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Mineralogical and petrographic characteristic of basic types of Turonian flints from the north-eastern margin of the Holy Cross Mountains: a preliminary report

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This paper presents the preliminary results of mineralogical-petrographical investigations of several basic types of Turonian flints from the north-eastern margin of the Holy Cross (Świętokrzyskie) Mountains. To discriminate the character and degree of individual types of flints based on their mineralogic and petrographic features, microscopic analyses of series of representative samples of flint raw material from different parts of the Turonian outcrops and archaeological artefacts found at the early Neolithic site in Tominy (Opatów district) were performed. The microscopic investigations were conducted using a petrographic microscope and a scanning electron microscope (SEM–EDS). The results enable us to determine some characteristic properties of individual types of Turonian flints, and to identify the so-called Zawada flint in the Linear Pottery Culture inventory from Tominy.

KEY-WORDS: Turonian flints, Holy Cross (Świętokrzyskie) Mountains, Early Neolithic, Linear Pottery Culture, Microscopic analysis, SEM–EDS

The presence of flint-bearing cretaceous sediments within the north-eastern, Mesozoic margin of the Holy Cross (Świętokrzyskie) Mountains is limited, in greatest extent, to its eastern part, covering the fairly small area on both sides of the middle reaches of Vistula River (Fig. 1). These sediments are represented primarily by white and gray siliceous limestones and, to a lesser extent, by detrital bryozoan limestones, genetically linked with the sedimentation processes of the Lower Turonian transgression (Samsonowicz 1934a: 46–49, 1934b; Pożaryski 1948: 36–39; Złonkiewicz 1994: 22–23; 1998; Michniak and Budziszewski 1995a: 15–17). The coexisting flint outcrops occur as

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Fig. 1. Geological structure of the sub-Quaternary sediments of the north-eastern margin of the Holy Cross Mountains (1 – Jurassic; 2 – Lower Cretaceous and Cenomanian; 3 – Turonian; 4 – Coniacian-Upper Maastrichtian) with the range of occurrence of the basic types of Turonian flints with the location of analysed samples (5 – Gościeradów flint; 6 – Świeciechów flint; 7 – Janików flint; 8 – Ożarów flint; 9 – Zawada flint). Places of origin of analysed samples: a – surface concentraction of flint raw material; b – quarry; c – prehistorical flint mines; d – archaeological site 6 in Tominy (geological structure: after Pożaryski 1997, with Authors modifications; location of the outcrops of flints: after Samsonowicz 1934b; Michniak and Budziszewski 1995: Fig. 6; Libera and Zakościelna 2002: Ryc. 1).

primary deposits (within fairly numerous quarries) and as secondary surface deposits, defaulting in ceiling parts of Turonian rocks. These secondary deposits were a significant source of archaeological material during the Neolithic and Bronze Age (Balcer 1971: 118; 1976: 192–195; Budziszewski 1980: 604; 1986: 75–78; Fig. I).

Despite the limited geographic range and lithological variety of Turonian sediments in the studied area, flints occurring therein show a significant degree of macroscopic diversity, particularly in color and transparency of primary mass and the nature and thickness of the cortex. These features were the primary criterion for almost all previous attempts of classification of this group of raw materials (Krukowski 1920: 195; Krzak 1965: 222, 1970: 292–295; Balcer 1975: 45–53; Libera and Zakościelna 2002: 96–100), leading to isolation of many local visual varieties (e.g. Gościeradów flint, Janików flint, Ożarów flint, Świeciechów flint or Zawada flint) with very diverse range of occurrence

Number of sample	Type of sample	Place of origin of samples	Macroscopic clas- sification of flint raw material		
G 1–3	Fragments of natural flint concretions	Świeciechów, surface of a prehistoric flint mine	- Gościeradów flint		
G 4		Gościeradów, surface concentration of flint raw material			
J 1-5		Janików, a present quarry	Janików flint		
О 1-4		Ożarów, 'Za Garncarzami' field, surface of a prehistoric flint mine	Ożarów flint		
Ś 1–4		Świeciechów, surface of a prehistoric flint mine	Świeciechów flint		
Z I–II		Zawada, surface concentration of flint raw material	Zawada flint		
Т І–ІІ	Flint artefacts (flakes)	Tominy site 6, settlement objects from the early Neolithic (Linear Pottery Culture)	Zawada flint (?)		

Table 1. Basic	data	for	analysed	samples	of	Turonian	flints	of	the	north-eastern	margin	of	the	Holy
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(Fig. 1). Unfortunately, only a few of them can generally be identified in archaeological contexts, regardless of state of preservation and size of the artifacts, because of unclear and ambiguous descriptions of their physical properties. The existing classifications only occasionally were confirmed by petrographic analyses and, however its range was limited, focusing only on selected varieties of flints from areas located on the left (Stawin 1970: tab. 7–8; Michniak 1980: 85–86) or right (Balcer 1975: 47–48) bank of Vistula did not lead to identification of diagnostic differences between particular varieties.

THE AIM OF THE STUDY

The main objective of the study presented here was to achieve a preliminary mineralogical and petrographic characterization of these varieties of Turonian flints, processing and use of which was confirmed in the Stone Age and Bronze Age¹. The present study was based on the analysis of 28 geological samples (i.e. concretions fragments) representing the following varieties: Gościeradów flint (G), Janików flint (J) Ożarów

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flint (O), Świeciechów flint (S) and Zawada flint (Z; Table I). Particular emphasis was directed toward identification of all diagnostic properties of particular visual varieties from the standpoint of their suitability for the identification in archaeological materials. For this purpose, further analysis was conducted on an additional II artifacts ('T' samples) from the settlement of Linear Pottery culture (hereinafter LPC) site 6 in Tominy, Opatów district (Fig. 1) because the macroscopic properties of these artifacts suggested that they may have come from the outcrop in Zawada, about 100 m to the north (Fig. 2g–h). The objective of the study was to verify this convergence at the mineralogical and petrographic level in regard to Zawada flint samples.

MATERIALS AND METHODS

Thin sections and polished sections from 28 samples were analyzed. They were examined using a Leica DM 2500 P petrographic microscope in reflected and transmitted light, and also by use of a Hitachi SU6600 with EDS addition scanning electron microscope without spraying (SEM–EDS). The aim of the analyses was to determine the structure, texture and mineralogical composition of samples and to obtain information about the presence of mineral inclusions.

RESULTS OF THE STUDY

Gościeradów flint (G1–G4 samples)

This flint variety occurs on the right bank of the Vistula River, most commonly in the vicinity of Gościeradów, Kraśnik district (Fig. 1), in various physical sizes as surface concentrations of fragmented concretions eroded from primary deposits, often accompanied by rubble of weathered rocks (Balcer 1971: 75; Libera and Zakościelna 1987: 40). Concretions are large (up to 30–40 cm in diameter) ovoid in shape, and relatively thick with smooth cortex. The basic mass of silica is dark gray with the presence of very slight, bright spotting and – especially characteristic – whitish and ashy irregular spots with diameters of several tens of millimeters. The basic mass and cortex are separated by a light gray transition zone with a thickness of 0,5–1 cm.

of the Holy Cross Mountains: A – Gościeradów flint; B – Janików flint; C – Ożarów flint;

A-B, E - photo: M. Szeliga; C-D, F-H - photo: T. Wiśniewski.

Fig. 2. Macroscopic differentiation of basic types of Turonian flints of the north-eastern margin

D–E – Świeciechów flint; F – Zawada flint; G – selected polished cross-sections obtained from concretions of the Zawada flint; H – selected polished cross-sections obtained from the flint artefacts (flakes) found on the archaeological site 6 in Tominy.



Previous archaeological studies indicated the use of this variety of flint from the late Paleolithic to Neolithic (Kadrow and Kłosińska 1989: tab. 2; Zakościelna 1996: tab. 43; Libera 2002: 31–44). The samples analyzed and reported here were obtained from the surface of prehistoric flint mine in Świeciechów, Kraśnik district (GI–3) and arable fields in the area of Gościeradów (G4; Fig. 1; Table 1).

Microscopic analysis revealed the presence of oxides and hydroxides of iron and manganese oxides, forming black 'dots' in tested samples (Fig. 3a). These are the two types of grains: small spherical aggregations and single, recrystallized grains, which are accompanied by pyrite. In the background also are crystals of chalcedony, calcite and microfossils (Fig. 3a). Chalcedonic aggregates form spherically-shaped concentrations possibly corresponding to discoloration visible in macroscopic images of tested samples. Foraminifera bioclasts also were found (Fig. 3a) and in the cortical area there also are a number of calcite grains. These data were confirmed by SEM–EDS research, which also identified the presence of aluminosilicates and undefined compounds of iron and manganese, infrequent (especially in non-cortical areas of samples) scattered grains of dolomite and calcite, as well as sulfate (gypsum).

Janików flint (J1–J5 samples)

The occurrence of this flint coincides with the range of detrital bryozoan limestones, defaulting on a small area of Turonian outcrops on the left bank of the Vistula (Fig. 1). The largest exposed deposits occur at a quarry in Janików, Opatów district, where concretions form a series of a dozen of regular layers, each separated by intervals of several tens of centimeters (e.g. Samsonowicz 1934a: 46–47; Pożaryski 1948: 37; Balcer 1975: 53; Michniak 1980: 83; Michniak and Budziszewski 1995b: 50, Fig. 25). Concretions are oval or strongly flattened, irregular and frequently occur in gnarled or jagged shapes. They are covered by quite thick, rough, light beige cortex, with the primary mass of silica a graybrown or brown color containing numerous bright spots up to several millimeters in diameter (Fig. 2b). Previous studies suggested incidental and only local use of this variety of flint during the Late Neolithic and Early Bronze Age (Balcer 1975: 53; Michniak and Budziszewski 1995b: 52). All tested samples were obtained from quarry in Janików (Table 1).

Macroscopic analysis revealed opal-chalcedony background of these rocks containing fossils (foraminifera) that often were filled with iron oxides and hydroxides (Fig. 3b) or carbonates (especially closer to the cortex). SEM–EDS research indicates the presence of alkali feldspar (Fig. 4: a) and calcite, impurities of manganese and iron oxides and small undefined admixture of barite (Fig. 4d).



Fig. 3. Microphotographs of selected samples of Gościeradów flint (A), Janików flint (B), Ożarów flint (C), Zawada flint (D–E) and selected artefact from Tominy (F) made using a polarized microscope in transmitted (B) and reflected light (A, C–D; A–D – crossed Nicols), as well as using back-scattered electrons (BSE) technique by scanning electron microscope with location of places of punctual analyses (E–F). Symbols: brt – barite, cal – calcite, cha – chalcedony, hem ? – iron compounds, probably hematite, Mn – compounds of manganese, py – pyrite. Individual sample numbers appear in the top right corners of each microphotograph. Photo: M. Huber.



Fig. 4. EDS spectra of selected samples of Janików flint (a, d), Ożarów flint (b–c, e) and Świeciechów flint (f–h). Prepared by: M. Huber.

Ożarów flint (O1–O4 samples)

This flint occurs within a small area on left bank of Vistula River, in the immediate vicinity of Ożarów, Opatów district (Fig. 1), in large and very large² oval or flattened concretions with thick and rough white colored cortex (Krzak 1970: 292–295; Budziszewski 1995: 45, Fig. 23). It is has an uneven primary mass color, including different shades of gray which creates a distinctive mottled texture (Fig. 2c). Incidental and only local processing and use of this material has been suggested for early Neolithic with the greatest popularity attained during the early Bronze Age, during which relics of quarry and related workshops for the manufacture of sickles and axes were discovered at the site 'Za Garncarzami' in Ożarów, Opatów district (Budziszewski 1980: 604; 1986: 75–78). All samples analyzed for purposes of this article were obtained from fragments of natural concretions defaulting on the surface of this site (Table 1).

Analysis of samples in transmitted and reflected light revealed spherical and streak concentrations of chalcedony of varying degree of crystallinity and the presence of small inclusions of iron and manganese oxides, as well as bioclastic (crushed detritus of mollusk shells) constituents. In the background rhombic grains of calcite were identified, as well as single grains of iron and manganese oxides (Fig. 3c). SEM– EDS research confirmed the presence of K-feldspar-like phase, mica and apatite inclusions, and also iron compounds (oxides, hydroxides). In the case of sample O2 the analysis also revealed a high amount of fluorine, with a minimal calcium content (Fig. 4c). Determination of its nature and phase requires further research. Small amounts of titanium, nickel, zinc, and copper of indeterminate phase (Fig. 4b, e) also was identified in the samples tested. The titanium and nickel may derive from silicates or oxides, whose presence was recognized in a previous study of this kind of flint (Michniak 1980: Tab. 1).

Świeciechów flint (Ś1–Ś4 samples)

This variety of flint occurs on the right bank of Vistula River, where there are several known areas of secondary, surface concentrations of concretions³. The largest of them (about 180 ha) was recorded in Świeciechów, Kraśnik district, while

² Concretions usually reach a diameter of up to several tens of centimeters. In the profile of one of the quarries in Karsy, north of Ożarów, flattened concretions at least 12 meters long and several tens of centimeters thick have been documented (Budziszewski 1980: Fig. 618).

³ Primary deposits of this raw material were documented during geological surveys in 1st half of 20th century. In one of the Świeciechów quarries it occurred as large (diameter up to 50 cm), flattened concretions, occurring in the rock about 2 meters below the surface (Samsonowicz 1924: 100). The primary deposit also was observed in a road cut near the local cemetery (Pożaryski 1948: 47). Despite later attempts (Balcer 1971: 93, 1976: 184), we failed to re-locate the primary deposits of this variety of flint and verify previous observations.

the remaining, much smaller, is located near Rachów Nowy, Kraśnik district, and Wymysłów, Ostrowiec Świętokrzyski district (Fig. 1). Outcrops in Świeciechów and in Rachów Nowy were connected with relics of intensive mining and processing activity in the Neolithic, Bronze Age and early Iron Age (Balcer 1971: 118; 1976: 192–195; Bargieł and Libera 1996: 37–38).

The silica primary mass of this flint is characterized by gray and olive-gray colors (Fig. 2d) and various shades of gray often co-occur with each other (Fig. 2e). The primary mass contains white or yellowish calcite precipitates (Balcer 1975: 47-48) and slightly less frequently, small spots a few millimeters diameter. The contact juncture between the silica primary mass and the surrounding rock has a rough textured white or light beige surface of varying thickness (Fig. 2d). Fragments of concretions found on the surface may not preserve this contact juncture because of smoothing and polishing resulting from aeolian processes. The presence of aeolized natural faces is one of the diagnostic macroscopic properties of this variety (Samsonowicz 1924: 99–100; Krzak 1965: 220; Balcer 1971: 110–111, 1975: 50; Libera and Zakościelna 2002: 96). The use of Świeciechów flint has been suggested for the Paleolithic and Mesolithic (e.g. Cyrek 1981: Fig. 24-25; Libera, 2002: 31-34; Kaczanowska and Kozłowski 2005: Fig. 5-6), but the most intense processing and use took place in the Neolithic, during development of the Funnel Baker Culture (Balcer 1976: 192–195). All samples of this variety analyzed in the present study, represent fragments of natural concretions collected from the surface of a Neolithic quarry in Świeciechów (Table 1).

SEM analysis of the background of silica represented by concentrations of chalcedony (in varying degree of crystallinity) revealed visible fillings composed by chalcedony with admixture of opal. Calcite and clay minerals crystals occur in the background, forming irregular concentrations, along with a few ore minerals (pyrite and iron oxides), isolated parts of bioclastic content (e.g. foraminifera, needles of sponges) and single grains of feldspar. In some specimens micro-faults filled by recrystallized quartz and opaque minerals are visible. Therefore, characteristic discolorations occurring in these flints may be the result of concentration of clay minerals, carbonate crystals and secondary epigenetic quartz (with a much lower share of coloring iron compounds). SEM–EDS analyses also revealed the presence of apatite, aluminosilicates and manganese oxides. In some samples were found small admixture of barite and titanium (Fig. 4g), as well as lead and zinc - most likely as sulphides (Fig. 4f, h).

Zawada flint (Z1–Z11 samples)

So far, the existence of this variety of flint is confined to a small quarry in Zawada, Opatów district (Budziszewski and Michniak 1989: 151; Szeliga 2014: 90, Fig. 5: A–B), backfilled during 2006. At the present time only a concentration of eroded



Fig. 5. EDS spectra of selected samples of Zawada flint (a–c, e) and artifacts ('T' samples) from site 6 in Tominy (d, f–g). Prepared by: M. Huber.

flint concretion fragments remain, on the surface in the immediate vicinity of the quarry (Fig. 1). These fragments represent quite regular, round and slightly flattened shape of concretions, often reaching considerable size and weight, covered with thick (although easy to rub away) white and yellowish cortex. The primary mass of silica is dark gray, occasionally brightened toward the centre of the concretion, where it is noticeably tarnished. Within it occur quite regular, bright spots, up to several millimeters in size, sometimes taking the form of intensive mottling. Its characteristic feature is the presence of a thin (2–4 mm), generally brown or dark purple (sometimes black) layer between the primary mass and cortex (Fig. 2f–g). The presence of this feature provided the basis for our idenfication of this material in LPC flint inventory from nearby site 6 in Tominy (Fig. 1; 2h). All analyzed geological samples of this variety come from the surface outcrop in Zawada (Table I).

The analysis of microscopic images revealed the presence of sulphides (pyrite occurring as euhedral crystals accompanyied by oxides and hydroxides of iron) visible on the background of numerous concentrations of chalcedony, including siliphicated organic fossils (foraminifera). In the background are crystals of calcite, along with single grains of feldspar (plagioclase) and quartz. SEM–EDS analysis also revealed the presence of apatite, pyrite and barite (Fig. 3d–e; 5a–b), with barite between the grains of pyrite and iron compounds. In those rocks there are also carbonates of glauconite-like phase. Inclusions of fluorspar also were identified, and certain undefined admixture-of titanium and nickel (Fig. 5c, e).

Archaeological samples from Tominy (T1–T11)

SEM analysis was conducted on 11 flakes from LPC context at site 6 in Tominy, located approx. 100 m south of the outcrop in Zawada (Fig. 1), verify to the distinct convergence of macroscopic features of artifacts with Zawada flint.

Microscopic analyses revealed a large amount of pyrite, visible on the background of studied samples (similar to samples of Ożarów flint), although in this case the sulphides have anhedral forms. Oxides and hydroxides of iron and numerous concentrations of chalcedony were observed, as well as calcite and quartz. SEM–EDS analysis also revealed the admixture of barite, which lie between the grains of pyrite and iron compounds (Fig. 3f; 5d). The presence of undefined admixture of cobalt, bismuth, titanium and strontium also was confirmed, as well as concentrations of barite, phosphates and aluminosilicates (Fig. 5d, f–g).

CONCLUSIONS

The results of microscopic research reported here, including SEM–EDS, are in accord with the results of previous mineralogical analyses (Stawin 1970: Table 7–8; Balcer

1975: 47–48; Michniak 1980: 85–86). Despite the increasing knowledge about the nature and mineralogical and petrographic diversity of basic varieties of Turonian flints in the north-eastern margin of the Holy Cross (Świętokrzyskie) Mountains, microscopic analyses failed to capture the significant differences in mineral composition of analyzed samples, each time revealing the dominant content of silica group minerals, and presence of different admixture, represented by oxides and hydroxides of iron and manganese, as well as aluminosilicates, apatite, pyrite, and also calcite (especially nearby the cortex). Current data do not reveal unequivocal diagnostic features to allow unambiguous correspondences between archaeological materials and flint raw materials originating in particular exposures of Turonian outcrops.

Despite the large mineralogical convergence of tested samples, this study captured some individual features, manifested by greater or lesser intensity within different varieties of Turonian flints. These data allow positive promise for the future, revealing at the same time the need to broaden the scope of research with more samples and additional chemical analyzes (e.g. ICP-MS), aimed at more precise specification and quantification of the admixture-of various minerals (sulphide, sulphate, including barite) and metals (e.g. manganese, lead, zinc, copper and strontium; see Fig: 4b, d, f, h; 5) identified here. The need for more precise, quantified corroborative analyses also applies to the observed mineralogical convergence between the samples of Zawada flint (ZI-II) and a series of archaeological samples from site in Tominy (TI-II), manifested primarily in the coexistence of pyrite grains with characteristic skeleton-like shape filled with barite, the presence of undefined compounds of titanium and other metals (Fig. 5), and enhanced of transition zone between the cortex and primary mass of silica by oxides and hydroxides of iron. Regardless, we conclude that the convergence of these microscopically generated features are sufficient to identify the outcrop in Zawada as the place of origin of the raw materials used in production of artifacts analyzed from site 6 at Tominy, confirming the knowledge, processing and use of Zawada flint by local early agrarian communities at the turn of the 6th and 5th millennia BC. This findings from Tominy represents the only instrumentally-based confirmation of processing and use of Zawada flint in prehistory.

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