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Siliceous rocks from the southern part of the Kraków-Częstochowa Upland (Southern Poland) as potential raw materials in the manufacture of stone tools – A characterization and possibilities of identification

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ABSTRACT

The Upper Jurassic sediments from the Kraków-Częstochowa Upland developed as carbonates, mostly limestones, which represent the microbial-sponge megafacies that were typical of the northern margin of the Tethys Ocean. Outcrops of these rocks are scattered along an extended belt ranging from Portugal to the Caucasus Mts. One of the common features of the bedded limestones belonging to this megafacies is the local occurrence of chert concretions.

The siliceous rocks embedded within the Upper Jurassic sequences from the southern part of the Kraków-Częstochowa Upland originated from a broad spectrum of limestone silicification processes and are represented by chert concretions, bedded cherts and epigenetic siliceous rocks. These rocks served as raw materials in the production of stone tools from the Middle Palaeolithic onwards. These tools have been studied extensively by archaeologists.

This paper presents the identification of the above mentioned types of siliceous rocks used in the manufacture of tools based upon (i) microscopic observations of thin sections which reveal the primary microfacies of limestones subjected to silicification, and (ii) X-ray diffraction analyses, including the determination of the crystallinity index of SiO_2. Unfortunately, the available research methods do not permit the identification of the varieties of chert concretions to an extent that would permit them to be even roughly connected with particular outcrops or, at least, with particular regions of the occurrence of siliceous raw materials on a Pan-European scale. Macroscopic criteria have recently been adopted in archaeological classifications aimed at determining the origin of stone tools and drawing conclusions as to the source outcrops or regions. However, regrettably, these have proven groundless from the geological point of view.

1. Introduction

The Upper Jurassic sediments from the southern part of the Kraków-Częstochowa Upland (KCU) (Fig. 1) are home to a variety of siliceous rocks. In the Middle Palaeolithic, this region was settled by humans who used these rocks for the manufacture of stone tools. In archaeological research, these siliceous raw materials are mostly categorized according to their macroscopic features and they are usually linked with particular regions or even particular outcrops.

The following paper provides a mineralogical and petrographic characterization of siliceous rocks in Upper Jurassic limestones from the southern part of the KCU. This characterization is closely related to the development of the primary depositional structures of their limestone hosts. The methodology includes macroscopic and microscopic descriptions, microfacial analysis and X-ray diffraction patterns. This attempt enabled the authors to relate the types of siliceous rocks distinguished to particular stratigraphic intervals of the Upper Jurassic succession from the southern KCU. Moreover, it was possible to determine the provenance of the siliceous raw materials from which the tools were made based upon these macroscopic features.

The methods used in the identification of the different types of siliceous rocks presented in this paper belong to the destructive analyses group. The research results obtained with these methods enabled us to delineate different types of silication, but for valuable stone tool collections it is necessary to develop non-destructive methods of analysis. Both the detailed results and the interpretation of the geochemical

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no.	name	chronology	type of archaeological site
1	Czernichów	Upper Palaeolithic	chert outcrop and chert workshop
2	Wołowice	Upper Palaeolithic-Neolithic	chert mine and chert workshop
3	Dąbrowa Szlachecka	Upper Palaeolithic	chert outcrop and chert workshop
4	Ściejowice	Upper Palaeolithic-Mezolithic	chert outcrop and chert workshop
5	Piekary	Upper Palaeolithic and Bronze Age	open site on loess and chert workshop
6	Kraków-Spadzista	Upper Palaeolithic	killing, butchering site and chert workshop
7	Kraków-Sowiniec	Middle Palaeolithic	open site on loess
8	Brzoskwinia	Upper Palaeolithic-Neolithic and Bronze Age	chert mine and chert workshop
9	Mammoth Cave	Upper Palaeolithic	settlement cave site
10	Wierzchowska Górna Cave	Upper Palaeolithic and Bronze Age	settlement cave site
11	Nietoperzowa Cave	Upper Palaeolithic and Bronze Age	settlement cave site
12	Ciemna Cave	Middle Palaeolithic-Medieval Age	settlement cave site
13	Owczary	Mezolithic	chert outcrop and probably mine
14	Iwanowice	Neolithic-Bronze Age	settlement site

Fig. 1. Location of the study area against the bedrock geological map after Gradziński (2009) with archaeological sites marked and types of silicification identified.

analyses of major elements and REE, stable isotope analyses and nuclear magnetic resonance spectroscopy of siliceous rocks and hosting limestones are far beyond the volume and the scope of this publication and thus, together with the results of non-destructive methods (e.g. Raman spectroscopy), will be presented in separate forthcoming papers. Here, only the data which is crucial to the argumentation are included.

2. History of research

The southern part of the KCU has been inhabited since at least the Middle Palaeolithic (Fig. 1), with one of the primary reasons being the availability and high quality of the local siliceous raw materials. There is a remarkable correlation between regions rich in siliceous raw materials and human settlements representing successive archaeological cultures.

2.1. Archaeological studies of siliceous rocks

The first geoarchaeological paper which attempted to indicate the geological sources of archaeological artefacts was published by Krukowski (1920), who distinguished two types of cherts based upon the transparency, fracture, colour and character of the cortex. These cherts originated from regoliths but, unfortunately, Krukowski did not provide detailed locations for his chert specimens.

Kaczanowska and Kozłowski (1976) published an attempt at the categorization of siliceous raw materials from the southern KCU based upon colour, the character of the matrix, cortex and their mutual relationships. Their analysis dealt mostly with stone inventories from both the Neolithic and Eneolithic sites in Nowa Huta (an eastern suburb of Kraków) and did not include the characterization of siliceous rocks in outcrops. Kaczanowska and Kozłowski (1976) distinguished six varieties of Jurassic cherts. However, the subjective nature of the criteria they applied to the evaluation of archaeological artefacts was criticized by Morawski (1976), rendering this categorization problematic.

Important geoarchaeological information was provided by Lech (1980), who characterized siliceous raw materials from the Kraków region and discussed the accessibility of chert deposits in prehistory, as well as the forms of their accumulations in sediments. He also noticed that a significant proportion of geological publications have never been (and are still not) cited in the archaeological literature, nor have their results ever been used in archaeological research. Geoarchaeological information, together with the results of laboratory methods (X-ray, IR and chemical analyses as well as microscopic observations), has only occasionally been utilized (see e.g. Lech et al., 1976; Kaczanowska et al., 1979; Małecka-Kukawka et al., 2016). However, these authors did not draw any conclusions and only postulated the necessity for further study.

Most opinions on the origin of cherts collected at archaeological sites, whether expressed in monographs (e.g. Ginter and Kozłowski, 1990) or specialist papers devoted to particular sites and their stone inventories in the KCU, advocate the provenance of the cherts from the three regions: southern KCU (i.e. the Kraków area), central KCU and northern KCU. The siliceous rocks from the southern KCU are usually termed in the archaeological literature as “Jurassic flint” or the “Jurassic flint around Kraków” (e.g. Lech, 1981; Ginter and Kozłowski, 1990; Přichystal, 2013, 2018; Brandl 2013–2014; Brandl et al., 2016).

The most recent summary of the petrography and varieties of Jurassic cherts from around Kraków can be found in Přichystal (2013). This paper provides some new data, despite being based on the articles mentioned previously. In comparison with earlier archaeological publications, its novelty lies in the use of the Munsell colour chart to define the colours of A-G chert varieties.

2.2. Geological and mineralogical studies

Geological studies of siliceous rocks from the Upper Jurassic successions of the KCU date back to the early 19th century. However, these were mostly conceptual works based upon macroscopic descriptions, whereas studies including geochemical analyses and/or mineralogical/petrographic observations were rare. The same is true of investigations in which the development and origin of siliceous rocks was presented against the background of the lithology of their limestone hosts.

Considering the historical context, the products of the silicification of Upper Jurassic limestones from both the southern and the central parts of the KCU were described by e.g. Staszic (1815), Zejszner (1841), Roemer (1870), Wiśniowski (1888), Zaręczny (1894), Gaweł (1925), Sujkowski (1926, 1937) and Kuźniar and Żelechowski (1927). Although these authors focused mostly on the macroscopic description of chert concretions, Zejszner (1841) noticed that the types of chert concretions are related more to the lithostratigraphy of the host limestones than to their geographical distribution. Unfortunately, publications dealing directly with Jurassic cherts, including their genesis, development, petrography, geochemistry, weathering and fossils were limited in both number and extent (see e.g. Ehrenberg, 1854; Wiśniowski, 1888; Miernik, 1911; Gaweł, 1925).

Modern publications presenting the studies on siliceous rocks from the KCU include: Dżułyński (1952), Bukowy (1956), Alexandrowicz (1960), Rajchel (1970), Pawlikowski et al. (1978), Matyszkiewicz (1987, 1989, 1996), Michniak (1989), Świerczewska (1997), Sitarz et al. (2014) and Matyszkiewicz et al. (2015a, 2015b, 2016). However, most of them deal rather with the regional geology, the analysis of the development of Upper Jurassic limestones or are detailed mineralogical works devoid of a more general geological context.

A separate group of publications describes the potential industrial utilization of the cherts. The earliest papers date back to the 19th century (Roemer 1870), when cherts from the KCU were proposed as a potential raw material (Flintenstein) for the production of flintlocks for firearms. In the second half of the 20th century, studies were conducted on the utilization of chert concretions for the production of abrasives for e.g. pebble mills (see e.g. Chudziński and Stawin, 1965; Morawiecki, 1965; Ruśkiewicz, 1968; Stawin, 1969, 1970). However, these industrial papers mostly provided the results of technological analyses of siliceous raw-materials.

3. Methods, materials and terminology

The following paper provides largely new results of recent studies, although it also includes some previously published data (Matyszkiewicz, 1987, 1996; Świerczewska, 1997).

The sample set comprises 84 samples collected from 21 outcrops located in the southern part of the KCU, comprising: 10 outcrops of chert concretions (52 samples), 3 outcrops of bedded cherts (18 samples) and 8 outcrops of epigenetic siliceous rocks (ESR) (15 samples). From the collected samples, 39 polished sections and 53 thin sections were prepared, which were subsequently examined for microfacial analysis using an OLYMPUS SZX9 microscope connected to a 5 MpixFireWire camera.

The selected samples of chert concretions (16 items) and bedded cherts (4 items) were analysed at the Laboratory of Phase, Structural, Textural and Geochemical Analyses of the Faculty of Geology, Geophysics and Environmental Protection, AGH University of Science and Technology in Kraków. The X-ray diffraction method (DSH) was applied in order to establish their mineral composition and the crystallinity index of the quartz. Samples were analysed with a Rigaku SmartLab diffractometer under the following analytical conditions: graphite-monochromatized $Cu_{K\alpha}$ radiation (voltage of 45 kV, current of 200 mA), step 0.05° 2Θ, counting rate 1 sec/step. The identification of mineral phases was based upon the interplanar spacings determined from diffractograms using the ICDD (2014) catalogue of diffraction data and the XRAYAN software. The crystallinity index (CI) of quartz was determined in accordance with the procedure described by Murata and Norman (1976). The method used to determine the crystallinity index of quartz (CI) after Murata and Norman (1976) was critically assessed by Marinoni and Broekmans (2013), since it does not express the alkali potential reactivity of quartz (cf. Broekmans, 2012). However, from a practical point of view, the CI-index allows early diagenetic chert concretions to be distinguished, as well as bedded cherts from the ESR.

For the description of SiO_2 phases, the simplified terminology was applied after Folk and Pittman (1971) and Klein and Hurlbut (1985), in which chalcedony, lutecite, quartzine and microflamboyant quartz (=flamboyant lutecite) were ascribed to a single group of fibrous varieties of SiO_2. In this group, moganite (microcrystalline SiO_2 polymorph) was identified by means of X-ray diffractometry (cf. Flörke et al., 1984; Miehe et al., 1984; Heaney and Post, 1992; Miehe and Grätsch, 1992; Grätsch and Grünberg, 2012; Zhang and Moxon, 2014).

Moreover, the cryptocrystalline silica (< 4 µm in diameter), microquartz (grains between 4 and 20 µm in diameter) and megaquartz (the term used for crystals over 20 µm in diameter) were also identified.

4. Geological setting

The siliceous rocks described below occur mostly within the Upper Jurassic normal facies (= bedded facies), sediments belonging to the so-called microbial megafacies (Gwinner, 1971), which occupies an extended area in Europe, from Portugal through Spain, France, Germany, Poland, Romania, Crimea and reaching as far as the Caucasus Mts. These megafacies were formed in the Late Jurassic over the northern margin of the Tethys Ocean. The analysis of publications describing the lithology of these megafacies in various parts of Europe (e.g. Gwinner, 1976; Flügel and Steiger, 1981; Gaillard, 1983; Geyer and Gwinner, 1984; Keupp et al., 1990; Dromart et al., 1994; Leinfelder and Keupp, 1995; Herrmann, 1996; Reolid, 2007; Krajewski, 2010; Matyszkiewicz et al., 2012, 2015a) together with rare comparative papers (Leinfelder et al. 1994; Matyszkiewicz, 1997) demonstrate a significant similarity between the lithologies of these sediments. One of their typical features is the presence of chert concretions in bedded limestones.

The Upper Jurassic (Lower Oxfordian-Lower Kimmeridgian) succession from the southern KCU (Fig. 1) comprises carbonates of a total thickness up to about 250 m. Three principal facies varieties are known: bedded facies, massive facies and submarine gravity flows facies (Fig. 2). In these facies, three silicification types and products were distinguished, presumably of diverse origin and with an unclear source of silica. The most common silicification, Type I, is responsible for the chert nodules, which occur chiefly in Middle Oxfordian-Lower Kimmeridgian bedded limestones, and only rarely in platy limestones occupying the lower, Middle Oxfordian part of the Upper Jurassic succession. Much less frequent is Type II silicification, which resulted in the bedded cherts whose occurrence is limited to calciturbidites deposited at the Oxfordian/Kimmeridgian turn. Both silicification types (and products) reveal clear relationships to limestone facies. The epigenetic Type III silicification and its products – the ESR are unrelated to any defined limestone facies. These are irregular silica bodies observed exclusively in the topmost part of the Upper Jurassic succession.

5. Chert concretions

5.1. Macroscopic features

The chert concretions (Figs. 3a and b, 4a, b, e, f) form ellipsoidal, spheroidal or, less commonly, irregular nodules, up to dozens of centimetres across (on average, up to about 10 cm). Their colours vary from grey through dark-grey, bluish and brownish to black. Usually, the nodules are covered with bright-grey or white rims which are several millimetres thick. Locally, in the topmost part of the Upper Jurassic succession and close to fault zones, chert concretions are overgrown by grey, brownish or creme, silicified rims (Type III silicification), up to several centimetres thick (Fig. 4f). The fracture of concretions is conchoidal, i.e., smoothly carved, with typical, arched surfaces. Locally, the fracture surfaces disclose relics of limestones in the form of bioclasts (mostly fragments of skeletons or spicules of siliceous sponges, echinoderms and/or fine brachiopods) and larger intraclasts (Fig. 4a and b). In the vicinity of fault zones, chert concretions are sometimes strongly fractured (Fig. 5c and d). Chert concretions from bedded limestones occasionally contain geodes, some centimetres across, filled partly or completely with coarse-crystalline quartz (Fig. 5b) or internal carbonate sediment. Chert concretions hosted in calciturbidites do not contain geodes (Fig. 6d).

5.2. Host rocks

The chert concretions occur mainly in the sediments of the normal facies, usually in bedded limestones (Fig. 3a and b) located in both the middle and the upper parts of the Upper Jurassic succession (Dżułyński, 1952; Matyszkiewicz, 1989, 1997; Matyszkiewicz et al., 2015a). These sediments reveal high contents of $CaCO_3$ but low amounts of Al_2O_3. Locally, in the upper part of the succession, bedded limestones are dolomitized. It is interesting that such dolomitized limestones do not contain chert concretions (Łaptaś, 1974). The lower parts of the Upper Jurassic succession from the southern KCU comprise so-called platy limestones with marl intercalations that are several-millimetres-thick. These sediments have higher contents of Al_2O_3 and usually do not host chert concretions.

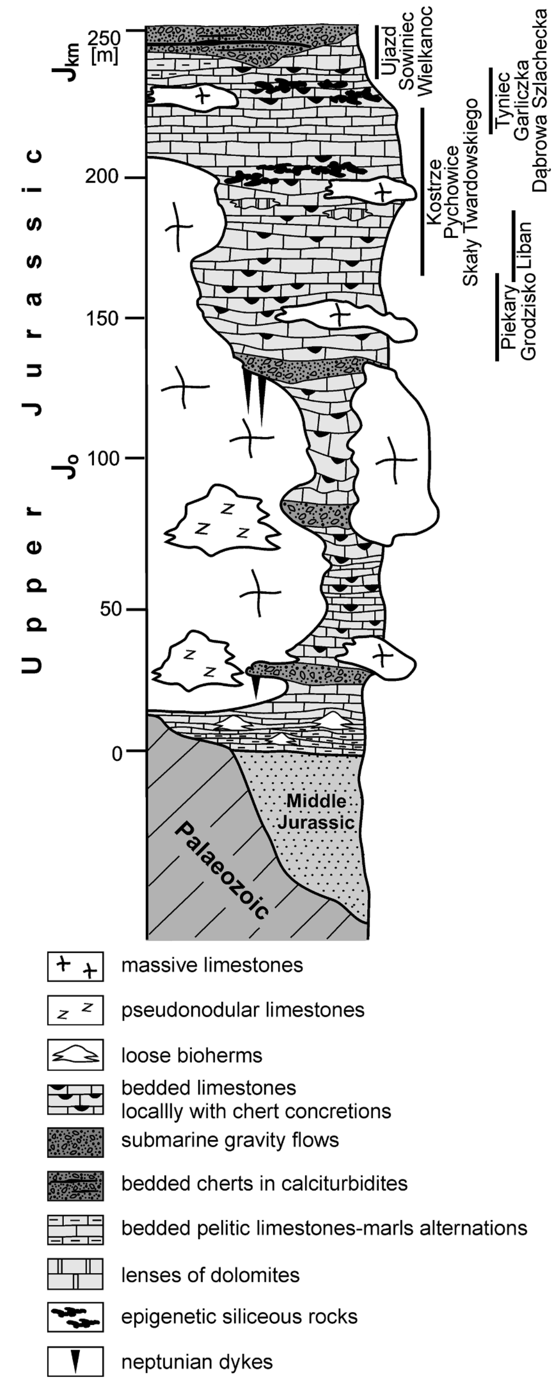

Fig. 2. Lithostratigraphic column of the Upper Jurassic (Oxfordian (J_o) – Kimmeridgian (J_{km})) sediments from the southern KCU with the approximate stratigraphic position of outcrops with siliceous rocks described in the text (after Matyszkiewicz et al., 2012, partly modified).

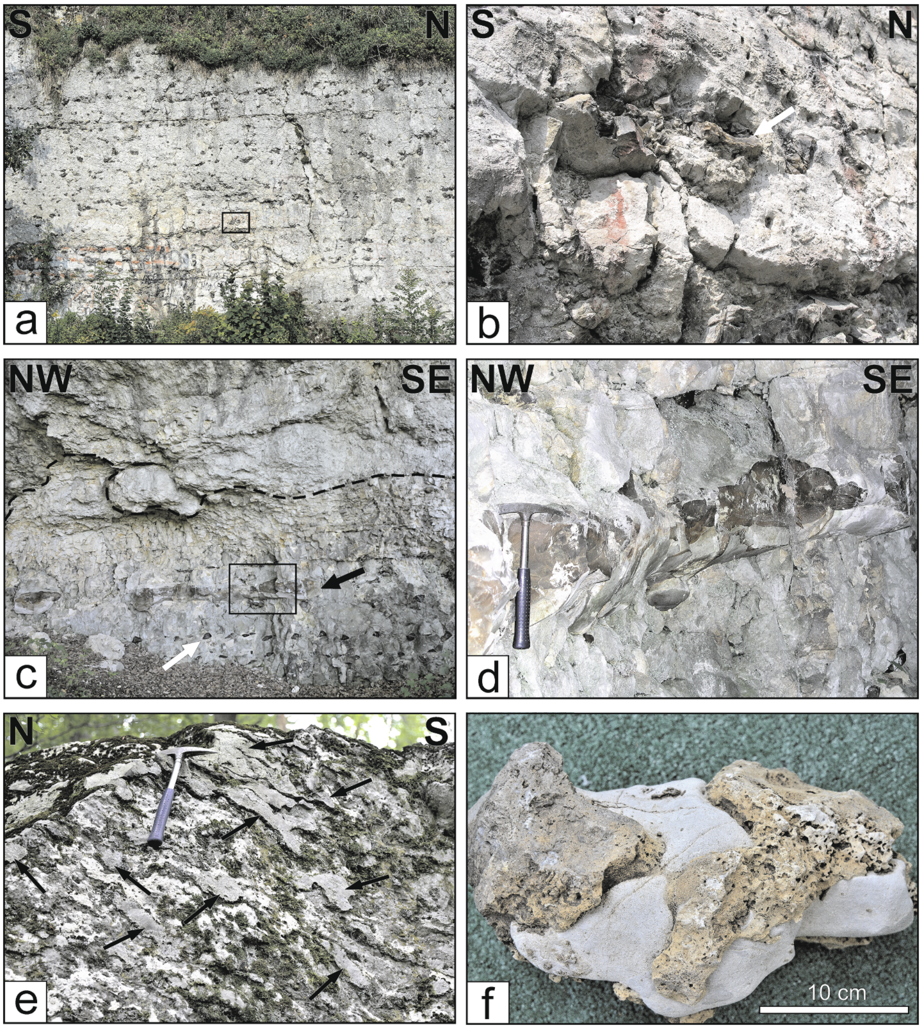

Fig. 3. (a) Horizons of chert concretions distributed in parallel to the bedding planes in bedded limestones. The area in the rectangle is shown in Fig. 3b. Piekary, Upper Oxfordian. (b) Fragment of outcrop wall from Fig. 3a with chert concretions. (c) Calciturbidite (bottom, marked by a dashed line) covered by debris flow. The black arrow indicates the bedded chert layer in calciturbidite. A white arrow marks the horizon of chert concretions located beneath the bedded chert and arranged parallelly to bedding plane. The area in the rectangle is shown in Fig. 3d. Ujazd, uppermost Oxfordian-lowermost Kimmeridgian. (d) Details of a bedded chert horizon in calciturbidite from Fig. 3c. (e) ESR developed as irregular nests in the topmost part of bedded limestone (arrows). Garliczka Valley, Upper Oxfordian. (f) A fragment of limestone with ESR. Sample from the regolith covering the top surface of the Upper Oxfordian limestones in the Garliczka Valley.

The bedded limestones are mostly wackestones or packstones (terminology after Dunham, 1962) and represent detrital sediment deposited on the seafloor in areas between the so-called carbonate buildups. These are chiefly composed of microbialites with subordinate siliceous sponges (Matyszkiewicz et al., 2012; Krajewski et al., 2018). Occasionally, bedded limestones developed as biostromes built of boundstones, which contain calcified siliceous sponges and microbial structures (Matyszkiewicz, 1989; Matyszkiewicz et al., 2012). In bedded limestones, chert concretions are usually distributed in horizons which are in parallel to the bedding planes (Fig. 3a). In transition zones from bedded to massive limestones, cherts are randomly scattered within host rocks and gradually disappear (Matyszkiewicz, 1989).

Occasionally, chert concretions can be encountered in submarine gravity flow deposits, which dominate the upper parts of the Upper Jurassic succession. One can observe chert nodules randomly scattered within the sediment or distributed within the calciturbidites (Figs. 3c, 4d). In the latter sediments, chert concretions occur in bedded limestones, precisely in both the grainstones-packstones located beneath the bedded cherts and in the wackestones-mudstones located above the bedded cherts. In calciturbidites, chert concretions are distributed in horizons which are parallel to the bedding planes (Fig. 3c; Matyszkiewicz, 1996; Matyszkiewicz et al., 2015a).

5.3. Microscopic features

The chert concretions are composed of several SiO_2 varieties: mostly cryptocrystalline but also fibrous, locally even of micro- and megaquartz. Also common are relics of silicified limestone components (Fig. 5a and b), such as bioclasts, and also, albeit more exceptionally, microbial structures. The silicified bioclasts, usually with well-visible contours, are built of fibrous SiO_2 microcrystalline quartz or even megaquartz. Sometimes, chert concretions host geodes up to several centimetres across, the walls of which are covered with quartz crystals several-millimetres-long. Some geodes are filled entirely with megaquartz crystals (Fig. 5b) or laminated carbonate internal sediments. The fractures typical of chert concretions hosted in limestones from large fault zones are filled with carbonate sediments which are partly silicified with cryptocrystalline SiO_2.

The lithostatic stylolites which are common in limestones are absent from the chert concretions. However, if such concretions have rims produced by epigenetic silicification, the stylolites are also silicified (Fig. 5e and f).

5.4. Mineral composition and crystallinity index of SiO_2

The main components of the chert concretions are fibrous varieties of SiO_2 including moganite, identified by means of X-ray diffractometry (Fig. 8; Table 1; cf. Sitarz et al., 2014). Moganite domains alternated with quartz compose the fibrous varieties of SiO_2. Locally, quartz was observed together with accessory calcite and Fe-oxides. The CI values of the chert concretions are diverse, but some regularities can be discerned (Table 1). The highest CI value of about 2.3 (on average, 1.1) is encountered in concretions from horizons in bedded limestones originated from the areas where extended fault zones are absent. In samples from

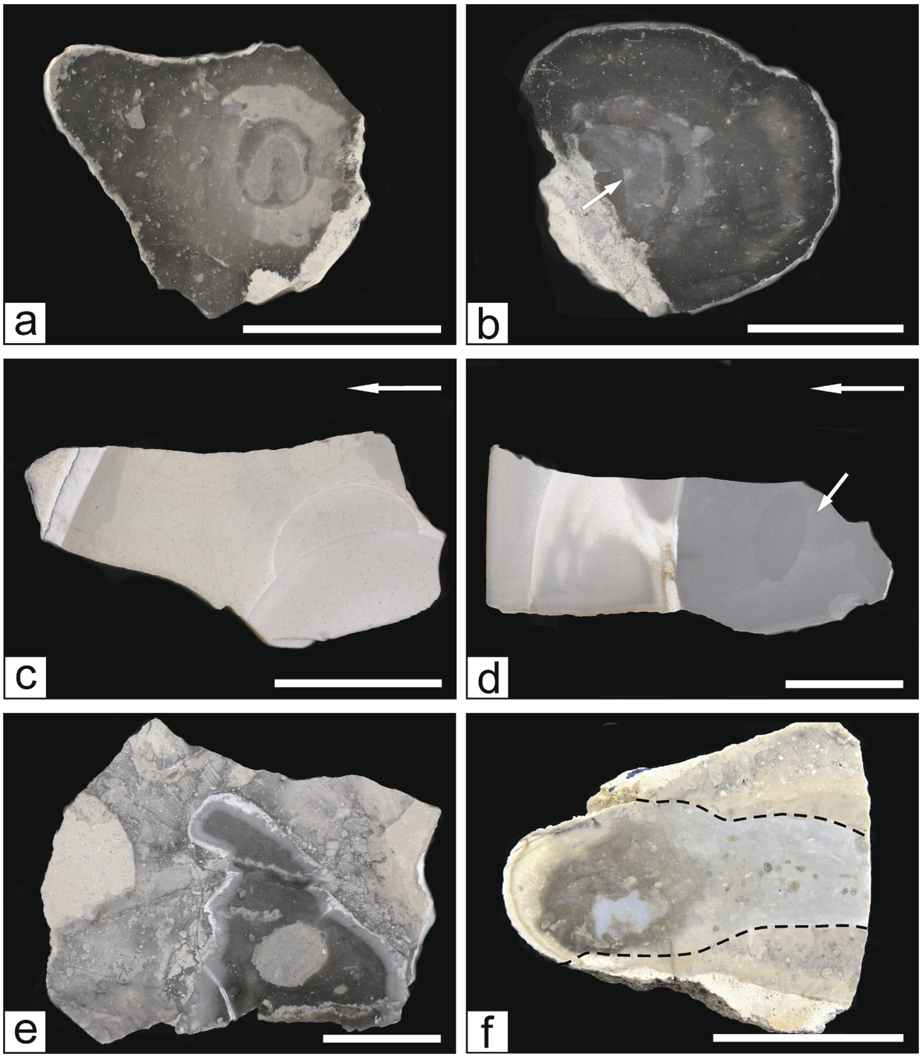

Fig. 4. (a) Chert concretion from bedded limestone. On the right – preserved skeleton of siliceous sponge with another sponge skeleton inside. To the left of the sponges – fragments of fine bioclasts and single intraclasts embedded in a brownish SiO_2 matrix. Sample KA-6, Piekary, Upper Oxfordian. (b) Chert concretion from bedded limestone with diverse colours. On the left – two dish-shaped siliceous sponges (arrows) seen at the contact point with the limestone. Sample KA-7, Piekary, Upper Oxfordian. (c) Bedded chert from calciturbidite. On the right – a fragment of an occluded chert concretion overgrown by concentric growth layers. The arrow marks the position of the top surface. Sample SB-1, Sowiniec Horst, uppermost Oxfordian-lowermost Kimmeridgian. (d) Bedded chert from calciturbidite. On the right – a chert concretion (arrow) on which the bedded chert developed. Concentric growth layers are visible. On the left – a spotty pattern of various colouration in the brighter part of bedded chert, related to selective silicification of bioturbated sediment. An arrow marks the position of the top surface. Sample SA-2, Ujazd, uppermost Oxfordian-lowermost Kimmeridgian. (e) Epigenetic silicification of a chert concretion – the primary sedimentary structure of limestone is well-visible in the epigenetically silicified portion but it is almost completely obliterated in the chert concretion. Small fragments of non-silicified limestone can be seen on both the right and left sides of polished section. Sample KB-7, Dąbrowa Szlachecka, Upper Oxfordian. (f) The rim of the epigenetically silicified limestone developed around the chert concretion. A dashed line separates the chert concretion from the epigenetic silicification zone. Fragments of non-silicified limestone seen on the top and at the bottom of polished section. Sample DG-1, Garliczka Valley, Upper Oxfordian. The scale bar in all photographs a-f is 5 cm.

large fault zones, where the only SiO_2 variety is quartz, CI values rise to even 5.4.

5.5. Genesis of cherts and a plausible source of SiO_2

The generally accepted opinion as to the plausible source of SiO_2 in chert concretions from bedded limestones of the Upper Jurassic microbial-sponge megafacies are skeletons of sponges primarily composed of opal-A. However, in the southern KCU, chert concretions occur not only in sediments rich in siliceous sponges (recently calcified) but also in deposits composed mostly of microbial structures with only a minor contribution from siliceous sponges, or even their complete absence.

In calciturbidites from the topmost part of the Upper Jurassic succession (Fig. 2), siliceous sponges practically do not occur and chert concretions are accumulated both beneath and above the bedded cherts. On the contrary, in the middle part of the succession, in the so-called pseudonodular limestones containing abundant calcified siliceous sponges, chert concretions practically do not occur (Matyszkiewicz and Kochman, 2016). These facts suggest that in various time periods, the diagenetic environment of the bedded limestones was invaded by episodic influxes of SiO_2-rich fluids, not necessarily related to the periodic abundance of siliceous sponges.

According to Migaszewski et al. (2006), the sources of SiO_2 for Middle Oxfordian-Lower Kimmeridgian chert nodules from the Holy-Cross Mts. (south-central Poland) were sea-floor outflows of hydrothermal fluids genetically linked to extensional tectonics. In both the southern and the central part of the KCU, manifestations of hydrothermal activity in the Upper Jurassic sediments were described by Matyszkiewicz (1987), Gołębiowska et al. (2010), and Matyszkiewicz et al. (2015b, 2016). Moreover, accumulations of hydrothermal megaquartz were described from the Oxfordian neptunian dykes, which are unambiguously related to Late Jurassic extensional tectonic events (Matyszkiewicz et al., 2016). Hence, an abiotic source of SiO_2 for the Upper Jurassic chert concretions and multistage silicification processes cannot be ruled out at this stage.

6. Bedded cherts

6.1. Macroscopic features

The bedded cherts form layers, up to 0.4 m thick and up to several meters long (Fig. 3c and d). In the outcrops, a maximum number of three layers was observed, as in the Wielkanoc Quarry (Fig. 1). The bottom surface of the bedded cherts is irregular and shows some bulges (Fig. 3d) whereas the top surface is usually flat. Within the bedded chert layers, occluded chert concretions and concentric growth layers can be observed (Fig. 4c and d). Colours of particular growth layers vary from creamy, grey or brownish to dark-creamy. Generally, the colours of central parts of chert beds are darker and grade outside to more brighter ones (Fig. 4c and d). Locally, the outer parts of the bedded cherts show silicified bioturbations (Fig. 4d). Due to their higher resistance to weathering, the bedded cherts protrude from outcrop walls

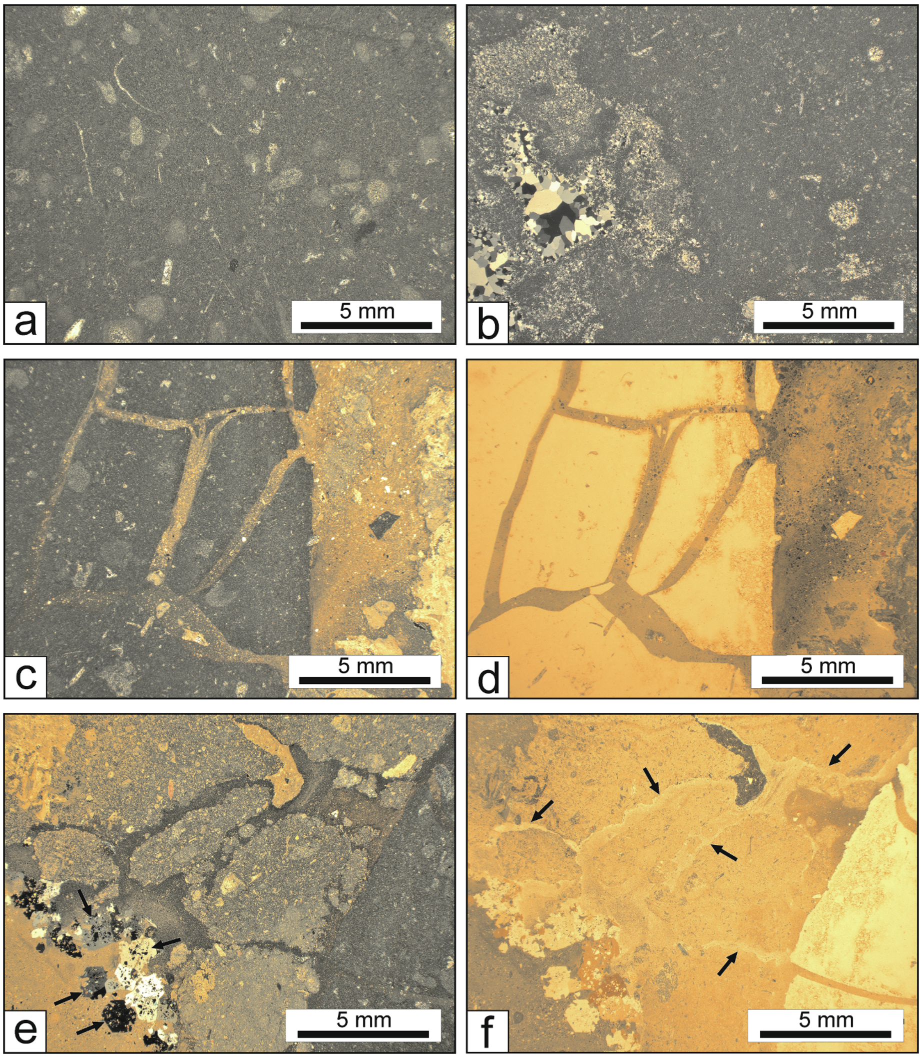

Fig. 5. (a) A typical microscopic image of chert concretion. Limestone is practically completely replaced by SiO_2. Fine brighter spots are fragments of bioclasts (mostly thin-shelled bivalves, brachiopods, bryozoans, fragments of echinoderms and sponge spicules) filled with microcrystalline quartz or fibrous SiO_2. Polarized transmitted light, crossed nicols, sample KD-1, Liban Quarry, Upper Oxfordian. (b) Chert concretion with a preserved siliceous sponge skeleton (left). In the center of the sponge skeleton, a geode filled with megaquartz crystals is visible. Polarized transmitted light, crossed nicols, sample KA-3, Piekary, Upper Oxfordian, (c) Fractured chert concretion in bedded limestone from a fault zone. Fractures are filled with partly silicified carbonate sediment. Partly silicified limestone is visible to the right of the chert concretion. Polarized transmitted light, crossed nicols, sample KB-5, Dąbrowa Szlachecka, Upper Oxfordian. (d) Chert concretion from Fig. 5c examined under non-polarized light. Practically, the whole sediment within the chert concretion is silicified but the deposit infilling the fractures and limestone at the contact with chert concretion (right) are less intensively silicified. (e) Chert concretion (far right) with epigenetic silicification in the interior (center). Non-silicified limestone is visible only in bottom left corner. Epigenetically silicified limestone shows dissolution seams. At the point of contact between the limestone and the epigenetically silicified zone, euhedral quartz crystals (arrows) are present, full of FI. Polarized transmitted light, crossed nicols, sample KB-7B, Dąbrowa Szlachecka, Upper Oxfordian. (f) Sediments shown in Fig. 5e examined under non-polarized light. The colour intensity corresponds to the intensity of silicification. Apart from chert concretion, intensive silicification proceeds along dissolution seams (arrow). The brownish part within the epigenetic silicification zone is filled with Fe-oxides.

(Fig. 3d). Similarly to chert concretions, bedded cherts show conchoidal fractures. No geodes filled with quartz were encountered.

6.2. Host rocks

The bedded cherts are hosted in calciturbidites which represent gravity flow deposits (Matyszkiewicz, 1996; Matyszkiewicz et al., 2015a). Calciturbidites are bedded and very well sorted detrital limestones (Fig. 3c) with graded bedding and well-visible Bouma sequences (Bouma, 1962; cf. Meischner, 1964; Matyszkiewicz, 1996). Bedded limestone located beneath the bedded cherts (T_a layer in Bouma sequence) is a packstone-grainstone whereas bedded limestone above the bedded chert is a wackestone grading up the sequence to mudstone (T_b and T_c layers in Bouma sequence). The main components of these limestones are fragments of planktonic echinoderms (*Saccocoma* sp.), the diameters of which decrease towards the top of the outcrop, and micropeloids, sponge spicules, and other small fauna species: benthic and planktonic foraminifers, holothurian sclerites, bivalve and brachiopod fragments (Fig. 6a–f). In the overlying mudstones and marls, calcareous spheres, coccoliths and radiolarians were identified (Matyszkiewicz, 1996).

6.3. Microscopic features

The bedded cherts occur in T_a and T_b layers of the Bouma sequence, which both reveal distinct normal graded bedding. The structure of the silicified limestone is well preserved, with primary depositional structures well visible, i.e. the directional arrangement of bioclasts. The cryptocrystalline SiO_2 and microcrystalline quartz replace both the fine-grained matrix and the bioclasts, mostly fragments of *Saccocoma* sp. and micropeloids (Fig. 6a-f). However, some micropeloids and fragments of echinoderms remain unsilicified. In the more coarse-grained layers of the Bouma sequence (T_a, partly also T_b) developed as grainstones-packstones, quartz aggregates are observed as the fillings of larger pore voids (about 0.2 mm across) and replacements for bioclasts. In very fine-grained sediments which were penetrated by burrows after deposition, the whole rock is subjected to silicification. This process is more intensive in burrowed portions of the sediments (Fig. 6e and f). Along the boundaries of concentric growth layers, complete silicification of sediments is observed in approximately 1-mm-thick zones (Fig. 6a–d), which totally obliterates the depositional structures. Occasionally, this boundary is highlighted by Fe-oxides (Fig. 6a and b).

6.4. Mineral composition and crystallinity index of SiO_2

The main component of bedded cherts is microcrystalline quartz, whereas pore voids are filled with megaquartz, particularly in the T_a layer of the Bouma sequence. Locally observed relic calcite is related to unsilicified micropeloids and fragments of echinoderms (Fig. 6c and d). Along the boundaries of growth layers, in zones with strongly obliterated primary depositional structures, the fibrous varieties of SiO_2 dominate. The CI values of bedded cherts which do not embrace

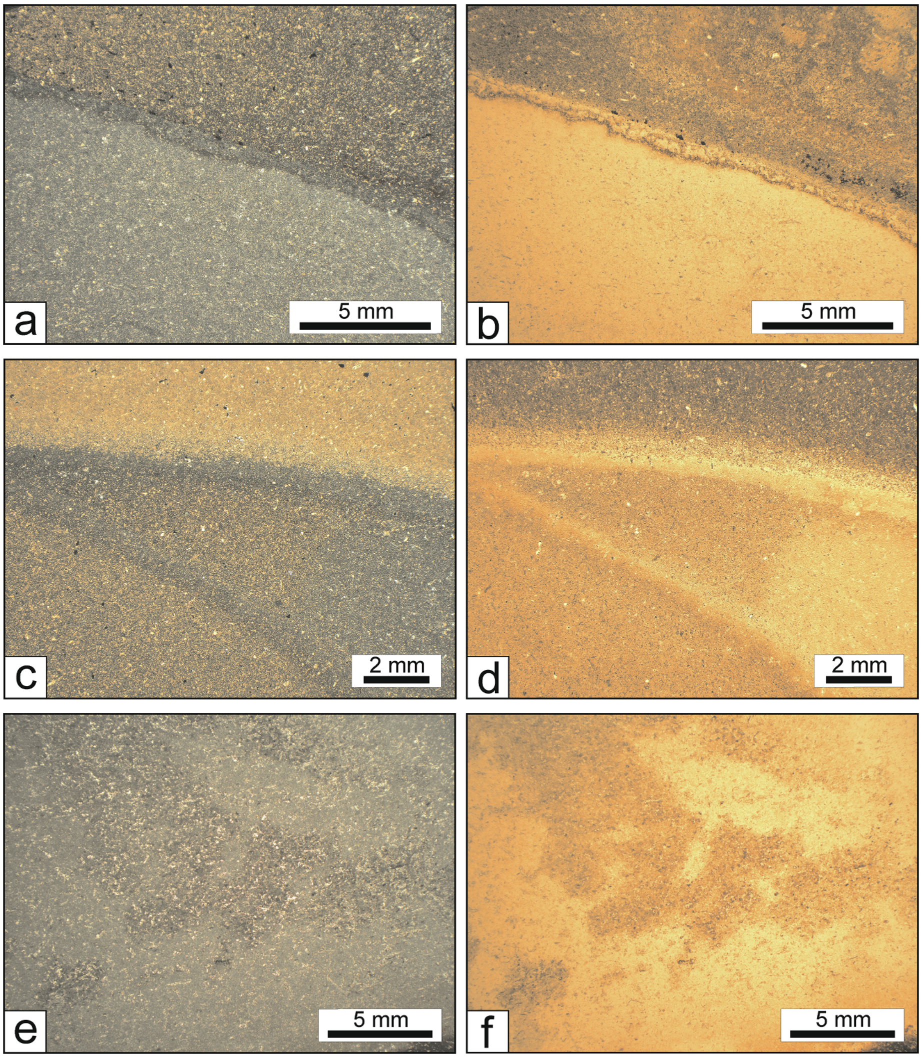

Fig. 6. (a) Bedded chert (bottom) at the contact with calciturbidite (top). The perfect sorting of limestone is evident. Fine, bright spots in limestone are fragments of planktonic crinoids *Saccocoma* sp. Polarized transmitted light, crossed nicols, sample SA-6, Ujazd, uppermost Oxfordian-lowermost Kimmeridgian. (b) Sediments from Fig. 6a examined under non-polarized light. The brownish lamina enriched in Fe-oxides is seen at the boundary of the bedded chert. (c) Contact between the bedded chert, having concentric growth layers (bottom and center), with non-silicified calciturbidite (top). Polarized transmitted light, crossed nicols, sample SB-1A, Sowiniec Horst, uppermost Oxfordian-lowermost Kimmeridgian. (d) Sediments from Fig. 6c examined under non-polarized light. Strong obliteration is evidence of the primary sedimentary structure of limestone at the boundaries of concentric growth layers. The diverse intensity of silicification is visible in the growth layers of the bedded chert. (e) Bedded chert in calciturbidite. The spotted structure results from the bioturbation of primary carbonate sediment. The primary sedimentary structure of limestone is only locally preserved. Polarized transmitted light, crossed nicols, sample SC-4, Wielkanoc Quarry, uppermost Oxfordian-lowermost Kimmeridgian. (f) Sediments shown in Fig. 6e examined under non-polarized light. Intensively yellow areas are bioturbated parts of sediment which have then been more intensively silicified.

occluded chert concretions vary from 5.1 to 7.1 (Table 2), which results from the domination of quartz in their mineral composition. In bedded cherts comprising chert concretions, CI values are distinctly lower (0.9–3.3; Table 2), which is an effect of the presence of moganite.

6.5. Genesis of cherts and plausible source of SiO_2

According to Matyszkiewicz (1996), the bedded cherts from calciturbidite sequences were caused by siliceous skeletons of radiolaria. This concept is consistent with the silicification model proposed by Bustillo and Ruiz-Ortiz (1987), which assumes the abrupt burial of the sea floor by calciturbidites, below which siliceous tests of radiolaria or skeletons of other organisms were deposited. This might have prevented the slow dissolution of the silica and its gradual migration into the sea water. Thus, the silica captured in the sediment might have precipitated in its most porous parts as bedded cherts.

In the top part of the calciturbidite sequence, single radiolaria tests were observed, but the significant thickness of the bedded chert layers suggests the contribution from other, more effective SiO_2 sources. Theoretically, these might have been skeletons or spicules of siliceous sponges but in the topmost part of the Upper Jurassic sequence from the southern KCU, siliceous sponges are rather rare (Matyszkiewicz et al., 2012). Additionally, the presence of concentric growth layers indicates multistage silicification instead of a single episode, although it does not preclude the participation of another external source of SiO_2, perhaps of an abiotic origin.

7. Epigenetic siliceous rocks

7.1. Macroscopic features

The ESR reveal a variety of shapes, sizes and colours (Matyszkiewicz, 1987; Rajchel, 1970; Kuźniar and Żelechowski, 1927). The ESR comprise: (i) concentric or irregular rims of silicified limestone growing on the cherts (Fig. 4e and f), (ii) irregular bodies embedded in limestones from the top part of the Upper Jurassic succession (Fig. 3e), immediately beneath the Upper Cretaceous sediments, (iii) coatings growing on fault surfaces or on the Cenomanian abrasion surface, cutting the Upper Jurassic strata and (iv) loose fragments of silicified limestones embedded within the regolith developed on the Upper Jurassic sediments (Fig. 3f).

The concentric rims of silicified limestone growing on the cherts (i) are up to about 5 cm thick. The colours of their weathered surface are dark-grey or dark-brown, which stems from their greater porosity in comparison with chert concretions which are brighter. The colour of the fresh fractures is even brighter (Fig. 4f). The contact with chert is sharp, whereas the outer surface is irregular (Fig. 4e) and comprises numerous protrusions several-millimetres-long penetrating the surrounding limestone. The irregular bodies embedded within the limestones (ii) have dark-brownish, dark-grey, brownish or cherry-red colours. They form irregular nests, from about a dozen to several dozen centimetres in diameter (Fig. 3e). Some such nests contain cherts and/or limestone fragments. Weathered nests are highly porous, with pore walls covered

Table 1
Values of crystallinity index CI (after Murata and Norman, 1976) and main SiO_2 phases in chert concretions from the southern KCU.

Sample	Locality	CI index	SiO_2 polymorphs	Host rock
KA-2	Piekary*	0.4	fibrous varieties,	bedded limestones
KA-5		1.1	moganite,	
KA-6		0.4	quartz	
KB-1	Dąbrowa	1.2		indistinctly bedded limestones and debris flows at huge fault zone
KB-8	Szlachecka**	3.5		
KB-10		3.3		
KC-2	Tyniec***	4.5		bedded limestones at huge fault zone
KC-4		5.4	quartz	
KC-5		2.4	fibrous varieties,	
KD-1	Liban***	1.9	moganite,	bedded limestones
KD-2		0.5	quartz	
SA-2A	Ujazd****	1.7		within a layer of bedded chert in calciturbidites
SA-3		2.3		beneath a layer of bedded chert in calciturbidites
SA-7		0.6		above a layer of bedded chert in calciturbidites
SB-1C	Sikornik*****	3.3		within a layer of bedded chert
SC-1B	Wielkanoc*****	0.9		

Detailed descriptions of outcrops and/or mineralogical data after: * Alexandrowicz (1955; 1960); Matyszkiewicz (1989), ** Sitarz et al. (2014), *** this paper, **** Matyszkiewicz, 1996; Matyszkiewicz et al., 2015a; Matyszkiewicz and Olszewska, 2007, ***** Matyszkiewicz (1996).

Table 2
Values of crystallinity index CI (after Murata and Norman, 1976) and main SiO_2 phases of bedded cherts from the southern KCU.

Sample	Locality	CI index	SiO_2 polymorph	Host rock
archival data	Ujazd*	5.1	quartz	calciturbidites
SB-1B	Sikornik**	5.6		
SB-2		6.3		
SC-2	Wielkanoc**	6.3		
SC-4		7.1		

Detailed descriptions of outcrops and/or mineralogical data after: * Świerczewska (1997), ** Matyszkiewicz (1996).

with crystalline quartz. Quartz aggregates up to several millimetres in size are also visible. The outer surfaces of the ESR bodies are very rough, which presumably results from the dissolution of the host limestone during weathering. The crusts growing onto the fault surfaces and onto the abrasion surface (iii) are up to several centimetres thick and are preserved as sheets not exceeding an area of 1 square metre. These accumulations are brownish or dark-grey and highly porous. The regolith (iv), which covers the top surface of the Upper Jurassic sediments contains fragments of completely silicified Upper Jurassic limestones, up to several dozen centimetres across, showing brownish, dark-grey or sometimes cherry-red colours. Similarly to the other ESR types, such limestone fragments are highly porous but their edges are usually rounded. The fragments may contain macroscopic-size limestone pieces (Fig. 3f).

7.2. Host rock

The limestones which host the ESR nests or whose surface is covered by siliceous crusts reveal a considerable diversity in term of facies. These may be bedded limestones, with chert concretions developed as wackestones-packstones-grainstones, or massive facies developed as boundstones. In the bedded facies, the ESR usually form rims overgrowing the chert concretions or nests embedded within the limestone. In the massive facies, however, they occur as crusts growing on the upper limestone surface, from several to a dozen centimetres thick. If the bedded or the massive limestones are cut by faults, or if they are worn down by abrasion, the ESR are preserved as sheets, some dozens of centimetres in diameter (Matyszkiewicz, 1987).

7.3. Microscopic features

The ESR are dominated by micro- and megaquartz, which crosscuts the primary sedimentary structures during their replacement with SiO_2. In contrast to chert concretions and bedded cherts, the boundaries between the ESR and limestones are not sharp (Fig. 7a–f). Silica selectively penetrates the surrounding limestone, replacing the fine-crystalline matrix first. Sometimes, this penetration proceeds along the stylolites (Fig. 5e and f). Fine pores in the limestones are filled by microcrystalline quartz and megaquartz, which also precipitate on the walls of larger pores and replaces calcite cement in larger bioclasts. Locally, numerous euhedral quartz crystals are observed, over 1 mm in size, full of fluid inclusions (FI) (Fig. 5e and f). Temperature determinations revealed the formation of a FI well of about 100 °C (Matyszkiewicz, 1987).

7.4. Mineral composition and crystallinity index of SiO_2

The ESR are dominated by quartz with a highly variable crystal size: from submicroscopic to large, euhedral specimens of over 1 mm in diameter. In comparison with both the chert concretions and the bedded cherts, quartz from the ESR shows remarkably higher CI values (over 9) (Matyszkiewicz, 1987). Locally, particularly in nests or crusts deposited in the joint sets or fault surfaces (Fig. 5e) as well as on abrasion surfaces and loose fragments embedded within the regolith (Fig. 5f), several percent of Fe-oxides may occur (Matyszkiewicz, 1987).

7.5. Genesis of ESR and plausible source of SiO_2

The high CI values, together with the high FI formation temperatures in quartz, clearly indicates a hydrothermal source of SiO_2 (Matyszkiewicz, 1987). This conclusion is supported by the FI temperatures of approximately 100 °C measured in quartz aggregates from the neptunian dykes penetrating the Oxfordian limestones (Matyszkiewicz et al., 2016). Therefore, this silicification can unambiguously be linked to extensional tectonics, which were active in the southern KCU from at least the Late Jurassic period.

8. Possible identification of the origin of siliceous raw materials used in the manufacture of stone tools − Conclusions

8.1. Chert concretions

The principal constraint affecting the identification of siliceous raw

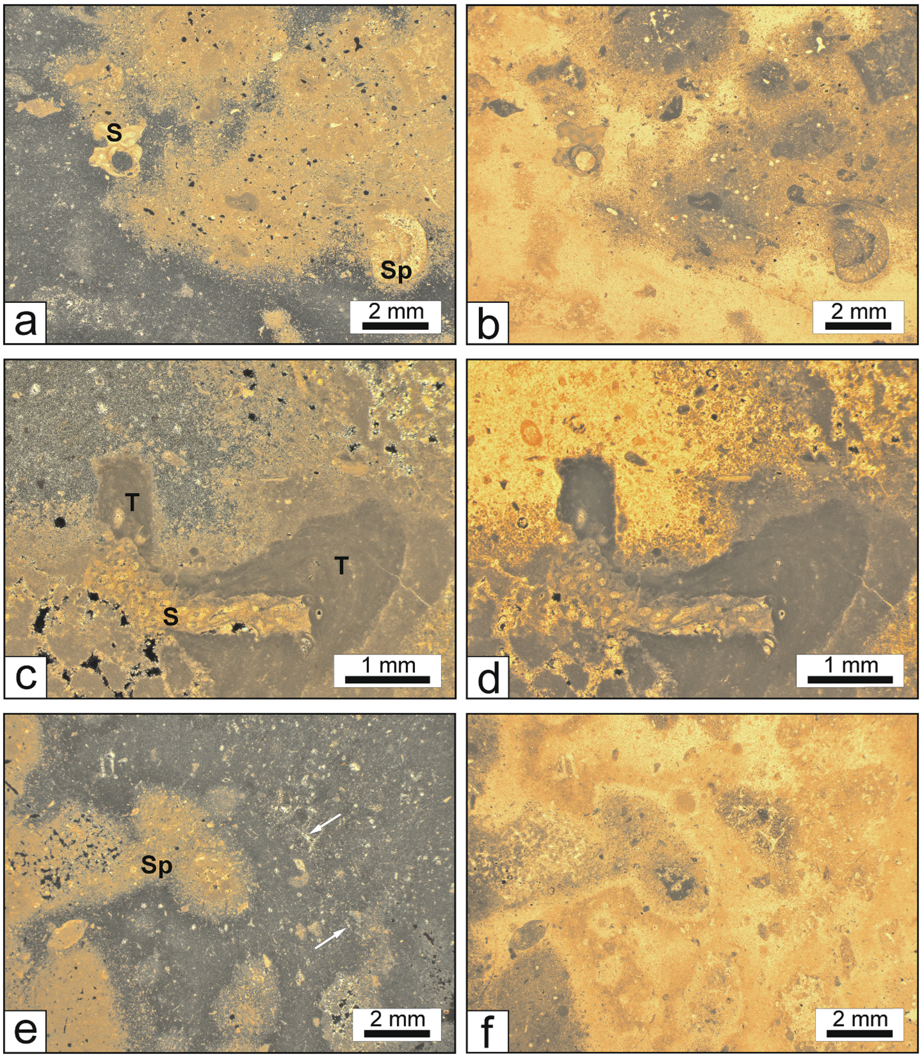

Fig. 7. (a) The irregular contact of the epigenetic siliceous crust (left and bottom) with limestone. Silicification selectively penetrates limestone in which single, larger bioclasts are visible (Sp – calcified siliceous sponge, S – serpula fragment). Polarized, transmitted light, crossed nicols, sample KES-1, Pychowice, Cretaceous abrasion surface developed on Upper Oxfordian massive limestone. (b) Sediments shown in Fig. 7a were examined under non-polarized light. The intensity of colour reflects the intensity of silicification. Numerous bright spots in the center are primary pores of limestones unfilled with SiO_2. (c) Contact of epigenetically silicified nest of (upper left) with limestone. Silicification selectively affects mostly the carbonate matrix. In the center, non-silicified microbial structures are visible of thrombolite type (T) growing onto serpules colony (S). Polarized transmitted light, crossed nicols, sample KES-3, Skały Twardowskiego. Upper Oxfordian. (d) Sediments shown in Fig. 7c examined under the non-polarized light. Intensively yellow areas reflect epigenetic silicification. The walls of the fine pores are covered with quartz crystals (upper right). (e) Irregular contact of epigenetic silicification zone (right side and center) with limestone. Selective silicification of limestone is visible together with a fragment of calcified porous skeleton of siliceous sponge (left, Sp). Within the silicified zone, single spicules of siliceous sponges are visible (arrows) filled with granular quartz. Polarized transmitted light, crossed nicols, sample KES-4, Skały Twardowskiego. Upper Oxfordian. (f) Sediments shown in Fig. 7e examined under the non-polarized light. The intensity of the yellow colour reflects the intensity of the silicification. Edges of larger bioclasts (mostly fragments of siliceous sponges) are surrounded by intensively yellow rims, in which the primary sedimentary structure of limestone is completely obliterated.

materials used in the manufacture of stone tools is the relatively limited volume of the study materials available (cf. Bustillo et al., 2009). The protoliths of the chert concretions, which were mostly bedded limestones, reveal a high degree of non-homogeneity which decisively influenced the development of concretions.

Regretfully, colour, which appears to be the principal criterion of archaeological categorization systems, is rather meaningless from the geological point of view as the chert colours result from local pigmentation with various compounds, e.g. finely disseminated pyrite, Fe-hydroxides and/or organic matter. The variability of colours is observed not only in particular outcrops but even within individual chert concretions (Fig. 4b). The results of the microfacial analysis of the Upper Jurassic limestones from the southern KCU suggest that the utmost caution is required when the skeletons of siliceous sponges are proposed as the main or even the only source of SiO_2 in the cherts. Instead, the seepages of hydrothermal fluids triggered by extensional tectonic events must be taken into consideration, which far extends the possible inventory of substances which might have coloured the chert concretions.

It is highly probable that the macroscopic observations will allow us to distinguish between tools manufactured from chert concretions and those from bedded cherts. The principal criterion must be the presence of limestone relics, i.e. the bioclasts of silicified microfauna, several millimetres across, or their fragments, and the intraclasts. However, this criterion does not apply to chert concretions hosted in calciturbidite sequences, which resulted from the silicification of homogenous sediment and thus they do not contain macroscopic relics of limestones and fossils.

The attempts to link the chert concretions found at archaeological sites with particular outcrops or at least particular regions (e.g. northern, central or southern parts of the KCU) based upon analysis of colours, shapes, sizes, etc. are, unfortunately, groundless from the geological point of view. The chert concretions do not reveal any specific features which would enable us to indicate their source outcrop or even only the source stratigraphic interval of the Upper Jurassic succession, especially over the full range of the occurrence of microbial-sponge megafacies in Europe. Only cherts embedded within calciturbidite layers, if correctly identified, may undoubtedly indicate the topmost part of the Upper Jurassic succession from the southern KCU.

More suitable information for the indication of chert sources is provided by microfacial analysis combined with X-ray diffractometry. These tools enable us to identify chert concretions almost unambiguously using the character of the limestone silicification process. The principal feature of chert concretions is their non-homogenous structure with bioclasts and/or their fragments preserved as carbonate relics or filled with granular quartz. However, this criterion does not apply to chert concretions from calciturbidite layers which, as quoted above, originated from the silicification of homogenous sediments.

The results of the X-ray diffraction analyses of chert concretions document the presence of quartz and moganite, the latter being only a trace component of bedded cherts and which is absent from the ESR. Also good indicators are low CI values of quartz.

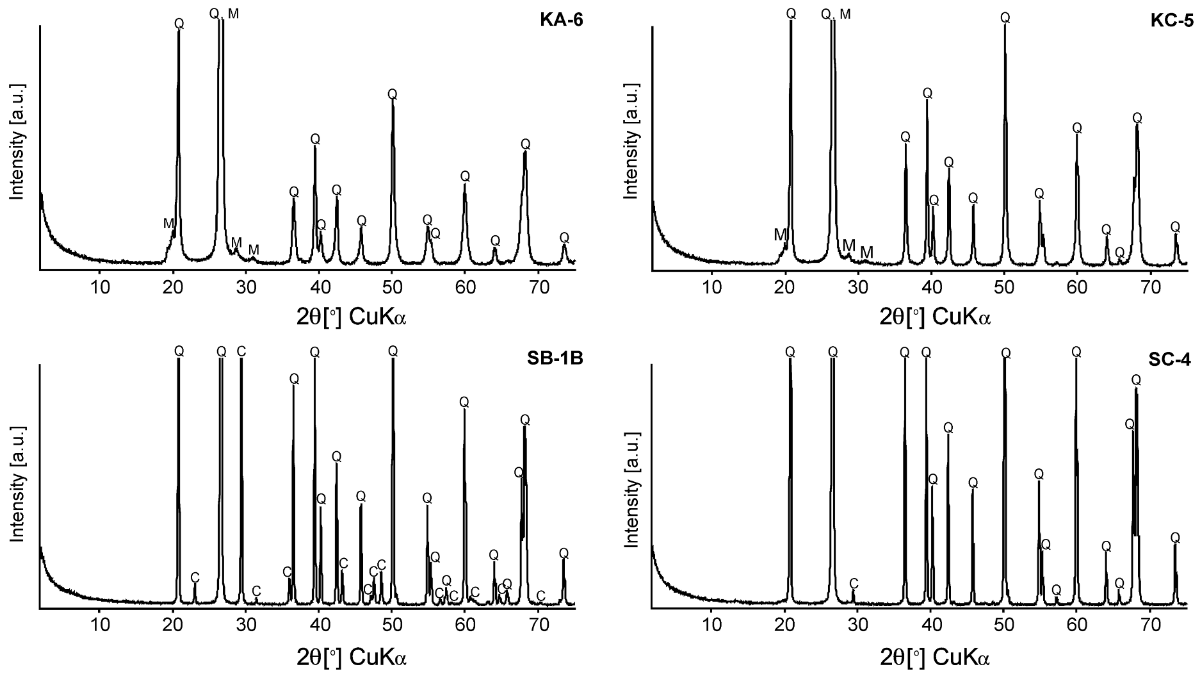

Fig. 8. Examples of X-ray diffractograms of chert concretions (samples KA-6 and KC-5) and bedded cherts (samples SB-1B and SC-4). Sample symbols, locations and supplementary data as in Tables 1 and 2; Q – quartz, M – moganite, C – calcite.

8.2. Bedded cherts

The macroscopic identification of stone tools manufactured from bedded cherts is primarily possible due to the significant homogeneity of the silicified limestone protoliths and the presence of characteristic concentric growth layers. Moreover, in the bedded cherts, no relics of non-silicified limestones have ever been observed and the silicified macrofauna is also practically absent.

Microfacial analysis enabled the researchers to unambiguously identify bedded cherts, which typically reveal a perfect sorting of primary carbonate sediment. Moreover, the directional arrangement of bioclasts should be noticed and graded bedding cannot be precluded, being particularly well-visible in larger tools. It must be emphasized that only chert concretions from calciturbidites display a similar development.

The results of X-ray diffraction analyses reveal the presence of quartz and only insignificant amounts of moganite, but CI values are distinctly higher than those from chert concretions and significantly lower than those from the ESR.

8.3. Epigenetic siliceous rocks

The tools manufactured from ESR rocks should be easy to distinguish. The principal petrographic feature of this raw material is the very well-preserved primary sedimentary structure of silicified limestone, which is an effect of its selective replacement. Silicification is much easier in a fine-crystalline matrix whereas the larger components (bioclasts, intraclasts, microbial structures) is more resistant to replacement. Moreover, the ESR are highly porous, which is partly inherited from the primary, porous limestones and partly produced by the dissolution of minute calcareous enclaves during karstification.

The microfacial analysis enables the researchers to unequivocally identify the raw materials based upon the character of the chert/limestone boundary. In contrast to chert concretions and bedded cherts, this boundary in the ESR is not sharp but rather irregular due to the selective nature of limestone replacement by SiO_2. If the chert/limestone contact cannot be observed, an identification criterion may be the large number of non-silicified limestone enclaves within the ESR and the simultaneous presence of abundant fragments built of quartz rich in FI. In contrast to both the chert concretions and the bedded cherts, epigenetic silicification also affects the stylolites, which clearly argues for the late-diagenetic provenance of the replacement processes.

In terms of mineral composition, the ESR are composed exclusively of quartz, as confirmed by the CI values which are significantly higher than those in both chert concretions and bedded cherts.

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CRediT authorship contribution statement

Alicja Kochman: Conceptualization, Methodology, Formal analysis, Investigation, Resources, Writing - original draft, Writing - review & editing, Visualization, Supervision, Project administration, Funding acquisition. **Jacek Matyszkiewicz:** Conceptualization, Methodology, Formal analysis, Investigation, Resources, Writing - original draft, Supervision, Project administration, Funding acquisition. **Michał Wasilewski:** Resources.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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