzen pottery	+	-
	B-1	14153	fine/semi-fine	-	traces of red pigment	+	+
8	B-2	14154	coarse	-	litzen pottery	+	+
vsk	B-3	14155	semi-fine / fine	-		+	+
arje	B-4	14156	semi-fine	pot		+	-
Ščit	B-5	14157	semi-fine	pot		+	+
ica	B-6	14158	semi-fine	pot		+	+
eln	B-7	14159	fine/semi-fine	-		+	+
S	B-8	14160	semi-fine	-		+	+
	B-9	14161	coarse/semi-fine	pot		+	+

*Table 1. Pottery samples from Turopolje selected for archaeometric analyses (XRD – X-ray diffraction analysis; OM – optical micros-copy).* 

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Site	Sample	Lab. number	Ceramic texture	Type of vessel	Remarks	XRD	ОМ
Jablanec	C-16	14177	fine/semi-fine	bowl	vessel type 14 - bowl with inverted rim	+	+
	C-1	14162	semi-fine	jug		+	+
	C-2	14163	coarse	portable furnace		+	+
	C-3	14164	semi-fine			+	+
Podgorica	C-4	14165	semi-fine			+	+
	C-5	14166	fine	bowl		+	+
	C-9	14170	fine	bowl		+	+
	C-10	14171	coarse	pot		+	+
-	C-6	14167	semi-fine	pot		+	+
	C-7	14168	fine/semi-fine	pot		+	+
Podyratnec	C-11	14172	semi-fine / fine	bowl	vessel type 19	+	+
Touviatilee	C-12	14173	semi-fine	bowl	vessel type 19	+	+
	C-13	14174	fine	bowl	vessel type 5, 17 - small bowl with wart	+	+
Vratnec I	C-14	14175	fine/semi-fine	bowl	vessel type 5, 17 - small bowl with wart	+	+
Podpanje	C-15	14176	semi-fine	bowl	vessel type 5, 17 - small bowl with wart	+	+
Mažaan	C-17	14178	semi-fine	bowl	vessel type 14 - bowl with inverted rim	+	+
wocvar	C-8	14169	fine	bowl	vessel type 14 - bowl with inverted rim	+	+

Table 2. Pottery samples from Podravina selected for archaeometric analyses (XRD – X-ray diffraction analysis; OM – optical microscopy).

Region	Sample number	Lab. number	Number of samples	Coordinates	Distance from the archaeological site	Location of the sample	Colour
Turopolje	М	14124	1	45.657977 16.084164	4 km	Brickyard Mraclin	brown yellow
Turopolje	В	14125	1	45.657977 16.084164	4 km	Brickyard Mraclin	very light gray
Turopolje	G	14126	1	45.700522 16.063316	0,6 km	near settlement remains	gray brown
Turopolje	К	14127	1	45.690340 16.071262	0,5 km	near settlement remains	gray brown
Podravina	J	14866 14867	2	46.247667 16.802674	30-70 m	near settlement remains	gray brown
Podravina	PG	14870 14871	2	46.181544 16.892333	30-70 m	near settlement remains	gray with yellow spots
Podravina	РР	14872 14873	2	46.210707 16.884622	30-70 m	near settlement remains	gray brown
Podravina	PV	14868 14869	2	46.221772 16.848861	30-70 m	near settlement remains	gray brown

Table 3. Clay samples from Turopolje and Podravina (M and B - clays from brickyard Mraclin; G - clays north from the Kurilovec-Belinščica site; K - clays south from the Kurilovec-Belinščica site; J-Jablanec site; PG-Podgorica site; PP-Podpanje site; PV-Podvratnec site).

#### CLAYS

The term "clay" is applied both to materials having a particle size of less than 2 micrometers and to the family of minerals that has similar chemical compositions and common crystal structural characteristics (Velde 1995). While clay minerals have a wide range of particle sizes (from tens of angstroms to millimetres), clays may be composed of mixtures of finer grained clay minerals and clay-sized crystals of other minerals such as quartz, carbonates, oxides or non-clay silicates. In this paper term "clay" is used for fine-grained natural rock or soil material that: (1) contains dominantly one or more clay minerals, other minerals in smaller amount and organic matter; (2) is plastic due to their water content; and (3) become hard, brittle and non-plastic upon drying or firing (Gugenheim & Martin 1995).

Altogether 12 samples of clays from Turopolje and Podravina region were selected and analysed (tab. 3). Possible raw material is represented by 4 samples of clayey material collected from 3 locations in the vicinity of the archaeological site Kurilovec-Belinščica. Two samples were collected in brickyard Mraclin, which is located 4 km from the site. Other two samples were collected close to the site (600 m to the northeast and 500 m to the south, respectively). Eight samples of clayey material were collected in Podravina at 4 locations next to the remains of the prehistoric settlements (within a radius of approximately 50 meters around the selected settlements).

#### ANALYTICAL METHODS

Petrographic analysis of Bronze Age potsherds was performed on thin sections using polarised transmitted light microscope Leica (type DMLSP) at the Department of Mineralogy, Petrology and Mineral deposits at Faculty of Mining, Geology and Petroleum Engineering (University of Zagreb). Photomicrographs were taken using microscope Zeiss Axio Imager.A2m, camera AxioCam Icc 5 and software Zen (Blue Edition), Carl Zeiss Microscopy GmbH, 2011.<sup>2</sup> Petrographic analysis includes determination of samples' colour; texture; matrix birefringence; mineral composition; presence of natural and anthropogenic inclusion, voids and pores; microstructure; and microtexture. For two potsherds it was not possible to prepare thin sections because of highlevel fragility and porous texture of the sample.

Mineral composition of pottery and clays from the surrounding of archaeological sites was determined by X-ray powder diffraction analysis (XRPD) using XPERT-PRO PANalytical (goniometer: PW3050/60, Theta/Theta, Cu/Kα radiation, automatic divergent slit). Prior to analysis samples were dried and ground to powder in agate mill. This technique gives the results of the bulk mineral composition of a sample. Unlike petrographic analysis, it cannot give other important parameters of the sample (eg. structure, texture, presence and type of rock particles, changes on mineral grains...). However XRPD the advantage of this analysis is unambiguous and direct determination of certain clay minerals, which cannot be determined by other physical methods, especially in the case of poly-phase mixtures.

In addition, detailed clay mineral analysis is performed on the < 2  $\mu$ m fraction of the possible raw material samples. The XRD patterns of oriented <2 $\mu$ m fraction were taken after the following treatments: (a) Mg-saturation, (b) K-saturation, (c) Mgsaturation and ethylene glycol solvation, (d) Mgsaturation and ethylene glycol solvation, (e) K-saturation and DMSO solvation, and (f) heating for two hours at 550°C. The identification of clay minerals was generally based on the methods outlined by Brown (1961), Brindley & Brown (1980), and Moore & Reynolds (1989).

#### RESULTS

#### POTTERY

Mineral composition of pottery samples from Turopolje determined by XRPD is presented in tab. 4. The dominant mineral phase is quartz. Plagioclase, K-feldspar and mica are present in minor amount in almost all samples. Mineral phases such as hematite and calcite are determined in some samples. While hematite is determined in some of Kurilovec-Beliniščica as well as Selnica Ščitarjevska samples, calcite is present only in some of Selnica Ščitarjevska samples. Some samples contain 14Å and 12Å mineral. Optical microscopy helped to distinguish micas (muscovite and/or biotite). Mineral phases identified only by optical microscopy are pyroxenes (ortopyroxene) and zircon.

Along with crystaloclast, lithoclascts and grog (ceramoclasts) are also present (tab. 5). Lithoclasts are

<sup>&</sup>lt;sup>2</sup> Microphotographs are taken at Babes-Bolyai University, Cluj-Napoca, Romania during CEEPUS exchange. We thank Prof. Corina Ionescu for invitation of Associate Professor Marta Mileusnić and given opportunity to use her equipment.

Site	Lab. number	Qtz	Pl	Kfs	Мса	Hem	Cal	~14Å	~12Å
	14128	+	*	*	+	-	-	+	?
	14130	+	*	*	+	?	-	+	+
	14131	+	?	+	+	+	-	-	-
	14132	+	*	?	+	?	-	-	-
	14133	+	*	*	+	?	-	?	?
	14134	+	*	*	+	-	-	+	+
	14135	+	?	?	+	-	-	-	-
	14136	+	?	*	+	-	-	-	-
	14137	+	*	*	+	-	-	?	+
ica	14138	+	*	*	+	-	-	+	?
inšč	14139	+	*	*	+	?	-	-	-
- Bel	14140	+	*	*	+	?	-	?	+
vec -	14141	+	*	*	+	-	-	-	?
urilo.	14142	+	*	*	+	-	-	+	?
Ku	14144	+	?	*	+	-	-	?	?
	14145	+	*	*	+	?	-	-	-
	14146	+	*	*	+	-	-	+	+
	14147	+	*	*	+	-	-	+	+
	14148	+	+	*	+	-	-	+	?
	14149	+	?	*	*	+	-	-	-
	14150	+	*	*	+	-	-	+	+
	14151	+	?	*	+	-	-	-	-
	14152	+	*	*	+	?	-	-	?
	14223	+	+	*	+	-	-	+	+
	14153	+	*	*	*	*	+	?	+
	14154	+	+	*	+	-	-	+	+
ska	14155	+	+	*	+	-	-	+	+
arjev	14156	+	*	+	*	-	*	-	-
Ščita	14157	+	*	+	*	-	+	-	?
nica	14158	+	?	*	*	-	?	-	*
Sel	14159	+	*	*	*	-	-	-	+
	14160	+	*	*	*	-	-	-	*
	14161	+	*	*	*	+	+	-	*

Table 4. The qualitative mineral content of pottery samples from Turopolje (Qtz - quartz; Pl - plagioclase; Kfs – K-feldspar; Mca – micaceous material; Hem – hematite; Cal - calcite;-14Å - mineral with diffraction maximum at 14 angstrem;-12Å - mineral with diffraction maximum at 12 angstrem; – not identified; ? – indications of presence; \* - minor quantities; + – present)

present in most of the samples and they are represented dominantly by quartzite. Chert is also often present. Sandstone particles are present in some samples as well. Some lithoclast are relatively large in size. Grog is present in various quantities in all analysed samples. Grog grains are angular to sub rounded, but majority of grog grains are sub angular with sizes between 0.2 and 4 mm. Clay pellets are present in many samples as well. In all analysed samples pores are clearly visible. They are mostly elongated and parallel to the vessel walls. There are also visible pores that do not show preferred orientation, as well as rounded pores. Matrix is in the broad range from amorphous to microcrystalline. All but one sample from Selnica Ščitarjevska have microcrystalline matrix. Birefringence of the matrix is in the range from low to high. Middle to high birefringence prevail in Turopolje samples. eral is probably chlorite (probably of primary origin - from clays). ~12Å mineral is probably secondary phase (formed by rehydroxylation or incorporated in the sample during its burial) Sample 14171 contain significant amount of spinel. Optical microscopy helped to distinguish micas. Most of the samples contain muscovite and/or biotite. Mineral phases identified only by optical microscopy are pyroxenes (ortopyroxene) and zircon.

Mineral composition of pottery samples from Podravina determined by XRPD is similar (tab. 6).

Site	Lab. number	Matrix	Bire fringence	Cristalo- clasts	Litho- clasts	Grog	Pores
	14128	M-A	HB	Qtz;Kfs;Ms;Bt	Cht	+	EP
	14131	A	LB	Qtz;Kfs;Pl;Bt;Px;Zr	Qtzt;Cht;Sst	+	RNO
	14132	A	MB-LB	Qtz;Kfs;Pl;Ms;Px	Qtzt	+	EP; RNO
	14133	A-M	HB	Qtz;Ms	-	+	EP; RNO
	14135	А	LB	Qtz;Ms;Px	Qtzt	+	RNO
ca	14136	A-M	MB	Qtz;Ms;Px	-	+	-
nšči	14139	A-M	MB	Qtz;Kfs;Px;Zr	Qtzt;Cht	+	RNO
Selin	14142	A-V	LB	Qtz;Ms	Cht	-	EP
	14144	М	HB	Qtz;Kfs;Ms;Px;Zr	-	+	EP
Deve	14145	М	HB	Qtz;Kfs;Pl;Px	Qtzt;Sst	+	EP
Irile	14146	М	HB	Qtz;Ms,Bt;Px;Zr	Qtzt	+	EP
K	14147	M-A	HB-MB	Qtz;Pl;Px	Qtzt	+	RNO
	14148	M-A	HB	Qtz;Kfs;Ms;Bt;Px	Qtzt	+	EP
	14149	A-V	LB	Qtz;Ms;Px	Qtzt	+	RNO
	14150	M-A	MB	Qtz;Px	Cht	+	EP
	14151	М	LB	Qtz;Px	Qtzt	+	EP
	14152	М	HB	Qtz;Px	Qtzt	+	PO
	14153	М	MB	Qtz;Kfs; Px; Zr	Qtzt;Cht	+	RNO
ika	14154	М	HB	Qtz;Kfs;Pl;Ms;Px; Zr	Qtz;Cht;Sst;Sch	+	EP
jevs	14155	М	HB	Qtz;Ms;Px	-	+	EP
itar	14157	М	MB	Qtz;Ms;Bt;Px;Zr	-	+	EP; RNO
a Šč	14158	М	MB	Qtz;Kfs;Ms;Bt;Px;Zr	Qtzt;Sst	+	EP; RNO
nica	14159	M-A	MB-LB	Qtz;Kfs,Ms,Px	Qtzt	+	EP; RNO
Sel	14160	М	HB	Qtz;Kfs;Pl;Ms;Bt; Px; Zr	Qtzt;Cht;Sst	+	EP; RNO
	14161	М	MB-HB	Qtz;Kfs;Px	Qtzt;Cht	+	EP; RNO

Table 5. The results of the optical microscopy of the pottery samples from Turopolje region (<u>Matrix:</u> M – microcrystalline; A – amorphous; V - vitreous; <u>Birefringence</u>: LB - low; MB – middle; HB – high; <u>Crystaloclasts</u>: Qtz – quartz; Kfs – K-feldspar; Ms – muscovite; Bt – biotite; Pl – plagioclase; Px – pyroxene; Zr –zircon; <u>Lithoclasts</u>: Cht – chert; Qtzt – quartzite; Sch – schist; Sst - sandstone; Pores: EP – elongated and parallel; RNO – round and not oriented; PO – partially oriented)

Only sample 14171 stands out. The dominant mineral phase is quartz. Plagioclase, K-feldspar and mica are present in minor amount in all samples. Mineral phases such as hematite, calcite and dolomite are determined in some samples. Some samples contain ~14Å mineral and/or ~12Å. ~14Å min-

Along with crystaloclast, lithoclascts and grog (ceramoclasts) are also present (tab. 7). Lithoclasts are present in most of the samples and they are represented mostly by quartzite and chert. Sandstone particles are present in only two samples. Grog is present in various quantities in all analysed sam-

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Site	Lab. number	Qtz	Pl	Kfs	Мса	Hem	Cal	Dol	Spl	~14Å	~12Å
Podgorica	14162	+	*	*	*	-	+	+	-	-	-
Podgorica	14163	+	*	*	*	+	?	?	-	-	-
Podgorica	14164	+	*	*	+	-	-	-	-	+	+
Podgorica	14165	+	*	*	*	+	-	-	-	+	+
Podgorica	14166	+	*	*	*	+	-	-	-	-	+
Podvratnec	14167	+	*	*	+	-	-	?	-	-	-
Podvratnec	14168	+	*	*	+	-	-	?	?	?	-
Močvar	14169	+	+	*	+	+	-	-	?	+	-
Podgorica	14170	+	*	*	*	-	-	-	-	?	?
Podgorica	14171	+	?	-	-	-	-	-	+	-	-
Podvratnec	14172	+	*	*	*	-	?	?	?	-	-
Podvratnec	14173	+	*	*	+	-	-	-	?	+	*
Podvratnec	14174	+	*	*	*	-	-	-	-	-	?
Vratnec I	14175	+	*	*	+	-	-	-	-	+	?
Podpanje	14176	+	*	*	*	+	+	+	-	-	-
Jablanec	14177	+	*	*	+	+	?	?	-	+	-
Močvar	14178	+	*	*	+	+	-	-	-	+	+

Table 6. The qualitative mineral content of pottery samples from Podravina (Qtz - quartz; Pl - plagioclase; Kfs – K-feldspar; MM – micaceous material; Hem – hematite; Cal - calcite; Dol - dolomite; Spl - spinel;-14Å - mineral with diffraction maximum at 14 angstrem;-12Å - mineral with diffraction maximum at 12 angstrem; – not identified; ? – indications of presence; \* - minor quantities; + – present)

Site	Lab. number	Matrix	Bire fringence	Cristalo- clasts	Litho- Clasts	Grog	Pores
Podgorica	14162	A	LB	Qtz; Ms; Bt; Px	Qtzt; Cht	+	EP; RNO
Podgorica	14163	М	MB	Qtz; Kfs; Bt; Px; Zr	Qtzt; Cht	+	EP; RNO
Podgorica	14164	M-A	MB-LB	Qtz; Kfs; Ms; Bt; Px; Zr	Qtzt	+	EP; RNO
Podgorica	14165	M-A	MB-LB	Qtz; Kfs; Pl; Ms; Bt; Zr	Qtzt; Cht; Sst	+	EP; RNO
Podgorica	14166	M-A	MB-LB	Qtz; Kfs; Ms; Bt; Px	Qtzt; Sst	+	EP; RNO
Podvratnec	14167	M-A	LB	Qtz; Kfs; Ms; Bt; Px; Zr	-	+	RNO
Podvratnec	14168	М	MB	Qtz; Kfs; Ms; Bt; Px; Zr	-	+	RNO
Močvar	14169	М	MB	Qtz; Kfs; Ms; Bt; Px; Zr Qtzt		+	RNO
Podgorica	14170	М	MB-LB	Qtz; Kfs; Ms; Bt; Px; Zr	Qtzt	+	EP; RNO
Podgorica	14171	A	LB	Qtz; Kfs; Pl	-	+	RNO
Podvratnec	14172	М	MB	Qtz; Kfs; Ms; Bt; Px; Zr	Qtzt	+	RNO
Podvratnec	14173	M-A	MB	Qtz; Kfs; Pl; Ms; Bt; Px; Zr	Qtzt	+	EP; RNO
Podvratnec	14174	M-A	MB-LB	Qtz; Kfs; Pl; Ms; Bt; Px; Zr	Cht	+	EP; RNO
Vratnec I	14175	A	LB	Qtz; Kfs; Ms; Bt	-	+	EP; RNO
Podpanje	14176	M-A	LB	Qtz; Kfs; Pl; Ms; Px	-	+	EP; RNO
Jablanec	14177	М	MB	Qtz; Kfs; Bt; Px	Qtzt; Cht	+	RNO
Močvar	14178	M-A	LB	Qtz; Kfs; Ms; Px; Zr	Qtzt; Cht	+	EP; RNO

Table 7. The results of the optical microscopy of the pottery samples from Podravina region (<u>Matrix</u>: M – microcrystalline; A - amorphous; <u>Birefringence</u>; LB - low; MB – middle; HB – high; <u>Crystaloclasts</u>: Qtz – quartz; Kfs – K-feldspar; Ms – muscovite; Bt – biotite; Pl – plagioclase; Px – pyroxene; Zr – zircon; <u>Lithoclasts</u>: Cht – chert; Qtzt – quartzite; Sst - sandstone; <u>Pores</u>: EP – elongated and parallel; RNO – round and not oriented)

ples. Grog grains are angular to sub rounded, but majority of grog grains are sub angular with sizes between 0.2 and 3.6 mm. In all analysed samples pores are clearly visible. They are mostly elongated and parallel to the vessel walls. There are also visible pores that do not show preferred orientation, as well as rounded pores. Matrix is in the range from amorphous, microcrystalline-amorphous, to amorphous. Birefringence of the matrix is in the range from low to middle.

#### CLAYS

According to the Shepard (1954) classification most of the clay samples from both regions are defined as silty clays and clayey silts (Fig. 2). All samples contain less than 20% of sand. In general, samples from Podravina are finer than samples from Turopolje, but differences are not significant.

Mineral composition of analysed clays from Turopolje is represented by quartz, plagioclase, K-feldspar, illitic material, kaolinite and chlorite (tab. 8). Only one sample contains carbonates in the form of dolomite, while two samples contain goethite. Mineral composition of analysed clays from Podravina is represented by the same minerals with addition of vermiculite in all analysed samples (tab. 8). Samples from the site Podvratnec (14868 and 14869) contain also carbonates in the form of calcite and possibly dolomite. Samples from the site Podgorica contain



Figure 2. Texture of the sampled clayey material (after Shepard, 1954). Legend: 1 – clay; 2 – sandy clay; 3 – silty clay; 4 – clayey sand; 5 – silty sand; 6 – sand; 7 – clayey silt; 8 – sandy silt; 9 – silt; 10 – sand silt clay; red circles – Turopolje samples; green circles – Podravina samples)

goethite. Illitic material is term that represents mixture of muscovite and/or illite and/or illite-smectite mixed layered mineral. Well-rounded iron-nodules in clays from Podravina can be seen by naked eye as well.

Region	Lab. number	Qtz	Pl	Kfs	Ill	Kln	Vrm	Chl	Dol	Cal	Gt
e	14124	+	+	*	+	+	-	+	-	-	+
lpodi	14125	+	+	*	+	+	-	+	+	?	+
Ture	14126	+	+	*	+	+	-	?	-	-	-
	14127	+	+	*	+	+	-	+	-	-	-
	14866	+	+	*	+	+	+	+	-	-	-
	14867	+	+	*	+	+	+	+	-	-	-
	14868	+	+	*	+	+	+	+	?	+	-
avina	14869	+	+	*	+	+	+	+	?	+	-
odr	14870	+	+	?	+	+	+	+	-	-	+
	14871	+	+	*	+	+	+	+	-	-	+
	14872	+	+	*	+	+	+	+	-	-	-
	14873	+	+	*	+	+	+	+	-	-	-

Table 8. The qualitative mineral content of the clays from Turopolje and Podravina (Legend: Qtz - quartz; Pl - plagioclase; Kfs - K-feldspar; III - iIlitic material; KIn - kaolinite; Vrm - vermiculite; Chl - chlorite; Dol - dolomite; Cal - calcite; Gt - goethite; - not identified; ? - indications of presence; \* - minor quantities; + - present)

#### DISCUSSION

#### POSSIBLE RAW MATERIAL

The selection of the raw material in the landscape reveals a social understanding and using of parts of the landscape especially when it comes to procurement of clay for pottery making (Michelaki et al. 2014). Studied pottery is found among the remains of Bronze Age settlements which are located in lowland along the major rivers, near minor watercourses. Hence, the alluvial clays were available in significant amount and easily accessible in the vicinity of the village. Analysed clays from both regions do not contain any significant mineral which could undoubtedly approve that these clays were raw material for Bronze Age pottery production. However, considering the grain size and mineralogical content, clays from the immediate vicinity of Bronze Age settlements could be used for pottery making. The quality of clays from Podravina is lower than that from Turopolje because it contains vermiculite. Vermiculite is swelling clay mineral. It gain volume by mixing with water and shrink by losing water. Hence, it could cause breakage of the vessel during drying and firing. Some clay samples contain goethite. Large rounded grains of iron oxides can be found in Podravina clays and they are visible even with naked eyes. The pottery samples from Podravina region often contain larger grains of iron oxide (Fig. 3: d) indicating that those clays were used as a raw material. Iron oxide is also present in pottery from Turopolje region but much smaller in size. Analysed clays could be used directly after the exploitation in a wet condition due to its quality, plasticity and availability. The presence of certain lithoclast (chert, quartzite; Fig. 3: b) also suggest that the raw material for the pottery production in Turopolje was collected in the vicinity of the site (Šikić et al. 1979).



Figure 3. Microphotographs of thin sections of Bronze Age pottery (scale bar = 0.5 mm.); a) sample A-18 (Kurilovec-Belinščica site) with specified types crystalloclasts: Quartz (Qtz), K-feldspar (Kfs), Plagioclase (Pl), Orthopyroxene (Opx); lithoclast: Quartzite (Qtzt) and grog grain (G); b) sample A2 (Kurilovec-Belinščica site), chert; c) sample C6 (Podravina – Podvratnec site), distinct presence of pyroxene in the matrix; d) sample C10 (Podravina – Podgorica site), a well-rounded iron nodule of pedogenic origin in potsherd (by: M. Mileusnić, 2014).

are located. Hence, it can be presumed that grog is made from the same materials as pottery. It is very common to find older, recycled peace of grog grain which is clearly visible in the grog grain from fragments of vessels which points to a long tradition of recycling pottery (Fig. 4: b). In general, non-plastics are added to pure clays to reduce shrinkage and im-

#### PREPARATION OF CLAY PASTE

Larger quartz grains (Fig. 3: a) were recorded in some pottery samples only in small quantities, therefore it cannot be claim with certainty that potter was intentionally added quartz grains to the clay paste. On the other hand, the presence of grog in various quantities and sizes is recorded by optical microscopy in almost all samples (Fig. 4). Grog grains can be recognized by different birefringence of matrix which is lower than birefringence of matrix in the rest of the sample. Mineral composition and matrix of grog grains does not differ from the ceramic in which they

prove workability (Rye 1981: 39; Velde & Druc 1999). The prevailing opinion is that the use of grog has primary functional basis, and according to some authors for kaolinite clays a suitable proportion of added non-plastic temper is between 20 and 50 % (Rye 1981: 39). By adding a smaller amount of grog temper in clay paste resistance to thermal stress of the vessels is improved in contrast to vessels that do not contain temper (Rye 1981: 27; Rice 1987: 229; Velde & Druc 1999: 116). According to certain ethnographic examples, deliberately adding larger amounts of



grog in the clay mixture (more than 30%) enhance the physical and thermal properties of the vessel (Spindel 1989: 69; according to Kreiter 2007:117). However, advantages of the grog as temper material are primarily its availability, easy preparation and functional characteristics that ensure a vessel resistance to thermal stress and a variety of mechanical damage.

Based on macroscopic analysis of the examined pottery from NW Croatia (Kudelić 2015) grog was recorded in 99% of samples, used in various quantities and sizes and it was used for making all types of vessels. It was found that the potter added minimum 2 and maximum 40% of grog to a clay paste. However, the certain number of samples (between 5 and 15 %) contains a considerable amount of grog sometimes up to 30%. It was found that the occurrence of larger amounts of grog in the analysed potsherds cannot be connected with the vessels that were used solely for thermal processing of food. In fact, the appearance of larger amounts of grog is recorded in a relatively thin-walled vessels also that correspond to the typological forms as cups or small bowls and contain no indicators on the use of fire. On the other hand, in majority of samples (approximately 40%), the use of a small amount of grog (less than 7%) may be an indication that the potters may not have functionality in mind when they used grog temper.

Figure 4. Microphotographs of thin sections of Bronze Age pottery (scale bar = 0.5 mm.); a) sample B9 (Selnica-Ščitarjevska site), various grog grain (G) embedded in matrix; b) sample B1 (Selnica-Ščitarjevska site), recycled grog in the grog grain – bottom left; c) sample C7 (Podravina – Podvratnec site), various grog grains (G), pores and voids in the matrix; d) sample C9 (Podravina – Podgorica site), various grog grains (G), pores and voids in the matrix (by: M. Mileusnić, 2014).

According to these data the use of grog can be interpreted in a different way. Data from ethnographic sources and evidence of systematic recycling of pottery indicate strong elements of tradition in the selection of temper (Kreiter 2007).<sup>3</sup> In this regard this phenomenon could be considered from the symbolic aspects of incorporating older vessels in the new pot. That transformation presents a symbolic moment of maintaining the continuity and identity within the community (DeBoer 1974: 336; Smith 1989: 61; Chapman 2000; Brück 2006).

Pottery fragments from the studied sites are highly porous and easy to break. It is a result of low firing temperature and probably due to the addition of organic temper to a clay paste. Distinctly dark colour

<sup>&</sup>lt;sup>3</sup> As part of PhD thesis (Kudelić 2015) the social aspect of temper selection, as well as the breaking of vessels as part of the social practices within the cultural group is discussed. This practice is deeply connected with the maintenance of tradition (Chapman 2000; Chapman, Gajdarska 2006).

of the pottery fragments core is also an indication of presence of organic temper in the clay paste. The remains of organic temper are manifested in the form of voids and abundant elongated and rounded pores in cross section of the potsherd (Gibson & Woods 1990: 267). The analysed pottery samples have rounded and elongated pores and very dark grey core of the wall which is evidently a result from the combustion of organic temper. Organic inclusions are usually naturally occurring in the clays but they could be added as temper. Intentionally adding organic material (dry grass, hay, chaff, grained dung) in the clay paste has a positive effect on thermal stress, and few other functional characteristics (Rye 1981: 34; Schiffer & Skibo 1987; Skibo et al. 1989.).

#### FORMING TECHNIQUES

The shape and orientation of pores occur as a result of certain forming techniques used to shape the vessels. Elongated pores with parallel orientation can be result of a rotating device usage, while rounded pores can be result of handmade pottery. Parallel orientation can be result of both, handmade and wheel made, so definite conclusion cannot be made (Courty & Roux 1995; Velde & Druc 1999; Cuomo di Caprio 2007). The macroscopic analysis of the several potsherds in both regions showed that rotating device was used for final processing but the main forming technique was building up from coils or slabs (Kudelić 2015: 252–254)<sup>4</sup>.

#### FIRING

The vessels were usually fired under the reducing atmosphere and incomplete oxidising atmosphere, resulting in a predominantly dark grey core while the surface is dark brown, brown, or yellowbrown in colour. Birefringence is relatively high in most of the samples indicating relatively low firing temperatures. Firing temperature is determined on the basis of specific mineral phase in a specific temperature range and on the basis of colour, and changes in the micro-texture of pottery which occur during firing process. Lowest firing temperature can be estimated at 573°C because of the presence of the cracked quartz crystalloclasts. Hematite found in analysed pottery is very likely product of firing. Firing temperatures were probably higher than 600°C and lower than 850°C. This is consistent with firing temperatures usually evaluated for Bronze Age pottery (Kreiter 2007: 44). Samples with low birefringence of matrix were fired at higher temperature than those with the higher birefringence of matrix. There is a significant difference in birefringence of matrix in samples from Turopolje and samples from Podravina. Pottery from Podravina region have much lower birefringence which can be result of different temperatures of firing.

The relatively low firing temperature and selection of firing atmosphere (reducing and incomplete oxidation atmosphere) are closely linked to a mixture of clay paste for making vessels. This particularly refers to presence of organic temper in clay paste. Pastes rich in organic matter has much higher rate of fractures when they are slowly fired. Rapid firing is more appropriate for organic tempered pastes (Albero Santacreu 2014: 99). Therefore such pastes are an indicator of the use of low firing temperatures (550-650°C), short time of exposure to highest temperature and short firing cycle.

Few pottery samples from Selnica Ščitarjevska in Turopolje and from Podravina contain minor amount of calcite which was determined by XRPD. Calcite can be natural component of the clay used for pottery making (in the form of limestone particles) or it can be precipitate of calcium carbonate from soil in which pottery samples were found. As there was no indication of calcite by optical microscopy (probably because of its negligible amount), it is not possible to determine if it is of primary or secondary origin. If calcite is primary constituent of the clay, it indicates low temperature of firing. As there was no indication of calcite by optical microscopy, it is not possible to determine if it is of primary or secondary origin (for sure bellow 800°C). On the other hand, sample 14 171 from Podravina contain spinel. Spinel formation start at 800°C (Kirsch 1968). Amorphous matrix, as well as low birefringence of this samples indicate higher temperature of firing.

#### CONCLUSION

Based on the conducted analyses, it was possible to reconstruct certain activities as part of the operational sequence of pottery production process. First of all, it refers to the selection and preparation of raw material for pottery production. The raw material was presumably purchased in the vicinity

<sup>&</sup>lt;sup>4</sup> As a part of the PhD thesis (Kudelić 2015) a reconstruction of the manufacturing process of cultural group Virovitica pottery was done. Detailed analysis of the building techniques, final processing of the vessels (slip, coloring, use of rotation device) and firing techniques were conducted as well.

of settlements and alluvial clays were of sufficient quality for pottery shaping. Potter had an easily accessible material that could have been exploited from shallow pits and was accessible in a moist condition which allowed the direct use of raw material.<sup>5</sup> Frequent occurrence of large iron oxides nodules in pottery from Podravina region is linked with geological characteristics of the landscape which is direct indicator of using a local raw material for the pottery production in the Bronze Age. After selection and procurement of raw material, potter prepared clay paste for building vessels by adding crushed pottery (grog) and presumably organic matter temper (dry grass, hay, chaff, grained dung). Grog was added to 99% of the samples which was visible already by macro-analysis of pottery. Grog grains were added to all types of ceramic vessels. It can be concluded that potters from both regions used a very similar recipe for the preparation of clay paste and similar raw material. According to the results of performed analyses, there is an indication of difference in technological choices, i.e. different practice related to firing techniques, between two micro-regions. Following the basic steps in the chain of operations potter is dealing with number of technological choices that allow us to understand his choice to use particular raw material, technique or tool which is directly related to cultural choices. Pottery production within the cultural group is generally very uniform, and this conclusion leads to the assumption of a unique cultural and spatial zone. Yet certain disparities in technology (firing techniques) between different micro-regions are manifested in a form that can also be recognized on the basis of macro-analysis of pottery. Therefore, to draw conclusions about regional differences and equalities, there is a need for additional experimental, archaeometric and macro-analysis of pottery and potential raw material. The scientific studies of ceramic material are rapidly evolving. Regardless of the technology currently available, valid answers cannot be obtained using only one analytical method. The study of pottery technology is multi-disciplinary process, and only a combination of different methods (archaeometry, ethnography, experiment, etc.) can make a certain shift in research, particular those related to dynamic processes of the production, use and exchange of the products.

<sup>&</sup>lt;sup>5</sup> According to some ethnographic examples in areas where deposits of alluvial clay raw materials is available and extracted near village, potter cleaned clay on the site (removing large inclusions of gravel or leaves) and formed clay into a balls and transported it to the village (Rice 1987: 121; Albero Santacreu 2014: 67).

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