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Abstract

At the end of the 19th century, between 1891 and 1893 the young Hungarian archaeologist Samuel Fenichel collected 315 polished stone implements (42 with wooden handle) in Astrolabe Bay, Northeast New Guinea. Because of his death in 1893, his work and his collection became unfinished. His compatriot, Lajos Bíró decided to carry on with his researches, previously on the same area, after on other parts of the island. He worked in New Guinea from 1896 until 1901. The stone implements disappeared in the meantime, so he didn't collect so many objects as his predecessor, but - while Fenichel hadn't any notices about the objects, he recorded all about the craftsmanship of stone adzes. The two collections together offer a unique opportunity to study the making and use of stone axes and adzes at the moment of their "transformation". The collection was never published: this paper is an attempt to present it.

Kivonat

A 19 század végén, 1891 és 1893 között a fiatal régész és néprajzkutató, Fenichel Sámuel 315 csiszolt kőeszközt (közülük 42 db nyéllel) gyűjtött az Astrolabe öbölben, Északkelet Új-Guineában. 1893-ban bekövetkezett halála miatt munkája és a gyűjtemény összeállítása befejezetlen maradt. Honfitársa, Bíró Lajos szánta el magát kutatásainak folytatására, előbb ugyanazon a területen, majd a sziget más vidékein. 1896-tól 1901-ig dolgozott Új-Guineában. Időközben a kőeszközök eltűntek, így ő már nem tudott annyi tárgyat összegyűjteni, mint elődje, de – míg Fenichel nem készített jegyzeteket a gyűjteményhez, addig Bíró szinte minden adatot feljegyzett a kőeszközökkel kapcsolatos mesterségbeli tudásról. A két gyűjtemény együtt egyedülálló lehetőséget nyújt a kőbalták és kő szalukapák készítéséről és használatáról abban a pillanatban, amikor felváltották őket a fémeszközök. A gyűjtemény máig közöletlen: jelen beszámoló célja az anyag bemutatása.

Keywords: New Guinea, polished stone axe/adze, ethnographical collection, Sámuel Fenichel, Lajos Bíró

KULCSSZAVAK: ÚJ-GUINEA, CSISZOLT KŐESZKÖZ, NÉPRAJZI GYŰJTEMÉNY, FENICHEL SÁMUEL, BÍRÓ LAJOS

The Museum of Ethnography in Budapest possesses a relatively large number of polished stone tools from North-East New Guinea collected more than hundred years ago. The material was never studied thoroughly and is unpublished until even today. The aim of this paper is to make experts acquainted with this material.

The story of the collection is as follows: at the end of the 19th century, between 1891 and 1893 young Hungarian archaeologist Samuel Fenichel (1863-1893) succeeded to reach North-East New Guinea, at that time part of the German colonial empire, under the name of Kaiser-Wilhelmsland.

Fenichel participated as preparator in the ornithological expedition organised and financed by a German agronomist, Albert Grubauer, who wanted to collect birds of paradise.

Grubauer, shortly after arriving to New Guinea, became ill, and returned to Germany, leaving Fenichel behind, without any financial assistance. Fenichel decided to stay in New Guinea, and he offered his services to the Hungarian National Museum, Department of Ethnography. The Museum accepted it and asked to turn his attention over the ethnographical material in the region and helped him to stabilise his financial situation, too.

Fenichel's working area was the Astrolabe Bay, where he collected more than 2000 ethnographical objects in the last 15 months of his life; among them, 315 polished and partly hafted (42 pieces) stone tools, too.

By this work he saved the stone tools used by the local people in the very last minute before the steel tools reached the coast.



Fig. 1.: Research areas of Samuel Fenichel and Lajos Bíró, New Guinea, North-eastern coast

1. ábra: Fenichel Sámuel és Bíró Lajos gyűjtési területei, ÉK-Új-Guinea partvidékén

After his death the Hungarian scholar, Lajos Bíró (1856-1931), who was mainly interested in natural history (entomology) undertook Fenichel's mission. He arrived in the Astrolabe Bay in 1896 and he worked in several parts of New Guinea until 1901.

In his extensive collection of ethnographical pieces (somewhat more than 5500 objects) there were only 30 polished stone tools (21 of them hafted) - because of the extremely rapid change from stone to steel. (Fig. 1.)

In contrast to Fenichel, who had very few notes about the objects, Bíró accompanied every type of objects and often some individual or special pieces with the most detailed descriptions ever made in this time in ethnography. His remarks, his inquiries about the manufacturing and using the stone tools can guide us to study the material of Fenichel, too.

The investigated area – Astrolabe Bay and the coastal region around it – is one of the earliest parts of New Guinea contacted by Europeans.

After the discovery of the island and the mapping of the coast between the 16th and 19th centuries, the colonization was started by different peoples. The first person who was interested in native population of North-East New Guinea is the Russian Nicholai Nicholaievich Mikluho-Maklay. He used to spend fifteen months there, between 1871 and 1876, mainly in the Astrolabe Bay and nearby. The German zoologist, Otto Finsch (1839-1917) was the next to visit the area, between 1884 and 1885: his publications helped Bíró in his researches.

At the time of the stay of Fenichel the stone tools were still in use, but three years later, when Bíró arrived, they were already out of use. He wrote: "I have not had the opportunity to see anyone using a stone axe. There are not any axes to be bought in Friedrich Wilhelmshafen or its vicinity. In Berlinhafen, similarly, there are not any." (Molnár-Bagley, E., 1993: 146)

The first polished stone blade he succeeded to collect at Bogadjim only after 8 months.

Beside the cultural and technological changes the lack of stone tools was due, as Bíró saw the situation, to the collectors (museum and private people) too, who, recognizing the importance of the ethnographical objects, as witnesses of the disappearing local culture, made every effort to collect them in large quantities. Bíró remarked the new trend, and he tells us, that around the Astrolabe Bay they can find no more stone tools because the people gathered all pieces to put on the market being much in demand by the Europeans.

Bíró could not collect any relevant information about the origin of the stone blades the local people had no longer any idea of them. They said to him, that the stone blades came from somewhere in the inland and they reached the coast by trading routes. As Bíró supposed, after the different types of rocks, the raw material must have been originated from different sources.

The interior of the island was discovered successively only after the visit of Fenichel and Bíró: the peoples living in the Highlands began to be in contact with the white people from the middle of the 1930s onwards.

From the Astrolabe Bay coast towards the inland we can find mainly volcanic and sedimentary rock types. The Highlands became known after the first patrol reports (1930: J. Taylor and M. Leahy, , 1939: L. G. Vial, Patrol Officer and L. C. Noakes, geologist, , 1943: J. A. Costelloe, Patrol Officer,) or by systematic investigations of scholars, studying this subject, like B. Blackwood, 1936-37 and J. Chappell, 1963-65 among others.

In the area of the Mount Hagen on the West, the Mount Michael on the East as well as between the Purari River on the South and the Ramu River on the North there are some important stone quarries known today.

Beside the smaller ones, the main quarries like Ganz-Tsenga, Dom, Abiamp or Kafetu supplied the mentioned area with raw material and stone blades which arrived by trade routes with all probability to the Astrolabe Bay as well.

Ian Hughes wrote in his work of capital importance about New Guinea Stone Age trade: "...polished stone blades of Central Highlands type appear in the early collections made around Astrolabe Bay." (Hughes, I., 1977, Vol.II.: 73.)

Chappell (1966) described the Central Highlands stone quarries and analysed their petrographical differences also. Because of the lack of any petrographical analyses in the material from Astrolabe Bay collected by Fenichel and Bíró until today, we had the possibility to establish only typological series – which is by far not enough for an appropriate study.

One of the aims of this paper is to find enthusiastic experts interested in different modern analyses on our pieces.

After the types of stone blades coming from different stone quarries it is very possible that about 60 blades originated from Central Highlands – mainly from Abiamp and Ganz-Tsenga, but, naturally, we cannot take it for certain on typological basis alone.

The blades in Fenichel's collection are in almost every case complete pieces – so we can study them after their formal characteristics and measures.

The length of the smallest ones (32 pieces) are between 4 and 6 cm, their width is between 3.4 - 5.2 cm. The medium size (258 pieces) is 6 - 14 cm x 3.6 - 7.2 cm, and the big ones (21 pieces) measures 14 - 22 cm x 5.9 - 7.8 cm. The biggest (1 piece) is 30 cm in length and 9.8 cm wide. (Width measured by the edge).

Their thickness alters between 0.7 - 3.4 cm as extreme data (0.7 - 0.9 cm for the smallest and 3.2 - 3.4 cm for the medium (biggest). For all the others vary between 1 - 2 cm, respectively, 2 - 3 cm sharing in proportion.

The blades, after their forms can be range into 9 clearly separate types, but from them, 4 types are represented only by 1 -1 examples. We can assign about 60 blades into 3 - 4 types: they are polished all over their surfaces, including their edges and traces of use are visible only on a few pieces.

The surfaces of the other blades are mainly polished only on the working edges, but in a small degree, only where it was really necessary, they aspire to make disappear the uneven parts of the blade.

I chose for a random test 70 blades to examine by magnifying glass (30x): on 32 pieces there were traces or rests of light brown, red or yellowish brown soil paint. On the surface of 3 blades I found resin-like material and some rests of fibres in four cases: they were with one exception together on the same object.

As Bíró wrote about his collection from Berlinhafen, the red paint or colour which can be observed mainly on the objects of wood, carvings and stone axes served probably protect them against insects and against crack. (Bíró, L. 1899: 34)

Because of the usefulness of the blade depends on its stability in the hafting, so they put a piece of *tapa* (bark-cloth) in the blade-holding socket or/and stick soil into it around the stone blade. (Bíró, L. & 1899: 36)

The collection consists of 42 hafted tools. In two examples there are shell blades and in two instances steel blades inserted instead of stone. In one case we have only the wooden socket, in five more, only the handle. All items represent the 2nd type of Crosby's hafting traditions: T-shaped haft with split sockets lashed to the head of the haft. (Crosby, E., 1977).

It is an important aspect for analysis what is the orientation of the blade or, more exactly, if the edge of the stone blade is parallel to the handle (axe) or perpendicular to it (adze).



Fig. 2.: Stone headed adzes and stone blades from the collection of Bíró (1) and Fenichel (2-4) Archive drawings in the Ethnographical Museum (hafted adzes) and J. Antoni (blades)

2. ábra: Nyelezett szalukapák és kőpengék Bíró (1) és Fenichel (2-4) gyűjtéséből. Eredeti rajzok a Néprajzi Múzeum leíró kartonjairól (nyelezett eszközök) és a pengékről (Antoni J. rajzai)

In the collection of Fenichel one can determine the situation with certainty only in 26 cases. They are preserved in their original condition, while the others are partially damaged: their blades fell out, sometimes lost, or their blades were set in by someone in the Museum during the last 100 years. (Fig. 2.)

The axe of the blade-edge was never parallel with the axe of the haft, but in 14 cases they were perpendicular to it and in 12 cases they stood obliquely. This latter is not by chance: in the collection of Bíró from Berlinhafen all the three possibilities occur and the local people had separate name for each position. (Figs. 3-4.)

The hafts are carved mainly from hard wood and their head, with the blade in the socket forms angles with the hafts of between 80-90 degrees.

The length of the haft is in general 55 cm, twice the length of the head. The measurements of the smallest tools are: length of the haft: 25 cm, length

of the head: 15 cm while for the biggest are: haft: 87 cm, head: 32 cm.

We can observe red colour on the surface of the hafts, too.

Bíró arrived in the last minute, when he still had the possibility to interrogate the old people about the names of different parts of stone tools. He collected all local words for craftsmanship in every village wherever he visited the area with all explications given by the people about their meaning.

He could not observe the manufacturing of the stone tools any more, but he had the chance to learn some information about the subject. The situation was similar although somewhat better in the case of the use of stone tools.

Bíró mentioned that they are different types of stone tools according to different use. He wrote:

"The natives... differentiate among the various kinds of stone axes by giving them different names. Different parts of the axes also have specific names.



Fig. 3.: Polished stone blades, hafted blades (as adzes) and wooden haft. Collection Bíró. Astrolabe Bay. Drawings by I. Nécsey (in: Bíró, 1901, p. 82, Fig.40.)

3.:ábra: Csiszolt kőpengék, nyelezett pengék és fa nyél Bíró L. gyűjtéséből az Astrolabe-öbölből. Nécsey I. rajzai (Bíró, 1901, p. 82, Fig.40.)



Fig. 4.: Stone-headed axes and adzes, shell-headed adze (4) and wooden socket with shell blade (7) from the collection of Bíró, Berlinhafen. Drawings by A. Richter (in: Bíró, 1899, Plate VIII.)

4. ábra: Kőpengével ellátott balták és szalukapák, kagylópengés szalukapa (4) és fa foglalat kagylópengével (7) Bíró L. gyűjtéséből (Berlinhafen). Richter A. rajzai (Bíró, 1899, VIII. tábla)

The types of axes are differentiated not so much on the basis of the type of stone or size of stone, the differentiation is based primarily on the stone's sharpness and positioning. This is certainly because these are things that determine the tools' use." (Molnár-Bagley, E., 1993: 146).

He remarked also that the stone tools in general were useful for all kinds of work, like the penknife: it was not a rarity to see someone trimming his finger-nails with them.

He organised a competition to know how much time was necessary to cut a tree using stone and steel tool: with stone bladed tool needed approximately three-times more than with a steel axe.

Biró remarked that the stone blade was not in circulation with sharpened edge: their owners sharpened them themselves and therefore they had everywhere a little grinding-stone in their bag.

About the secondary use of stone blades Bíró mentioned that the blades, being used up or because

of the continuous grinding became too small or damaged – useless in their original form – could survive as reamer, hammerstone or anvil, for example to produce little shell-discs or pearls, as we can see on some pieces collected by him.

The contacts with traders, missionaries, discoverers, colonizers and representatives of all sorts of sciences resulted in the successive disappearing of the ancient culture. In Papua New Guinea, which was born from former colonies as independent state, we cannot find any more people using stone age technology. Today the old traditions, objects of everyday life or objects of tribal art serve as attractions for tourists. However, in the western half of the island, actually part of Indonesia (Irian Jaya) about 30 years ago the scholars had the real chance to observe some people in manufacturing and using stone tools.

The Central Highlands of Irian Jaya remained relatively less frequented and the peoples living here had no direct contacts with strangers before 1961 or some groups even only 10-20 years later. The publications of ethnologists or archaeologists, who came to study the region and its peoples – with few exceptions – concentrated primarily on questions of sociology or art and less on technology.

After the first visits of Fenichel and Bíró on the eastern part and other scholars on the western part of New Guinea it was a great deal – not hundreds but several thousands – of stone tools exported from the island to museums (less in number) and to private people (much more pieces) all over the world. As Bíró bitterly remarked : "Objects are transported to museums but they lose the spirit which gives them life." (Molnár-Bagley, 1993, p.177: translated from Bodrogi, 1987, p.178). For the pieces taken away by private people the situation is even worse: they are probably lost for ever.

Pierre and Anne-Marie Pétrequin studied the stone tools first in their complexity, embedded in their natural environment and social context, following the steps of manufacturing technology "from the rock to the stone axe".

In their fundamental work (Pétrequin. P., -Pétrequin, A.-M., 1993) they succeeded to realize what was the most important for Bíró and for me, too: to give life to the "dead" objects...

I hope, with the help of experts in geology and petroarchaeology we can arrive at that point in our museum collection material also.

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RESULTS OF NON-DESTRUCTIVE SEM-EDX AND PGAA ANALYSES OF JADE AND ECLOGITE POLISHED STONE TOOLS IN HUNGARY MAGYARORSZÁGI JADE ÉS EKLOGIT NYERSANYAGÚ CSISZOLT KŐESZKÖZÖK RONCSOLÁSMENTES VIZSGÁLATÁNAK EREDMÉNYEI ZSOLT BENDŐ¹; GYÖRGY SZAKMÁNY¹; ZSOLT KASZTOVSZKY²; BOGLÁRKA MARÓTI²; SZANDRA SZILÁGYI²; VERONIKA SZILÁGYI²; KATALIN T. BIRÓ³

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Abstract

Good quality high pressure (HP) metaophiolites suitable for making stone implements, like jade and eclogite, are geologically absent in the Carpathian Basin and its surroundings. Therefore this raw material type was unknown among Hungarian findings for a long time, and henceforward this is one of the rarest types of raw material of polished stone implements in Hungary. Their investigation is very important because of their scarcity and distant origin. The nearest geological locality where these raw materials can be found is over 1000 kms away. The specific form and integrity of these stone implements indicate that they were transported as complete (finished) artefacts.

Petrological investigations of large collections (Miháldy and Ebenhöch collections) revealed their presence among Hungarian findings (Friedel et al. 2008, Szakmány et al. 2013). In 2013 and 2014, several new pieces of HP stone implements were found in museums of Hungary, most of them from known archaeological context. Most samples were found in Transdanubia, only one piece turned up in Eastern Hungary (**Fig. 3**.). The 7 pieces of known archaeological context are from four localities (Zengővárkony (3), Szombathely Oladi plató (2), Alsónyék (1), Gorzsa (1)). Other pieces are stray finds. The localities and the main features of the tools are summarized in **Table 1**. In addition to the 3 pieces presented in Szakmány et al. 2013, 10 pieces of stone implements made of HP metaophiolites are presented in this work (**Fig. 1**.).

Only non-destructive analytical methods were used in this study. Stone implements were divided into raw material type groups based on their mineral chemical and bulk rock analytical data. Our results are corresponding to results on HP metaophiolites of North-western Italy, obtained both on geological and archaeological samples (D'Amico et al. 2003, D'Amico 2012, Pétrequin et al. 2012). Based upon these facts, the HP metaophiolite stone implements in Hungary probably originated from the same raw material sources as Italian (and other Western European) HP metaophiolite stone tools. According to technical literature, these primary sources can be the Monviso, the Voltri Massif and secondary in the resedimented Oligocene conglomerates in Quaternary of River Po, Staffora and Curone equally (D'Amico et al. 2003, D'Amico & Starnini 2006, 2012, Pétrequin et al. 2012).

Kivonat

Jelenlegi ismereteink szerint a nagynyomású (HP) metaofiolit nyersanyagú eszközök igen csekély számban találhatók meg a magyarországi csiszolt kőeszközök leletegyütteseiben. Vizsgálatuk azért kiemelkedő fontosságú, mert nyersanyaguk a Kárpát-medencében, és annak közvetlen környezetében nem található meg. Legközelebbi, a neolitikumban bizonyítottan felhasznált nyersanyagforrásuk Magyarország jelenlegi területétől több, mint 1000 km távolságban található, ami, az eszközök formavilágát is figyelembe véve, arra utal, hogy ezek az eszközök kereskedelmi útvonalakon, használatra kész termékként érkeztek régészeti lelőhelyükre.

Magyarországi jelenlétükre a Miháldy- és az Ebenhöch-gyűjtemények kőzettani szempontú feldolgozása során derült fény (Friedel et al. 2008, Szakmány et al. 2013). 2013-ban és 2014-ben további leletek kerültek elő különböző múzeumok gyűjteményeiből, a JADE2 program keretében végzett szisztematikus kutatások eredményeképpen. A leletek legnagyobb része a Dunántúlról került elő, mindössze egy darab származik az ország keleti részéből (**3. ábra**). Hét darab kőeszköz került elő részletesen dokumentált régészeti ásatásból (Zengővárkony (3), Szombathely Oladi plató (2), Alsónyék (1), Gorzsa (1), a többi szórványlelet. Lelőhelyeik és legfontosabb adataik megtalálhatók az **1. tábláza**tban. Ebben a munkánkban a Szakmány és munkatársai (2013) cikkben ismertetett **3** kőeszközön felül 10 további HP metaofiolit nyersanyagú kőeszközt mutatunk be részletesen (**1. ábra**).

Munkánk során kizárólag roncsolásmentes analitikai módszereket használtunk. Eredményeink alapján kőzetszöveti, ásvány- és teljes kőzet kémiai adatok segítségével a nemzetközi archeometriai szakirodalomban található felosztás alapján csoportosítjuk a leleteket. Eredményeink jól egyeznek az Északnyugat Olaszország területéről leírt kőeszközök adataival (D'Amico et al. 2003, D'Amico 2012, Pétrequin et al. 2012), így az általunk vizsgált kőeszközök nyersanyagának forrásterülete feltehetőleg megegyezik azokéval. Ezek elsődleges előfordulása a Monviso, Voltri ill. az ÉNy-i Appenninek területe és másodlagosan az ezekről a területekről lepusztult, majd a negyedidőszakban áthalmozódott és a Pó síkságon, valamint a Curone és a Staffora folyók környezetében lerakódott konglomerátumok kavicsanyaga (D'Amico et al. 2003, D'Amico & Starnini 2006, 2012, Pétrequin et al. 2012).

KEYWORDS: POLISHED STONE TOOL, JADE, ECLOGITE, PROVENANCE, NON-DESTRUCTIVE ANALYSIS

KULCSSZAVAK: CSISZOLT KŐESZKÖZ, JADEITIT, EKLOGIT, NYERSANYAG FORRÁSTERÜLET, RONCSOLÁSMENTES ANYAGVIZSGÁLAT



Fig. 1.:

Stone implements from Hungary made of HP metaophiolite raw material. Most important form types can be seen on the picture.

1. ábra:

Néhány magyarországi HP metaofiolit anyagú kőeszköz. A képen az összes fontosabb alaki típus látható.

Introduction

Green or greenish colour is very popular among the prehistoric polished stone tools. This colour is characteristic for several different rock types, e.g. greenschist, contact metabasite, nephrite and serpentinite; however HP metaophiolites are outstanding among the 'greenstones' both by beauty and quality (Fig. 1.). Their elaboration is unique, in most of cases they have flat, elongated, lingular or triangular shape and they are of different shades of green and their surface is especially finely polished. These types of stone implements are very widespread all over Western Europe (D'Amico 2003, Pétrequin et al. 2011, Klassen 2012, Domínguez-Bella et al. 2015). Their penetration decreasing toward east, the easternmost known localities are in Bulgaria, however, they were

practically unknown in the Carpathian Basin until the recent past (Pétrequin et al. 2011, Szakmány et al. 2013).

Their importance is derived from their scarcity and distant origin (**Fig. 2.**). Their first recognition is connected with the study of major polished stone tool assemblages: Friedel et al. (2008) mentioned six pieces of HP metaophiolite stone implements from Ebenhöch collection without detailed description. Szakmány et al. (2013) gave detailed information on three pieces of stone implements made of HP raw material. These pieces are from the collections accumulated by F. Ebenhöch and I. Miháldy in the 19th century. Because of the afore mentioned article is in Hungarian, these important artefacts will be presented in this work again (see below) for a wider audience.



Fig. 2.: Raw material sources of HP metaophiolites. 1 – Primary sources; 2 – secondary/tertiary sources (Oligocene conglomerates and their redeposited sediments). After D'Amico & Starnini 2012.

2. ábra: A HP metaofiolit nyersanyagok előfordulási területei. 1 – elsődleges előfordulások; 2 – másodlagos/harmadlagos előfordulások (Oligocén konglomerátumokban és azok áthalmozott üledékeiben). D'Amico & Starnini (2012) alapján.

General characterisation of jade and eclogite raw material

HP metaophiolites can be divided into two groups based upon the garnet content of these rocks. The most widespread group (in both area and mass/amount) is the eclogite group, which contains large amount of garnets beyond the alkaline pyroxenes (at least 5 % according to D'Amico 2003). Among the alkaline pyroxenes, omphacite is much more abundant than the other Na-pyroxene types, however jadeite dominated eclogites do exist as well. The group of 'alkaline-pyroxenites' or 'jades' is much smaller. Dominant mineral phases are the different alkaline-pyroxenes in this group, like jadeite and omphacite, sometimes aegirine and/or aegirine-augite. The quantity of alkalinepyroxenes is more than 80% in these samples. In addition to the main phases, both groups contain some accessory minerals in these samples, e.g. rutile, zircon, ilmenite, apatite and titanite.

These two groups are formed under the same conditions, by high pressure (2-3 GPa) and low to medium temperature (500-600 °C) metamorphism from basaltic protoliths. Based upon their mineral and chemical composition, the HP metaophiolite

raw material of these stone implements were formed by the younger, Alpean stage of orogenesis, during the Eocene epoch (Compagnoni 2007; Beltrando et al. 2010).

The Alpine type HP metaophiolites suitable implements for raw material can be found both in primary, secondary (Oligocene) and tertiary (recent) positions in NW-Italy. This latter were redeposited in the Quaternary period, and Prehistoric people used this as raw material source (D'Amico & Starnini 2006). Primary occurrences can be found in the eastern range of the Western Alps, from Monviso till the Aosta valley, and in the Voltri Massif at the north-western end of Apennines (Monte Beigua, Fig. 2.). The formation of these units occurred after the subduction of the floor of Tethys-ocean. Both areas were uplifted in the Alpean orogenic stage, after the high pressure metamorphism (Compagnoni et al. 2007). Secondary raw material sources can be found at the nearby piedmonts of the Western Alps and Voltri Massif and in the Po plain. They were formed during Oligocene by sedimentation of eroded materials from the Western Alps and Voltri Massif. These types of raw material sources were described from recent sediments of Staffora and Curone rivers (D'Amico & Starnini, 2012), and at the upper Poriver, which originates from the Monviso region (Pétrequin et al. 2012) (**Fig. 2.**).

Material quality played an important role in the usage and spreading of these stone implements. Stone tools made of eclogite were used as woodworking tools by the local prehistoric people, however exceptionally shaped and finished stone implements made of jade can be found all over Europe. They are interpreted by the archaeologists as symbols of power and/or religion, and/or wealth rather than common work tools. The selection of raw material was really careful, it is indicated by the distribution of the quantity, the quality and the range of these stone implements. 'Jade' as raw material can be found at very limited area and in relatively small quantities, however, stone implements made of this raw material can be found more than 1500 km away from the source areas. On the other hand, the much more abundant eclogites were mostly locally (regionally) used as raw material for work tools (D'Amico 2005, D'Amico & Starnini 2012, Pétrequin et al. 2011).

Archaeological background

The analysed artefacts are from various localities within Hungary, some of them from archaeological excavations of prehistoric sites, others from recent field surveys or surface collections as stray finds in the old collection of various museums (**Table 1.**, **Fig. 3.**). Their description here follows roughly the temporal order of their recognition among the polished stone artefacts in Hungary (Szakmány 2009, Szakmány et al. 2013).

Miháldy collection

The collection accumulated by István Miháldy (1833-1901) contains 378 polished stone tools, now curated in the Laczkó Dezső Museum, Veszprém. Unfortunately, the information on the exact provenance of the pieces was lost during reinventorisation in the 1950-ies. We know, however, that all the samples were found at different sites in the Bakony Mts. and its surroundings (Horváth, T. 2001, Füri et al. 2004). One piece of HP metaophiolite artefact was found in this collection (sample 55.1276).

Ebenhöch collection

The collection was accumulated by Ferenc Ebenhöch (1821-1889), prebend of Győr, NE Hungary. His collection, partly donated, partly sold to the Hungarian National Museum contains more than 700 polished stone tools and it is now part of the Prehistoric Collection of the HNM, Budapest.



Fig. 3.: Archaeological localities of presented artefacts. 1 – Neszmély, 2 – Bakonypéterd, 3 – Szombathely, Olad-plateau, 4 – Zirc, 5 – Iszkaszentgyörgy, 6 – Bakony Mts., 7 – Hódmezővásárhely-Gorzsa, 8 – Lábod, 9 – Zengővárkony, 10 – Alsónyék.

3. ábra: A bemutatott kőeszközök régészeti lelőhelyei. 1 – Neszmély, 2 – Bakonypéterd, 3 – Szombathely, Oladi-plató, 4 – Zirc, 5 – Iszkaszentgyörgy, 6 – Bakony, 7 – Hódmezővásárhely-Gorzsa, 8 – Lábod, 9 – Zengővárkony, 10 – Alsónyék.

The artefacts were found in Western and Upper Hungary (latter is today Slovakia). The polished stone tools of this collection were studied in details by O. Friedel (Friedel et al. 2011). Two pieces of HP metaophiolite stone tools were investigated from this collection (samples 300/1876.247 and 300/1876.264). The localities involved are Bakonypéterd and Neszmély with no further archaeological context.

Gorzsa

Hódmezővásárhely-Gorzsa is a famous Late Neolithic (Tisza culture) tell settlement (Horváth, F. 2003, 2005). The site lies at the confluence of the Tisza and Maros rivers in the Great Hungarian Plain, north of Szeged. Approximately 1,000 square meters of the tell settlement were excavated and more than 1000 stone artefacts were collected from controlled stratigraphical context (Szakmány et al. 2008; Starnini et al 2015). One HP metaophiolite stone implement was identified in the rich the lithic assemblage, this is the easternmost example of HP stone tools in Hungary so far (sample 99.3.1863).

Szombathely (Olad plateau)

Olad plateau was a Late Neolithic settlement of the early Lengyel culture between the early and the classical periods (Lengyel Ib, Ilon 2011). Two very small fragments of HP metaophiolite stone implements were found in this site (samples Olad-321 and Olad-329). **Table 1.:** Physical properties and basic petrographic description of the investigated HP metaophiolite samples. Abbreviations: LDM, Vp. – Laczkó Dezső Museum, Veszprém, HNM, Bp. – Hungarian National Museum, Budapest, RRM – Rippl-Rónai Museum, EW – private collection of Ernő Wolf, WMM – Wosinsky Mór Museum, JPM – Janus Pannonius Museum, SM – Savaria Museum, MFM – Móra Ferenc Museum, coll. – collection, ND – not determined.

1. táblázat: A vizsgált HP metaofiolit anyagú kőeszközök fizikai tulajdonságai és alapszintű kőzettani leírásai. Rövidítések: LDM, Vp. – Laczkó Dezső Múzeum, Veszprém, HNM, Bp. – Magyar Nemzeti Múzeum, Budapest, RRM – Rippl-Rónai Múzeum, EW – Wolf Ernő magángyűjteménye, WMM – Wosinsky Mór Múzeum, JPM – Janus Pannonius Múzeum, SM – Savaria Múzeum, MFM – Móra Ferenc Múzeum, coll. – gyűjtemény, ND – nem mért.

	Locality	Museum (collection)	Inv. number	(mm)	w (mm)	H (MM)	MS (x10 ⁻³ SI)	shape	colour	archaeologi- cal context	macroscopic description
-	Bakony	LDM, (Mihâldy collection)	55.1276	95	40	15	0,17	flat, elongated, triangular adze	medium to dark green	stray find	homogenous, lighter green veins and patches on the darker green background, tiny (smaller than 1 mm), white subhedral-anhedral crystals
7	Neszmély	HNM, (Ebenhöch collection)	300/1876.247	40	28	7	0,47	flat adze	dark green	stray find	homogenous, pale green, greyish green, grey and locally very dark green patches and veins
3	Bakonypéterd	HNM, (Ebenhöch collection)	300/1876.264	56	25	∞	0,10	flat, elongated, lingular adze	pale green	stray find	homogenous, medium to dark green patches, locally rounded grains with darker and brighter shade than the matrix
4	Iszkaszent- györgy	HNM, (Prehistoric coll.)	39/1903	118	50	8	0,32	flat, strongly elongated, lingular adze	very dark green	stray find	homogenous, medium ét dark green patches, lot of relatively large rutile (larger than 1 mm), pale green-greenish white elongated crystals (jd?) locally
ŝ	Lábod	RRM, Kaposvár	3127	95	50	25	QN	thick, elongated, lingular adze	dark green	stray find	slightly inhomogenous, dark and medium green patches
9	Zirc	EW, Zirc	81/W2 5	36	29	01	0,19	flat, slightly elongated, triangular adze	pale green	stray find	slightly inhomogenous with brigther green patches
2	Alsónyék	WMM, Szekszárd	M6.2010.10 B.3060.3	116	50	22	0,95	thick, strongly elongated triangular adze	greenish black	Lengyel culture	roughly homogenous, sheared, bands of different shades of green changing, large (max. 2 mms) brown and green grains locally
×	Zengővárkon y	JPM, Pécs	N.5/47-1939	40	30	2	0,19	flat, slightly elongated, triangular adze	bluish black	Lengyel culture	roughly homogenous, slightly foliated, tiny, white subhedral-anhedral crystals, crystalagglomerates and green veins
6	Zengővárkon y	JPM, Pécs	N.1/81-1938	62	39	13	0,20	flat, elongated, triangular adze	green	Lengyel culture	roughly homogenous, patches with different sheds of green, some large (max. 5 mms) white patches, 1-2 mm long dark green subhedral-euhedral crystals locally
10	Zengővárkon y	JPM, Pécs	N.11/169- 1938	58	31	12	0,39	flat, strongly elongated, lingular adze	dark green	Lengyel culture	homogenous, medium and dark green patched, max. 2 mm long rutile crystals
Ξ	Szombathely, Olad	SM, Szombathely	Olad-321	18	20	9	0,03	fragment of flat axehead	medium green and red	Lengyel culture	roughly homogenous, red grains in medium green matrix
12	Szombathely, Olad	SM, Szombathely	Olad-329	20	27	<u>0</u>	0,03	fragment of flat axehead	medium green	Lengyel culture	roughly homogenous, pale and medium green patches, tiny, locally white crystals
13	Hódmező- vásárhely, Gorzsa	MFM, Szeged	99.3.1863	40	25	6	0,24	flat, slightly elongated, triangular adze	dark green	Tisza culture	very dark green veins and patches in the dark green matrix, some (4-5) garnets with 1 mm of max diameter, few, elongated (max. 1 mm long) white crystals (zm or an)



Fig. 4.: Artefacts presented in this work. Numbers are referring to first column on Table 1.4. ábra: Az összes bemutatott kőeszköz. A sorszámok az 1. táblázat 1. oszlopára vonatkoznak.

Zengővárkony

Classical settlement and cemetery of the Lengyel Culture, excavated and published by J. Dombay (Dombay 1939, 1960). The polished stone artefacts were investigated previously (Biró et al. 2003) and reconsidered for sourcing the HP metamorphites. Three HP metaophiolite artefacts were identified, all of them found in graves (samples N.1/81-1938, N.11/169-1938 and N.5/47-1939).

Alsónyék

The largest known cemetery of the Lengyel Culture, excavated in course of preventive rescue excavations. The elaboration of the site is in progress. One HP metaophiolite artefact was found in a princely grave (Grave 3060, sample M6.2010.10B.3060.3), published separately (Zalai-Gaál et al. 2011).

In addition, three stray finds were investigated, their localities: Iszkaszentgyörgy, Fejér county (HNM 39/1903); Lábod (3127), RMM Somogy county; Zirc (81/W2.5), private collection, Veszprém county. All the objects with documented archaeological context are from the Late Neolithic. Most of them belong to Lengyel culture (Zengővárkony, Olad-plateau and Alsónyék). The object from Gorzsa is assigned to the Tisza culture, coeval with the Lengyel culture.

Methods

The stone implements made of jade and related rocks are generally intact, therefore only nondestructive methods can be used for their investigation. These non-destructive methods included macroscopic analyses, stereomicroscopy, magnetic susceptibility measurement, geochemical analysis by prompt gamma activation analysis, nondestructive X-ray diffractometry (performed at Miskolc University and presented in the paper by Kristály (2014) in this volume) and original surface analysis, explained in details below.

At the same time, spectroradiometrical analyses (Errera et al. 2007) were performed on the same items by M. Errera that will be reported on elsewhere.

Loupe and stereomicroscope were used for the basic petrographic description. Magnetic susceptibility (MS) measurements were made with portable Kappameter KT-5. Real MS values were calculated by different corrections of size and thickness (Bradák et al. 2005, 2009).

All of the BSE images and EDX analyses were made by non-destructive original surface analysis method (Bendő et al. 2013) that was developed for textural and mineral chemistry analysis of stone tools. Besides the standards listed in Bendő et al. 2013, jadeite (SPI #AS1195-AB), omphacite (Smithsonian Microbeam Standard, SMS, NMNH 110607), garnet (SMS, NMNH 87375) and pyrope (SMS, NMNH 143968) mineral standards were used. These measurements were made at the Department of Petrology and Geochemistry, Institute of Geography and Earth Sciences, Eötvös Loránd University, Budapest. The instrument was an AMRAY 1830 scanning electron microscope equipped with EDAX PV9800 energy dispersive spectrometer. Conditions of analysis: accelerating potential: 20 kV, beam current: 1 nA, beam diameter: focused electron beam (~50-100 nm), measurement time: 100 sec (livetime).

Non-destructive PGAA was performed at the Budapest Research Reactor operated by the Centre for Energy Research, Hungarian Academy of Sciences. The method is suitable for the quantitative determination of the average concentrations of the major elements (SiO₂, TiO₂, Al₂O₃, Fe₂O₃^t, MnO, MgO, CaO, Na₂O, K₂O and H₂O) and some trace elements (e.g. B, Cl, Sm, Gd) in a few cm³ volume. Thermal equivalent beam intensity was 7.75×10^7 cm⁻²×s⁻¹. Calibrated Compton-suppressed HPGe detector was used to detect prompt gamma spectra, and Hypermet PC software was used for evaluation (Révay, 2009, Szentmiklósi et al. 2010, Szakmány et al. 2011).

Results

Physical properties and MS values of the stone implements are summarized in Table 1. with short macroscopic description of the samples. According to our results, the objects can be divided into six raw material groups. The first four groups are the 'Na-pyroxenites', those HP metaophiolite raw materials which do not contain significant amounts of garnet (less than 5 %). These groups are listed by increasing iron content. The fifth group contains the only glaucophane schist sample, and the last group comprises an iron eclogite specimen (Fig. 4.). Nomenclature diagram of alkaline pyroxenes (Morimoto et al. 1988) was used for the present mineral chemical grouping of samples. Garnets are presented also on a triangular diagram.

PGAA results are presented on multi-element diagrams normalized to Upper Continental Crust (UCC) data by McLennan (2001) for easier comparison. Data fields of Italian HP metaophiolite stone implements by D'Amico et al. (2003) are also presented on these diagrams for comparison.

Jadeitites

Two pieces of jadeitite stone implements were found until this time, both of them located in large old museum collections, one is from the Ebenhöch collection (300/1876.264), the other one is from the Miháldy collection (55.1276).

In respect of mineral composition, their raw material contains much more jadeite than omphacite (**Fig. 5.**). Both samples contain zircon and allanite as accessory minerals. Titanite and xenotime can also be found in them. The Na-pyroxenes are generally zoned, they have jadeite core and omphacite rim (**Fig. 6., Fig. 7.**). Deformation textures are missing in these samples.



Fig. 5.: Typical texture of jadeitites: small amount of omphacite is scattered among jadeite. According to the textural characteristics, omphacite was formed in the latest stage of metamorphism in this sample (300/1876.264, Bakonypéterd).

5. ábra: Tipikus jadeitit szövet: kevés omfacit helyezkedik el szétszórva a jadeit kristályok között. Szöveti jellegzetességei alapján az omfacit a metamorfózis legutolsó fázisában keletkezett ebben a mintában (300/1876.264, Bakonypéterd).



Fig. 6.: Zoned pyroxenes with jadeite core and omphacite rim. Sample 55.1276 (Bakony Mts.).

6. ábra: Zónás piroxénkristályok jadeit maggal és omfacit peremmel az 55.1276-os mintában (Bakony).



Fig. 7.: Pyroxene compositions in the jadeitite samples. Discrete compositions of jadeite and omphacite are very well visible.

7. ábra: A jadeitit minták piroxénjeinek összetétele. A jadeit és omfacit mezőkben elhelyezkedő összetételek határozott elkülönülést mutatnak.

Jadeitic pyroxenes are similar in composition in these two samples, but the omphacitic pyroxenes of sample 300/1876.264 contain much more omphacite than pyroxenes of sample 55.1276. Jadeitic pyroxenes appear along a steep trend line toward omphacitic pyroxenes but the continuity of this trend is doubtful based on these data (Fig. 7.).

Bulk-rock data are corresponding very well to data of D'Amico et al. (2003, grey field). According to these results these samples have fairly different calcium and magnesium content. This difference was probably caused by the higher omphacite content of sample 300/1876.264 (Fig. 8.).

Mixed jades

This jade group contains two pieces of stone tools. One of them was found at the excavations of Zengővárkony (N.1/81-1938), from documented archaeological context. The other one is a stray find from Zirc (81/W2.5).

Their raw material contains more jadeite than omphacite or aegirine-augite (**Fig. 9.**). Both samples contain rutile as accessory mineral. Zircon, allanite, titanite, xenotime and monazite can also be found in them. Retromorphic phases (albite, epidote) can be found in sample N.1/81-1938.



Fig. 8.: Bulk-rock compositions of jadeitite samples normalized to UCC by McLennan (2001). Results showing very well the correspondence to jadeitite compositions by D'Amico et al (2003). Differences in compositions are possibly caused by varieties in omphacite content of the samples.

8. ábra: A jadeitit minták teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). Az adatok nagyon jó egyezést mutatnak D'Amico et al. (2003) jadeitit adataival. A két minta közötti összetétel különbséget valószínűleg a minták különböző omfacit-tartalma okozza.



Fig. 9.: Texture of mixed jades with accessory and retromorphic minerals. Sample N.1/81-1938 from Zengővárkony.

9. ábra: Kevert jadeitit szövete, akcesszórikus és retromorf ásványai. N.1/81-1938 minta Zengővárkonyról.

Na-pyroxenes are frequently zoned, they have jadeite core and omphacite/aegirine augite rim (Fig. 10, Fig. 11.). Na-pyroxenes in sample 81/W2.5 have a 'radial' texture (Fig. 10.). Deformation structures are not present in these samples.



Fig. 10.: 'Radial' texture of alkaline pyroxenes in sample 81/W2.5 (Zirc). Pyroxenes shows 'normal' zonation, with jadeite core and omphacite rim.

10. ábra: Az alkáli piroxének "sugaras" szövete a 81/W2.5 mintában (Zirc). A piroxének normál zónásságot mutatnak, jadeit maggal és omfacit peremmel.



Fig. 11.: Pyroxene compositions in mixed jades. Jadeites have very similar composition in both samples, however omphacite compositions are rather different, omphacites of sample 81/W2.5 are richer in iron than omphacites of sample N.1/81-1938.

11. ábra: Piroxén összetételek a kevert jadeititekben. A jadeiteknek nagyon hasonló az összetétele a két mintában, míg az omfacitok összetétele meglehetősen eltérő, a 81/W2.5 mintában az omfacitok sokkal vasgazdagabbak, mint az N.1/81-1938 mintában.



Fig. 12.: Bulk-rock compositions of mixed jade samples normalized to UCC by McLennan (2001). Two datasets are very similar to each other, and their correspondence to mixed jade data of D'Amico et al. (2003) is good too.

12. ábra: A kevert jadeititek teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). A két adatsor nagyon hasonló, és jó egyezést mutatnak D'Amico et al. (2003) kevert jadeitit adataival is.

Composition of jadeitic pyroxenes of the two samples are very similar, they appear along a relatively steep trend line. Omphacitic pyroxenes are very different, they show two distinct trends. Sample N.1/81-1938 trends towards the calcium rich omphacites, 81/W2.5 towards the iron rich aegirine-augites. The continuity of these trends is doubtful based on these data (Fig. 11.).

Bulk-rock data are very similar to each other. Their correspondence to data of D'Amico (2003, grey field) is good, differences probably caused by the larger natural variety of the raw material of the earlier investigated samples (Fig. 12.).

Iron-mixed jades

This jade group contains three pieces of stone tools, all of them found at different archaeological excavations. N.11/169-1938 was found in Zengővárkony, 99.3.1863 was found in Gorzsa and Olad-329 was found at Szombathely, Olad-plateau.

Their raw material contains a lot of jadeite with large amounts of omphacite and/or Fe-jadeite and/or aegirine-augite (Fig. 13.). Accessory minerals are zircon, allanite, rutile, apatite, ilmenite and titanite. Retromorphic phase (chlorite) can be found in one sample (Olad-329).

Na-pyroxenes are frequently zoned, in most of cases they have a jadeite core and Fe-jadeite or omphacite or aegirine-augite rim (Fig. 14, Fig. 15.).



Fig. 13.: Textural view of an iron-mixed jade with accessory and retromorphic minerals. Sample Olad-329

13. ábra: Vas-kevert jadeitit szöveti képe, akcesszórikus és retromorf ásványai. Olad-329-es minta.



Fig. 14.: Zoned pyroxenes with jadeite core and iron-jadeite/aegirine-augite rim. Sample N.11/169-1938 (Zengővárkony).

14. ábra: Zónás piroxénkristályok jadeit maggal és vas-jadeit/egirinaugit peremmel. N.11/169-1938 minta (Zengővárkony).

The pyroxenes of these samples have rather different composition. Two of the stone tools contain three different compositional groups of pyroxene, one is near the clear jadeite composition, the second group is in the jadeite field with high iron content (iron-jadeite) and the third has omphacitic–aegirine-augitic composition. These pyroxenes have a flat trend-like line towards aegirine-augite across the omphacite field pyroxenes. Continuity of these trend lines are doubtful based on these data, again (Fig. 15.).

Bulk-rock data are corresponding very well to data published by D'Amico (2003, grey field). According to these results, these samples have fairly different manganese and magnesium content (**Fig. 16.**).



N.11/169-1938 (Zengővárkony)

Fig. 15.: Very diverse pyroxene compositions in iron-mixed jades. Sample 99.3.1863 contains four different pyroxenes: jadeite, iron-jadeite, omphacite and aegirine-augite.

15. ábra: A vas-kevert jadeititek erősen eltérő piroxén összetételei. A 99.3.1863-as minta négyféle piroxént tartalmaz: jadeitet, vas-jadeitet, omfacitot és egirinaugitot.



Fig. 16.: Bulk-rock compositions of iron-mixed jade samples normalized to UCC by McLennan (2001). Datasets are corresponding to iron-mixed jade data of D'Amico et al. (2003).

16. ábra: A vas-kevert jadeititek teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). Az eredmények jó egyezést mutatnak a D'Amico et al. (2003) által mért vas-kevert jadeititek értékeivel.

Iron jadeitite

Fe-jadeitite is a frequent raw material among the Hungarian HP metaophiolite stone tools, three pieces were found. Unfortunately all of them are stray finds without archaeological context. One piece is from the Ebenhöch Collection of the HNM (300/1876.247), the second is from Iszkaszentgyörgy (Prehistoric collection of Hungarian National Museum, Inv. nr. 39/1903), and the third is from Lábod from the collection of the Rippl-Rónai Museum, Kaposvár (Inv. nr. 3127).



Fig. 17.: Texture and accessory minerals of ironjadeite, sample 39/1903 (Iszkaszentgyörgy).

17. ábra: Vas jadeitit szöveti képe és akcesszórikus ásványai, 39/1903-as minta (Iszkaszentgyörgy).



Fig. 18.: Zoned pyroxenes with jadeite core and iron-jadeite rim. Sample 3127 from Lábod.

18. ábra: Zónás piroxének jadeit maggal és vasjadeit peremmel. 3127 minta, Lábod.

Their raw material contains jadeite and Fe-jadeite; while omphacite and aegirine-augite are absent (Fig. 17., Fig. 19.). Common accessory minerals are zircon and rutile. Titanite, allanite, ilmenite, xenotime and monazite can be found in them as well.

The Na-pyroxenes are generally zoned, they have jadeite core and Fe-jadeite rim (Fig. 18, Fig 19.). Deformation textures are absent from these samples.

Pyroxenes of sample 300/1876.247 contain more iron than pyroxenes of the other two samples. Pyroxenes appear along a very shallow/flat trend line towards aegirine (**Fig. 19.**).



Fig. 19.: Pyroxene compositions in iron jadeitites. All of results fall to the jadeite field, however compositions are varying between iron poor and iron rich jadeites.

19. ábra: A vas-jadeititek piroxénjeinek összetétele. Minden mért érték a jadeit mezőbe esik, de az összetételek a vasszegény és vasgazdag jadeitek között váltakoznak.



Fig. 20.: Bulk-rock compositions of iron jadeite samples normalized to UCC by McLennan (2001). Correspondence to iron jadeite data of D'Amico et al. (2003) is good.

20. ábra: A vas jadeititek teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). A mért adatok elég jól egyeznek D'Amico et al. (2003) vas-jadeitit adataival.

Correspondence of bulk-rock analyses to data of D'Amico (2003, grey field) is good, the variance is probably caused by the diversity of the raw material (**Fig. 20.**).

Glaucophane schist (retrograde omphacite schist)

This type of glaucophane schist is very rare among the Hungarian HP metaophiolite stone tools, only one piece was found until this time. This piece is from Zengővárkony archaeological site (N.5/47-1939).

Its raw material contains a lot of glaucophane and Na-pyroxenes as well. Most of the Na-pyroxenes are omphacite, jadeite occurs on the rim of omphacite crystals (**Fig. 21, Fig. 22.**). Accessory minerals are ilmenite, apatite, titanite (**Fig. 21.**).

Na-pyroxenes are zoned in this sample, but they show inverse zonation against the prior samples (**Fig. 21.**). According to the texture, glaucophane was formed in the latest, retrograde stage of metamorphism. N.5/47 has slightly foliated texture (**Fig. 21.**).

There are two compositional varieties of alkaline pyroxenes in this sample. The more abundant pyroxene is the omphacite, which has a steep trend line on the diagram. Jadeite compositions are much more close to each other, they appear in a pile and they do not form a trend line (**Fig. 22.**).

Bulk-rock data fit very well to glaucophane schist data of by D'Amico (2003, thick grey line). The only difference between the two compositions is the Na2O content, probably sample N.5/47-1939 has higher alkaline pyroxene content than the reference sample. Notably, the distribution of omphacite schist and glaucophane schist are very similar to each other (thick grey line and pale grey fields in **Fig. 23.**).



Fig. 21.: Texture of glaucophane schist with accessory minerals. Sample N.5/47-1939 (Zengővárkony).

21. ábra: Glaukofánpala szöveti képe és akcesszórikus ásványai. N.5/47-1939 minta (Zengővárkony).



Fig. 22.: Alkaline pyroxene compositions in glaucophane schist. Compositions are showing two distinct group of jadeite and omphacite.

22. ábra: Alkáli piroxén összetételek a glaukofánpalában. A mért eredmények két különálló halmazt képeznek, egyet a jadeit és egyet az omfacit mezőben.



Fig. 23.: Bulk-rock compositions of glaucophane schist sample normalized to UCC by McLennan (2001). Dataset are very similar to glaucophane schist (thick gray line) and omphacite schist (pale gray field) data of D'Amico et al. (2003).

23. ábra: A glaukofánpala teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). Az adatsor nagyon hasonlít D'Amico et al. (2003) glaukofánpala (vastag szürke vonal) és omfacitpala (világosszürke mező) eredményeire.

Iron eclogites

Eclogite is rare raw material among the investigated HP metaophiolite stone tools, only two pieces were found. Both of them were found on excavated archaeological sites, M6.2010.10B.3060.3 was

found in Alsónyék, Olad-321 was found at Szombathely, Olad plateau.

Their raw material contains a high amount of garnets in addition to alkaline pyroxene which is mainly omphacite (Fig. 24.). Accessory minerals are ilmenite, zircon, apatite and rutile. Retromorphic phase (epidote) occurs in the sample from Olad (Fig. 25.).

Alkaline pyroxenes seem to be homogenous in most of cases, but in some cases they have relatively jadeite-rich core (**Fig. 26.**).



Fig. 24.: Typical texture of iron eclogites with accessory minerals and large amount of garnets. Moderate inhomogeneity of pyroxenes can be observed on this sample (M6.2010.10B.3060.3, Alsónyék). Foliation is visible on the placements of ilmenite crystals.

24. ábra: Tipikus vas eklogit szövet akcesszórikus ásványokkal és sok gránáttal. A piroxének összetételében enyhe inhomogenitás figyelhető meg. Az enyhe foliáció az ilmenit kristályok elhelyezkedésén látható. M6.2010.10B.3060.3 minta, Alsónyék.



Fig. 25.: Elongated, homogenous omphacite crystals among garnets and ilmenites in the sample Olad-321.

25. ábra: Nyúlt, homogén omfacit kristályok a gránátok és az ilmenitek között az Olad-321 mintában.



Fig. 26.: Pyroxene compositions in the iron eclogite samples. Most of the compositions fall into the omphacite field, however there are some jadeite and aegirine-augite, also.

26. ábra: Alkáli piroxének összetétele a vas eklogitokban. A legtöbb esetben omfacitos összetételt mértünk, de előfordulnak jadeit és egirinaugit összetételű mérések is.



Fig. 27.: Garnet compositions in iron eclogite. Most of the compositions can be found in one narrow pile, however the distinct values are demonstrating the zonation of crystals.

27. ábra: Gránát összetételek a vas eklogitokban. A legtöbb érték egy szűk halmazban található, de néhány ettől eltérő összetétel is előfordul, amik a kristályok zónásságát bizonyítják.

Garnets are slightly zoned, probably, but this zonation is invisible on these rough surfaces (**Fig. 27.**). M6.2010.10B.3060.3 has slightly foliated texture (**Fig. 24.**).



Fig. 28.: Bulk-rock compositions of iron eclogite samples normalized to UCC by McLennan (2001). Two datasets are very similar to each other, and they are corresponding very well to iron eclogite data of D'Amico et al. (2003), also.

28. ábra: A vas eklogitok teljeskőzet-kémiai összetétele a kontinentális felső kéreg összetételre normálva (McLennan 2001). A két adatsor nagyon hasonló egymáshoz, és mindkettő nagyon jól egyezik D'Amico et al. 2003 vas eklogit adataival.

Pyroxene composition of these samples do not show any trends or alignment. Most of them are omphacite, however there are jadeitic and aegirineaugitic parts as well (**Fig. 26**.).

Most of the garnet compositions are in one pile, there are only two exceptions, and possibly they are indicating the zonation of the crystals (**Fig. 27**.).

Bulk-rock data are corresponding very well to data of D'Amico et al. (2003, grey field). According to these results these samples have very similar composition (**Fig. 28**.).

Discussion

Most of the investigated stone implements have characteristic, elongated, triangular or lingular shape which is similar to the stone implements described from Western Europe (D'Amico et al. 2003, D'Amico & Starnini 2006, Pétrequin et al. 2012). Instrumental analyses allow the grouping of these stone tools. According to our results, they can be divided into 6 raw material groups. These raw material groups are jadeitites, mixed jadeitites, ironmixed jadeitites, iron jadeitites, glaucophane schist and iron eclogites (Fig. 29.).







CAO : E. Gauthier and J. Desmeulles – University of Franche-Comté UMR CNRS 6249, MSHE C.N. Ledoux - January 2010

Fig. 30.: Distribution of HP metaophiolite stone implements in Europe completed with Hungarian artefacts. Archaeological excavations where the investigated finds were found are marked on map. Modified after

Pétrequin et al. 2011.**30. ábra:** HP metaofiolit nyersanyagú kőeszközök eloszlása Európában, kiegészítve a magyarországi adatokkal. A régészeti ásatások, melyek során a tanulmányozott leletek előkerültek, külön fel vannak tüntetve. Pétrequin et

al. (2011) alapján módosítva.

Based on their inventory number, they can be grouped accordingly:

- * Jadeitite: 55.1276 and 300/1876.264
- * Mixed jadeitite: N.1/81-1938 and Zirc 81/W2.5
- * Iron-mixed jadeitite: N.11/169-1938, 99.3.1863 and Olad-329
- * Fe-jadeitite: 300/1876.247, 39/1903 and 3127
- * Glaucophane schist: N.5/47-1939
- * Iron-eclogite: M6.2010.10B.3060.3 and Olad-321.

Based upon data from the technical literature (D'Amico et al. 2003, D'Amico & Starnini 2006, Pétrequin et al. 2012), these raw materials, based on their mineral assemblage, chemical composition and textural properties, are identical to the Northwestern Italian HP metaophiolites which were formed by the Alpine orogenic stage. We do not make claims on the exact localisation of the raw material source proper because the relevant areas known from publications (see Fig. 2.) are rather large, and they are all at least about 1000 km from Carpathian Basin, practically in the same direction. Therefore, from the respect of prehistoric contacts they have the same significance.

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Our results essentially increased the knowledge about the distribution range of these stone implements (Fig. 30.). Stone tools made of western Alpine HP metaophiolite raw material were formerly unknown in the Carpathian Basin, especially in Hungary. The artefacts presented here were mainly found in the Transdanubian region (West Hungary); there is one piece, however, from East Hungary as well (Fig. 30.).

addition, very important result is the In heterogeneity of lithotypes, not all of presented stone implements were made of the most widespread "pure jade" raw material, but eclogites and glaucophane schist are also occurring. This heterogeneity is explicable with several reasons. One possible scenario is the development of manufacturing and discovery of better and better quality sources for these stone implements in due time. Other probability is the changes in the transportation routes and/or procurement and contact modes in the function of time and/or regions. Last but not least these very distinct materials may be transported from multiple, coexistent sources/manufactures along multiple, coexistent trading routes.

Conclusion

In the last years there was a significant growth in the number of recognized and investigated stone implements made of HP metaophiolite raw material in Hungary (**Fig. 30**.). This growth is due to systematic quest for greenstones and the applied non-destructive methods (PGAA and original surface method by SEM-EDX). These methods proved their suitability to identify the raw material of stone implements and capability to provide good quality data for comparison with data in the international literature.

Artefacts from known archaeological context are from the Late Neolithic period and most of them belong to the Lengyel Culture. The only piece from East Hungary belongs to the Tisza Culture which is coeval to Lengyel Culture, and they were in regular contact with each other.

According to our results there is a large probability of finding other HP metaophiolite artefacts in Hungary, especially in the course of systematic petrologic investigations of findings of Lengyel and coeval cultures and from the old collections.

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Abbreviations on pictures

ab – albite aeg-au – aegirine-augite aln - allanite ap - apatitechl - chlorite ep – epidote Fe-jd - iron-jadeite gln – glaucophane grt - garnet ilm – ilmenite jd – jadeite omp-omphacite rt - rutile ttn - titanite xtm – xenotime zrn - zircon

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POSSIBLE PROVENANCES OF NEPHRITE ARTEFACTS FOUND ON HUNGARIAN ARCHAEOLOGICAL SITES

(PRELIMINARY RESULTS)

MAGYARORSZÁGI RÉGÉSZETI LELŐHELYEKEN TALÁLT NEFRIT ESZKÖZÖK ÉS EZEK LEHETSÉGES SZÁRMAZÁSI HELYE

(ELŐZETES EREDMÉNYEK)

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Abstract

Nephrite is mainly known in prehistoric context as raw material for polished stone tools. It is present among archaeological finds in Hungary only in a few numbers. They are known mostly from Transdanubian archaeological sites.

The general aim of our investigations is the detailed petrographic and geochemical examination of the nephrite artefacts found on Hungarian sites, and locating the origin of the raw materials. The material was basically investigated by non-destructive methods (PGAA, non-destructive SEM-EDX) to avoid invasive analyses on the complete artefacts. In this study, preliminary results are presented.

Based on their chemical composition, most of the artefacts measured so far belong to the S-type (serpentinite-related) nephrite deposits.

On the basis of their microscopic and mineral-chemical features, the artefacts investigated so far can be divided into five raw material types: (1) almost pure tremolite-nephrite with only a few fine grained magnetite or ilmenite grains and some pseudomorphs after pyroxenes; (2) almost pure actinolite-nephrite with only a few very fine grained magnetite or ilmenite grains; (3) almost pure tremolite-nephrite with a few chlorite and some pseudomorphs after pyroxenes; (4) actinolite-nephrite, with chlorite, relict clinopyroxenes (diopside), pseudomorphs after pyroxene, spinels and garnets. Magnetite, limonite, apatite and titanite also occur. There is a typical association of chromite spinel and grossular garnet in this type; (5) actinolite-nephrite – sometimes also tremolite - with chlorite, relict clinopyroxenes and spinel (chromite), but garnet is missing.

We have already built a database of the possible nephrite raw material sources of Europe - descriptions and survey data: mineral-, textural- and chemical composition (Péterdi et al., 2014.).

On the basis of our investigations the most probable raw-material sources are the following: type (1) and (3) belongs to Jordanów, Poland. The provenance of the other types is not so clear, but we have candidates from the Swiss Alps. There is a nephrite type in Jordanów, that looks very similar to type (4), but the main amphibole type is tremolite in all Jordanów samples, while actinolite in the type four artefacts.

Kivonat

Magyarországi régészeti leletanyagban nefritet csak kis számban ismerünk, főként csiszolt kőeszközök nyersanyagaként és többnyire dunántúli lelőhelyekről.

Munkánk célja a nefrit kőeszközök ismertetése, részletes kőzettani és geokémiai vizsgálata; nyersanyaguk szerinti csoportosítása, illetve a nyersanyagok származási helyére vonatkozó következtetések levonása. A kőeszközök épségének megőrzése érdekében csak roncsolásmentes vizsgálatokat alkalmaztunk (PGAA, roncsolásmentes SEM-EDX). Cikkünkben előzetes eredményeket közlünk.

Teljes kőzet kémiai összetételük alapján a már megvizsgált nefrit kőeszközök nagy része S-típusú (szerpentinesedett utrabázisos kőzetes-típusú) nefrit-lelőhelyekhez köthető.

Eredményeink alapján az eddig megvizsgált kőeszközök – nyersanyaguk mikroszkópos és ásványkémiai jellemzői alapján – öt csoportba sorolhatók: (1) szinte "tiszta" tremolit-nefrit, csak apró magnetit, ritkán ilmenit szemcséket tartalmaz, valamint piroxén utáni pszeudomorfózákat; (2) szinte "tiszta" aktinolit-nefrit, csak apró magnetit, ritkán ilmenit szemcséket tartalmaz; (3) szinte "tiszta" tremolit-nefrit, kevés klorittal és piroxén utáni pszeudomorfózával; (4) aktinolit-nefrit, klorittal, relikt klinopiroxénekkel (diopsziddal), piroxén utáni pszeudomorfózákkal, spinellekkel és gránátokkal. Magnetit, limonit, apatit és titanit szintén megtalálható ebben a típusban. Nagyon jellegzetes képet mutat a spinellek (krómit) és gránátok (grosszulár) együttes megjelenése. (5) aktinolit-nefrit (néhány esetben tremolit is megtalálható benne) klorittal, relikt klinopiroxénekkel és spinellekkel (krómittal); a gránátok ebből a típusból hiányoznak.

Az európai nefritlelőhelyek nyersanyagtípusairól – irodalmi adatok és általunk vizsgált minták alapján - általunk készített adatbázist (Péterdi et al. 2014) használtuk a lehetséges nyersanyagforrások azonosításához.

Az eddig megvizsgált nefrit kőeszközök makroszkópos megjelenése, ásványos összetétele, szövete, valamint teljes kőzet kémiai összetétele alapján a legvalószínűbb nyersanyag forrásterületek a következők:

Az (1) és (3) típus nyersanyagának forrása Jordanów (Alsó-Szilézia, Lengyelország). A többi típus nyersanyageredete még nem tisztázott, de egyes svájci lelőhelyek jellemzői nagy hasonlóságot mutatnak ezekkel a típusokkal. Az egyik jordanówi nefrit-típus nagyon hasonlít a (4) típusra, de a fő kőzetalkotó amfibol Jordanówban a tremolit, míg a 4. típusba tartozó régészeti leletek esetében aktonolit.

KEYWORDS: NEPHRITE, POLISHED STONE TOOL, PROVENANCE STUDIES

KULCSSZAVAK: NEFRIT, CSISZOLT KŐESZKÖZ, PROVENIENCIA-VIZSGÁLATOK

Archaeological background, aim of the study

Nephrite is mainly known in prehistoric context as raw material of polished stone tools. It is present among archaeological finds in Hungary only in few numbers. They are known mostly from Transdanubian archaeological sites, primarily in the material of old surface collections like the Miháldy Collection of Veszprém (Szakmány et al. 2001) and the Ebenhöch Collection of the Hungarian National Museum (Friedel 2008; Friedel et al. 2008; Friedel et al. 2011) that were systematically investigated from a petroarchaeological point of view.

The polished stone tools of these old collections typically cannot be identified according to either age or culture because of lack of information concerning their provenances (Horváth 2001). Recent surveys connected to modern excavations and archaeometrical programs (e.g. JADE 2 project), however, yielded some nephrite artefacts with known locality - and some cases with known context - as well (Table 1., Fig. 1.).

We have to emphasize that we do not know any nephrite sources in the Carpathian Basin, therefore the raw material must have been transported from distant source(s).

The general aim of our investigations is the detailed petrographic and geochemical examination of the

nephrite artefacts found at Hungarian sites, and locating the origin of the raw materials.

In this study preliminary results are presented (nephrite artefacts of the Miháldy Collection, and artefacts with known locality, see **Table 1**.) further we are planning to complete our data with the results of the investigation of the artefacts of the Ebenhöch Collection, and other archaeological assemblages, too.

Methods

Due to the complete artefacts, the material of the nephrite artefacts was basically investigated by non destructive methods. On some broken pieces, however, we could also perform destructive The first method was always analyses. macroscopical description. Results were completed with petrographic microscopical and geochemical analyses. For bulk-rock chemistry, Prompt-gamma activation analysis (PGAA) was used; for mineral chemistry Electron Probe Micro-Analysis performed in a Scanning Electron Miscroscope (EPMA, SEM-EDX)] was applied. The results were compared to published data (Péterdi et al. 2014).

PGAA measurements were performed at the HAS Centre for Energy Research, at the PGAA and NIPS-NORMA stations of the guided external cold neutron beam of the Budapest Neutron Centre.

Table 1.: Samples and analyses.

(Abbreviations: TLBC – Transdanubian Linear Pottery Culture; EH – Ebenhöch Collection; LDM – Laczkó Dezső Museum, Veszprém; MH – Miháldy Collection; HNM – Hungarian National Museum; PGAA - prompt-gamma activation analysis; RRM – Rippl-Rónai Museum, Kaposvár; SEM-EDX - non-destructive SEM-EDX, 'original surface investigation method'; SM – Savaria Museum, Szombathely)

1. táblázat: Minták és vizsgálatok

(Rövidítések: TLBC – Dunántúli Vonaldíszes Kerámia kultúra; EH – Ebenhöch gyűjtemény; LDM – Laczkó Dezső Múzeum, Veszprém; MH – Miháldy Gyűjtemény; HNM – Magyar Nemzeti Múzeum; PGAA - promptgamma aktivációs analízis; RRM – Rippl-Rónai Múzeum, Kaposvár; SEM-EDX – roncsolásmentes elektronmikroszkópos vizsgálat, 'eredeti felszín' vizsgálat; SM – Savaria Múzeum, Szombathely)

Sample	Locality	Culture	ID / inventory number	PGAA	SEM- EDX
Alattyán	Alattyán, Vízköz	Tisza Culture	private collection of Gy. Kerékgyártó (Jászberény)	+	+
Balatonőszöd	Balatonőszöd, Temetői-dűlő	Baden Culture IIB- III	B-991. pit (RRM, not inventorised)	+	+
Balatonszemes	Balatonszemes- Szemesi berek	Baden Culture or TLBC	18.3/696.1 (RRM)	+	+
Ikervár	Ikervár, Péterfa major	Baden Culture	4.12.5/3, 217 obj. (SM)	+	+
Gérce, Nemeshegy	Gérce, Nemeshegy alja	uncertain (field survey)	8.10.6/3 (SM)	+	+
Gérce, Római villa	Gérce, Római villa II.	uncertain (field survey)	Gy. 2004 (SM)	+	+
Lukácsháza	Lukácsháza	unknown	4150 (RRM)	+	+
Orci	Orci	unknown	4004 (RRM)	+	+
Szombathely	Szombathely, Táncsics M. u. 44.	Baden Culture	70. gödör (SM)	+	+
MH 1006	unknown	unknown	55.1006 (LDM)	+	-
MH 1010	unknown	unknown	55.1010 (LDM)	+	-
MH 1097	unknown	unknown	55.1097 (LDM)	+	+
MH 1109	unknown	unknown	55.1109 (LDM)	+	-
MH 1144	unknown	unknown	55.1144 (LDM)	+	+
MH 1145	unknown	unknown	55.1145 (LDM)	+	-
MH 1149	unknown	unknown	55.1149 (LDM)	+	+
MH 1152	unknown	unknown	55.1152 (LDM)	+	+
MH 1203	unknown	unknown	55.1203 (LDM)	+	+
MH 1275	unknown	unknown	55.1275 (LDM)	+	-
EH 248	unknown	unknown	300/876.248. (HNM)	+	-



Fig. 1.: Studied artefacts and sites (abbreviations: 1. Lukácsháza; 2. Szombathely; 3. Ikervár; 4. Gérce; 5. Balatonszemes; 6. Balatonőszöd; 7. Orci; 8. Alattyán; Ebenhöch – Ebenhöch Collection, Hungarian National Museum, Budapest; Miháldy - Miháldy Collection, Laczkó Dezső Museum, Veszprém)

1. ábra: A vizsgált régészeti leletek és lelőhelyek (rövidítések: 1. Lukácsháza; 2. Szombathely; 3. Ikervár; 4. Gérce; 5. Balatonszemes; 6. Balatonőszöd; 7. Orci; 8. Alattyán; Ebenhöch – Ebenhöch Gyűjtemény, Magyar Nemzeti Múzeum; Miháldy - Miháldy Gyűjtemény, Laczkó Dezső Múzeum, Veszprém)

The thermal equivalent intensities at the target positions are $7.75 \times 10^7 \text{ cm}^{-2} \text{s}^{-1}$ and $2.75 \times 10^7 \text{ cm}^{-2} \text{s}^{-1}$, respectively. The PGAA station is suitable to study objects that are not bigger than 5 cm×5 cm×10 cm. while at NIPS-NORMA station one can investigate objects up to 20 cm×20 cm×20 cm. In case of the second station, it is possible to perform 2D or 3D imaging with neutrons besides the bulk elemental investigations; moreover, elemental mapping of the objects can be implemented, too.. The cross-section of the neutron beam can be adjusted between 5 mm² an 400 mm², and a selected part of the objects can be studied (Kis et al., 2015). The neutrons can enter into the material in more cm depth, thus PGAA is regarded as bulk method. A precisely calibrated HPGe-BGO detector system and а 16k multichannel analyser are used for detection of prompt gamma photons. The experimental set-up was described by Szentmiklósi et al. (2010). Quantitative determination of chemical elements is made on the bases of the PGAA library that has been compiled during standardisation

measurements (Révay, 2009). In case of most geological samples, all major components and a few trace elements (B, Cl, Sc, V, Cr, Nd, Sm and Gd) can be quantified. PGAA is unique in the non-destructive determination of H and B in low concentrations.

Non-destructive mineral-chemical examination of the surface of the artefacts (SEM-EDX) was performed with the 'original surface investigation method' (Bendő et al., 2012) at the Department of Petrology and Geochemistry of the Institute of Geography and Earth Sciences of the Eötvös University (ELTE). The instrument is an AMRAY 1830 type SEM, with an EDAX PV9800 energy dispersive spectrometer. Conditions of analyses: accelerating potential: 20kV; beam current: 1nA; focused electron beam (diameter: ~50 nm). Fairly large samples can be placed into the sample chamber of this electron microscope so the stone implements could be placed into the sample chamber without intrusive preparation.



Fig. 2.: Bulk-rock chemistry (PGAA-results): studied artefacts. (Abbreviations: MH - Miháldy Collection, Laczkó Dezső Museum, Veszprém; EH - Ebenhöch Collection, Hungarian National Museum Museum, Budapest)

2. ábra: Teljes kőzet kémiai összetétel (PGAA-eredmények): vizsgált régészeti leletek. (Rövidítések: MH - Miháldy Gyűjtemény, Laczkó Dezső Múzeum, Veszprém; EH - Ebenhöch Gyűjtemény, Magyar Nemzeti Múzeum)

Results - macroscopic features

The colour of the studied nephrite artefacts is varied: white, shades of pale green to dark green. The surface of the white ones - as an effect of the weathering and burying - may become light or dark grey, in some cases bluish gray coloured. On some artefacts two or more basic colours can appear like white-pale green, pale bluish green colours. In the surface of every colour-variant reddish brown patches can occur, but these patches are absent in the interior parts (or on broken surfaces) of the artefacts. The characteristic texture is based on fibrous amphibols: these fibres are wavering and making fan-shaped structures ('nephrite-texture'). (**Fig. 1.**)

Results - bulk rock chemistry (PGAA)

Nephrites can be classified into two groups according to their formation: the first one is formed by contact metasomatism between intermediateacidic intrusions and dolomitic marbles (dolomiterelated deposits, D-type), the other type is formed by contact metasomatism between serpentinite and magmatic body (serpentinite-related deposits, S-type) (Zhang et al 2011.).

There is a significant difference in the chemical composition between the two types. The difference can be best expressed with the $Mg^{2+}/(Mg^{2+}+Fe^{2+(3+)})$ mol-ratio, witch varies between 0.930-1 in D-type and varies between 0.860-0.930 in S-type nephrites (Zhang et al 2011.). The marking of the boundary between the two types, however, was probably arbitrarily, because among the Chinese geological samples there were no values observed between 0.920-0.950.

Most of the artefacts measured so far belong to the S-type (**Fig. 2.**), only three belongs to the D-type (but the values of these not exceed 0.950). It must be pointed out that the geochemical boundary between the two nephrite types is not so sharp: because of the deviations of the non-destructive methods, the samples with values close to the boundary need individual consideration and comparison with results of other methods of investigations.



Fig. 3.: A-B) BSE-photomicrograph: rock texture (type 1). (Abbreviations: trem - tremolite, lim - limonite)
3. ábra: A-B) Visszaszórt elektronkép fotó: szöveti kép (1. típus). (Rövidítések: trem - tremolit, lim - limonit)



Fig. 4.: A) BSE-photomicrograph: rock texture (type 2). (Abbreviations: act – actinolite, lim – limonite) B) BSE-photomicrograph: rock texture (type 3). (Abbreviations: trem – tremolite, chl – chlorite)

4. ábra: A) Visszaszórt elektronkép fotó: (2. típus). (Rövidítések: act – aktinolit, lim – limonit)
B) Visszaszórt elektronkép fotó: szöveti kép (3. típus). (Rövidítések: trem – tremolit, chl – klorit)

Results - microscopic features (mineralcompositon, fabric etc.), mineralchemistry (non-destructive SEM-EDX)

Some of the artefacts (see **Table 1.**) were investigated with non-destructive SEM-EDX, with the '*original surface method*'. Based on these data, the artefacts investigated so far can be divided into five types.

The first type is almost pure tremolite-nephrite with only a few small magnetite, limonite or ilmenite grains and some pseudomorphs after pyroxene (**Fig. 3**.).

The second type is almost pure actinolite-nephrite with only a few minute magnetite, limonite or ilmenite grains (**Fig. 4A.**).

The third type is almost pure tremolite-nephrite with a few chlorite and some pseudomorphs after pyroxene (**Fig. 4B**.).

Type four is actinolite-nephrite, with chlorite, relict clinopyroxenes (diopside), pseudomorphs after pyroxene and relatively large enclosed minerals: spinels and garnets (**Fig. 5.**). In relict diopsides tremolite may occur as veins (**Fig. 5A.**). Magnetite, limonite, apatite and titanite also occur. There is a typical association of chromite spinel and grossular garnet in this nephrite type (**Fig. 5C-D**.).

Beside the major mechanism of nephrite formation (diopside recrystallization to tremolite / actinolite), there is another mechanism: garnet retrogression with chlorite and spinel as common products (Gil et al. 2015). In this type the relict garnets are still existing.



b) visszászort elektronkép 1010. szöven kép (4. upus). (Kövidnesek, act – aktinont, pspx – piroxen utal pszeudomorfóza)

C) Visszaszórt elektronkép fotó: (4. típus). (Rövidítések: grs – gránát, chr – krómit, ap – apatit)

D) Visszaszórt elektronkép fotó: szöveti kép (4. típus). (Rövidítések: grs – gránát, chr – krómit, act – aktinolit)

Type five is actinolite-nephrite with chlorite, relict clinopyroxenes and spinel (chromite). ; Tremolite was also measured, but garnet is missing (**Fig. 6.**).

In this type the relict garnets are missing, but spinel with chlorite are typical.

The main amphibole type (tremolite or actinolite) varies, even within one sample (**Fig. 7.**).

Possible source regions

Its pleasing aesthetic appearance and its toughness, ensured by the compact fabric consisting of interweaving and interlocking thin, fine amphibole fibres, makes nephrite an excellent raw material for polished stone tools, and so it became a widespread raw material all over Europe in the Neolithic Period and the Bronze Age.



Fig. 6.: A) BSE-photomicrograph: rock texture (type 5). (Abbreviations: act – actinolite, chl – chlorite, mt – magnetite)

B) BSE-photomicrograph: rock texture (type 5). (Abbreviations: chr – chromite, chl – chlorite)

6. ábra: A) Visszaszórt elektronkép fotó: szöveti kép (5. típus). (Rövidítések: act – aktinolit, chl – klorit, mt – magnetit)

B) Visszaszórt elektronkép fotó: szöveti kép (5. típus). (Rövidítések: chr - krómit, chl - klorit)



Fig. 7.: Main amphibol type (non-destructive SEM-EDX results): studied artefacts (diagram modified after Leake et al. (1997))

7. ábra: A fő kőzetalkotó amfibol típusa (roncsolásmentes SEM-EDX eredmények): régészeti leletek. (Amfibol-besorolás Leake et al. 1997 alapján.)

It was; however, not used (as raw material) in large quantities because of the small size and limited occurrence of the nephrite bodies.

The identification of the provenance for nephrite artefacts is rendered more difficult because far too many 'green-colour' rocks have been named nephrite in the course of time. For example, the labelling of amphibole-types has changed several times so in earlier literature we are to look for 'grammatite', even for 'hornblende' and not necessarily for tremolite or actinolite. Geological occurrences of nephrite are fairly rare in Europe. Known nephrite sources are the following: the Alps (on the territory of Switzerland, Italy, France, Germany and Austria), the Apennines (Liguria), the Harz Mts. and Scandinavia; also in the metamorphosed basic and ultrabasic complexes of the boundaries of the Bohemian Massif. Nephrite occurs among glacial erratics carried there by ice from Scandinavia in periods of glaciation on the Rügen-island, and in the environs of Potsdam and Leipzig (Gunia 2000). (**Fig. 8.**)

The so-called 'Mur Nockerls' – nephrite-gravels, nephrite-boulders originating from the alluvium of the river Mur between Leoben and Graz – also deserve mention. The Mur flows into the Drava so it is nearer the Carpathian Basin than the other known provenances. Along its upper course (before it is breaking through the Glein Alm) several serpentinized rock masses occur, but the parent material of the nephrite-gravels is unknown as yet (Giess, 2005). The 'Mur Nockerls' belong to the tremolite-nephrites (Giess, 2005), but detailed descriptions or analyses are not available.

Since a significant number of polished tools made of nephrite have been found on the Balkan Peninsula a raw material source for nephrite has Data available from two probable source areas consist of descriptions and survey data and concerns mineral-, textural- and chemical composition. These two regions are the Swiss Alps (canton Graubünden / Grisons) and the Boundaries of the Bohemian Massif. (Meyer 1884; Traube 1885a, 1885b, 1887; Heierli 1902; Sachs 1902; Kalkowsky 1906; Welter 1911a, 1911b; Schneider 1912; Staub 1915, 1917; Schmidt 1917; Preiswerk 1926; Dietrich and de Quervain 1968; Gunia 2000; D'Amico et al. 2003; Giess 2003, Gil 2013, Gil et al. 2015)

Here we show only a few examples (**Fig. 9-11.**). For the database and detailed descriptions see Péterdi et al. 2014.

From Polish Lower Silesia we have also new samples from Jordanów and Złoty Stok, with modern chemical analysis data (PGAA, EPMA and non-destructive SEM-EDX) (Gil 2013; Péterdi et al. 2014). Many types of nephrite can be found on these localities.



Fig. 8.: Known nephrite raw-material sources in Europe (yellow sings - nephrite sources with detailed descriptions and survey data; orange signs - nephrite sources without detailed data).

8. ábra: Ismert nefrit nyersanyag-források Európában (sárga jelzés – nefrit lelőhelyek részletes leírásokkal és elemzési adatokkal; narancs jelzés - nefrit lelőhelyek részletes adatok nélkül).


Fig. 9.: Example from our "nephrite-database" (Péterdi et al. 2014): Cuolms

9. ábra: Példa a "nefrit-adatbázisból" (Péterdi et al. 2014): Cuolms



Fig. 10.: Example from our "nephrite-database" (Péterdi et al. 2014): Mühlen, Salux **10. ábra:** Példa a "nefrit-adatbázisból" (Péterdi et al. 2014): Mühlen, Salux

26	Jordanów (Jordansmühl in Schlesien)			
Colour:	white, greenish-creamy, bright green (light green) to dark green, greyish-blue, blue, pink	Germany	Poland	В
Fabric	compact fabric or foliated or schistose		S OF	2
(microscopic fabric):	(typical non-directional, fibrous, parallely fibrous)			
Geochemical data:	+			1.5.0
Formation type:	S	المر المر المر المر المر المر المر المر	27 / 27	ART
Main amphibol type:	Т	X	111	1
Associated and	1: pure nephrite	25		(a
enclosed minerals:	2: pseudomorphs after pyroxene	S Cze	ch Republic	7
	3: diopside, chlorite (common)	A De la Caracteria		7
	4: grossular, hydrogrossular, prehnite, antigorite, Cr-spinel, titanite, apatite, monacite, zircon	h.	m	<i>, , , , , , , , , ,</i>
References:	Traube 1885a, 1885b; Sachs 1902; Gunia 2000; Mazur et al. 2006; Gil 2013			and the second

Fig. 11.: Example from our "nephrite-database" (Péterdi et al. 2014): Jordanów

11. ábra: Példa a "nefrit-adatbázisból" (Péterdi et al. 2014): Jordanów

Evaluation of the results, and the most probable sources

Fig. 12. shows the bulk rock chemical data of the measured artefacts and the possible raw-material sources. We used data of analyses from the literature and from our new investigations.

Fig. 13. shows the main amphibole types of Jordanów and Zloty Stok samples together with the artefacts.

On the basis of macroscopic appearance, mineral composition, fabric character and bulk chemical composition the most probable raw-material sources of the so far investigated artefacts are the following:

Type one and three belongs to Jordanów. The provenance of the other types are not so clear, but we have candidates from the Swiss Alps. (Table 2., Fig. 14.) Unfortunately about the source nearest to the Carpathian Basin i.e., the 'Mur Nockerls', there are no detailed descriptions or analyses available.

There is a nephrite type in Jordanów, that looks very similar to type four (for example the chromite-

grossular association), but the main amphibole type is tremolite in all Jordanów samples, while actinolite in the type four artefacts.

For green nephrite-types of Jordanów, however, the green colour can be associated with not only the small amount of chlorite in them, but also due to the presence of actinolite, occurring in small veins (Gil et al. 2015). So it is probable that the raw material of this type (type four) also belongs to Jordanów.

It must be pointed out that stone axes made of Jordanów nephrite were found about 15 km north of Jordanów (Neolithic); in the central part of Poland (Danubian culture); and also in Upper Silesia (Funnel Beaker culture, Corded Ware culture) (Foltyn et al., 2000; Gunia, 2000) and probably from a Late Neolithic Silezian site (Přichystal et al., 2012). We know cultural connections between this nephrite source and the Carpathian Basin in the Late Copper Age (Baden Culture) (Přichystal 2000).





Fig. 12.:

Bulk-rock chemistry (PGAA-results): studied artefacts; nephrite raw-material sources (PGAA-results and data from literature, references in the text).

12. ábra:

Teljes kőzet kémiai összetétel (PGAAeredmények): vizsgált régészeti leletek, nefritnyersanyagforrások (irodalmi adatok és saját PGAA-mérések, hivatkozásokat ld. a szövegben).



Fig. 13.: Main amphibol type: studied artefacts (non-destructive SEM-EDX results); nephrite raw-material sources (destructive and non-destructive SEM-EDX results, references in the text). (Diagram modified after Leake et al. (1997))

13. ábra: A fő kőzetalkotó amfibol típusa: régészeti leletek (roncsolásmentes SEM-EDX eredmények); nefritnyersanyagforrások (roncsolásos és roncsolásmentes SEM-EDX eredmények, hivatkozásokat ld. a szövegben). (Amfibol-besorolás Leake et al. 1997 alapján.)



Fig. 14.: Most probable raw-material sources (preliminary results).14. ábra: Legvalószínűbb nyersanyagforrások (előzetes eredmények).

Table 2.: Probable raw-material source-regions

2. táblázat: A nyersanyagtípusok valószínű származási helye

Туре	Probable source
type 1: "pure" tremolite + minor magnetite, limonite, ± ilmenite ± pseudomorphs after pyroxene	Jordanów (Jordansmühl in Schlesien) (Lower Silesia, Poland)
<pre>type 2: "pure" actinolite + minor magnetite, limonite, ± ilmenite</pre>	Cuolms? (Oberhalbstein (Alpi di Platta), Switzerland)
type 3: tremolite + minor chlorite ± pseudomorphs after pyroxene	Jordanów (Jordansmühl in Schlesien) (Lower Silesia, Poland)
type 4: actinolite + chlorite, relict clinopyroxenes (diopside), pseudomorphs after pyroxene, spinel (chromite), garnet (relict grossular) + minor magnetite, ilmenite, ± apatite, ± titanite	Val da Faller (Faller valley): Mühlen (Mulegns), Forschella-peak, (Oberhalbstein (Alpi di Platta), Switzerland) Salux? (Oberhalbstein (Alpi di Platta), Switzerland) Jordanów??? (Jordansmühl in Schlesien) (Lower Silesia, Poland)
type 5: actinolit and tremolite + chlorite, relict clinopyroxenes, spinel (chromite) + minor magnetite garnet is absent	??? (Crap Farreras?) (Oberhalbstein (Alpi di Platta), Switzerland)

Moreover, it has been proved that serpentinites containing nephrite-bodies from this area were used to make stone-axes in the Neolithic Age (Majerowicz et al., 2000; Skoczylas et al., 2000). territory's serpentinite quarried The was (Wojciechowski, 1995) and polished stone tools made of them got fairly far away (even to 340 kms) (Přichystal and Šebela, 1992.; Skoczylas et al., 2000). In the time of the Corded Ware culture, there was a centre for extracting and processing serpentinite on the territory; its most important products were the so-called 'Ślęża-type' shaft-hole axes (Skoczylas et al., 2000).

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RAPID NON-DESTRUCTIVE X-RAY DIFFRACTION INVESTIGATION OF POLISHED GREENSTONE TOOLS ZÖLDKŐ TÍPUSÚ CSISZOLT KŐESZKÖZÖK GYORS RONCSOLÁS MENTES RÖNTGENDIFFRAKCIÓS VIZSGÁLATA

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Abstract

During the research of polished stone tools we may frequently find rare, valuable and unique exemplars. These qualities in many cases are determined by the rock type the tool has been made from. Among Hungarian findings, high pressure metamorphic rock made tools are rare. These cannot be identified based on their macroscopic appearance, colour or texture. The precise mineralogical investigations needed for their identification can be made by X-ray diffraction. Since we have to deal with unique pieces, non-destructive technique must be applied. On a laboratory diffractometer this can be done with the use of Göbel mirror, in parallel beam geometry. With a scintillation detector measurement times up to 8 hours are necessary, but position sensitive detectors allow recording times of minutes. Our measurements were carried out with 15 minutes recording. Instrumental alignment and precision was tested with the use standards. Identification of rock forming components was possible with accuracy using Search/Match algorithm. Measurement times were reduced to even 1 minute, depending on measured surface and rock type. Our measurements revealed the existence of eclogite type omphacitic and jadeite bearing rocks, amphibolites and nephrites, chlorite schist and hornfels type contact metamorphite. According to our observations, textural features and orientation patterns can be extracted, if necessary.

Kivonat

A csiszolt kőeszközök kutatása során gyakran találkozunk ritka, értékes és egyedi példányokkal. Ezeket a tulajdonságokat sokszor a kőzet típusa határozza meg, amiből készült az eszköz. Magyarországi leletek között ritkák a nagy nyomású metamorfitokból készült balták és vésők. Makroszkópos megjelenésük, színük és szövetük alapján nehezen határozhatók meg. Az azonosításukhoz szükséges pontos ásványtani vizsgálatát röntgen diffrakcióval lehet végezni. Mivel egyedi példányokról beszélünk, roncsolásmentes eljárást kell alkalmazni. Laboratóriumi pordiffraktométeren ezt az eljárást Göbel tükörrel, párhuzamos nyaláb geometriában tudjuk elvégezni. Szcintillációs detektorral akár 8 órás mérésekre is szükség lehet, de helyzetérzékelő detektorral percek alatt elvégezhető a mérés. Vizsgálatainkat átlagosan 15 perces méréssel végeztük. A műszer beállításait és pontosságát standardokkal ellenőriztük. A kőzetalkotó ásványok azonosítását nagy pontossággal el tudtuk végezni a Search/Macth algoritmus alkalmazásával. Mintafelülettől és anyagtípustól függően 1 perces mérési időt is elegendőnek találtunk az azonosításhoz. A méréseink során eklogit típusú omfacitos és jadeites kőzeteket, amfibolitokat és nefriteket valamint kloritpalát és szaruszírt típusú kontakt metamorfitot azonosítottunk. Megfigyeléseink szerint, ha szükséges, szövet-szerkezeti, szöveti orientációs adatokat is ki tudunk nyerni.

KEYWORDS: NON-DESTRUCTIVE XRD, PYROXENITE, OMPHACITE SEARCH/MATCH, NEPHRITE, XRD

KULCSSZAVAK: RONCSOLÁSMENTES XRD, PIROXENIT, OMFACIT SEARCH/MATCH, NEFRIT, XRD

Introduction

X-ray diffraction investigations make the basis of materials science and related research fields since many decades. May we speak of single crystal, powder or micro diffraction, the structure (crystalline or not) of materials is mainly resolved by using X-rays. This is due to the elaboration of many laboratory scale instrument types and high performance laboratory X-ray sources. Perhaps the most widespread application is that of the Bragg-Brentano (or parafocusing) powder diffractometers, which gives the best resolution – intensity geometry (Brentano 1946). This is used to obtain data for crystal structure solution, quantitative mineralogical evaluation or simply identification of sample components. Powder diffractometers are also suitable for the investigation of "block" samples, if the means of sample surface quality and alignment requirements are satisfied (see metallurgical applications, Kocks et al. 2000). For a Bragg-Brentano diffractometer, this would mean a polished – planar surface, which can be adjusted at least with \pm 0.01 mm to the sample height reference plane of the goniometer circle.

This problem was overcome by developing parallel beam geometry (Schuster & Göbel 1995), with X-

ray optical attachments such as a Göbel mirror (Deslates et al. 1997). This way, surface roughness and sample height error or displacement is not a problem (Holz et al. 2000), and measurements can be carried out on any solid material, that can be adjusted in the sample chamber of the apparatus. By this application the X-ray diffraction evolved into a totally non-destructive analytical method.

In archaeometry, we encounter frequently valuable objects that cannot be investigated by traditional analytical methods (e.g. powder diffraction, thin sections, and solution based chemical analysis). Non-destructive XRD is currently the only available technique to investigate properties related to crystalline structure of such materials. The Göbel mirror solution of Bruker AXS Ltd. has been applied in several archaeological cases in the last decade, for various artefacts (Duran et al. 2008), glasses, ceramics (Kristály & Kovács 2011) and even stone tools (Chiari et al. 1996). We also have investigated the reliability of this technique by using powder, micro powder and sliced specimens of various stone tools. As it was expected, identification of crystalline components is possible, even at accessory minerals level. But several major issues may lead to false identifications. One of these is preferred orientation of crystals (or grains) mainly of fibrous and platy minerals. Also the base line profile is highly influenced by the shape of analyzed surface, permitting erroneous amorphous phase observations. But one of the major setbacks is measurement time, which is required to be up to 8 hours in most cases. This issue can be partly overcome with the use of position sensitive detectors, which reduce measurement times even to 1/10, although sample surface alignment requires more attention.

As we can learn from papers dedicated to stone tool research (Gan et al. 2010, Giustetto et al. 2008), many analytical techniques are useful and applicable non-destructive. as Chemical information is easier to obtain in a non-destructive way, and even mineralogical information can be extracted e.g. by Raman microscopy (Smith & Gedron 1997), but crystallographic data can only obtained by diffraction methods, like laboratory XRD. Microscopic and micro spectrometric solutions work well in the cases were grains and textures are distinguished. But if we have greenstone tools. with macroscopically homogeneous structure, we barely can make difference between omphacitite, jadeitite and other green stone materials. In these cases, even local (SEM+EDS, Bendő et al. 2013) and more or less bulk (PGAA, Szakmány & Kasztovszky 2004)

chemical results may be hard to interpret. The aim of this study is to show how useful non-destructive XRD technique can be, applied to identify the rock type of rare and special stone tools, with easy and simple measurement and evaluation procedure.

Although application of Search/Match evaluation technique and the use of ICDD PDF databases is a "last century's" invention, the success of evaluation depends much on how the user can maximize data quality and searching criteria. Relative peak intensities of searched phases are as important in Search/Match evaluation, as peak positions. But preferred orientation usually produces anisotropic distortion of pattern, i.e. several peaks will be measured smaller, while others higher, than theoretical values. When working with powder pattern, these peaks can easily be identified, and intensity distortions either disregarded e.g. in multiple iteration Search/Match, or corrected by profile or pattern fitting. In both cases we can stop with evaluation without seeking partial solutions in vain. In the case of block samples, where orientation of crystals is determined by rock texture, we may observe one or just a few peaks of one phase. In these cases finding that phase is improbable, and correction of intensities may also become impossible. None the less, reduction of measurement and data evaluation time is crucial in the lack of stone tools research dedicated laboratories.

Materials and methods

A Bruker D8 Advance powder diffractometer was used, equipped with Cu-K_{α} source (40kV, 40mA), Göbel mirror and Våntec1 PSD detector, on a 250 mm radius goniometer, with 50 cm sample plane height (allowing large sample introduction). Since the Göbel mirror removes K_{β} components of incident beam, filters or monochromators are not required. The Göbel mirror has $< 0.25^{\circ}$ primary equatorial divergence, thus an instrumental broadening of 0.145° (2 θ) can be obtained, using 0.6 mm exit slit and 1 mm detector slit. Instrumental broadening as a measure of resolution means the minimal separation that can be observed between two neighbouring peaks on an ideally crystalline material. This resolution is maintained with Våntec1 on smooth and plain surfaces, slightly increasing at high angles, up to 0.150° (20). For non-destructive application, change in resolution and displacements were verified on the NIST1976 α -Al₂O₃ calibration standard. The instrument is equipped with primary beam shield and detector side beam stop (Fig. 1.).



Fig. 1.: A: general view of the goniometer with sample mounted (1 - X-ray source, 2 - Göbel mirror, 3 - Vantec-1 PSD detector, 4 - incident beam cut-off knife edge, 5 - diffracted beam low angel cut-off shield), B and C: view on the analyzed surface, marked with an Al-foil

1. ábra: A: a goniométer és beállított minta általános nézete (1 – sugárforrás, 2 – Göbel tükör, 3 – Vantec-1 PSD detektor, 4 – primer nyaláb szabályozó késlemez, 5 – diffraktált nyaláb alacsony szögű szabályozó késlemez).

Several green stone tools were measured, on one or more selected surface spots, to identify mineralogical constituents. Description of tools by the means of macroscopic features, their origin, and petrographic description is given in Péterdi et al. (2015) and Bendő et al. (2014). In lack of dedicated sample mounting system, the tools were mounted and aligned by the use of an optical microscope with dismounted stage with micro adjustment screws and rotate-tilt stage (Fig. 1.). The only required sample preparation was the cleaning with acetone of the selected plane and possibly smooth surfaces to be analysed. The selected surface was aligned in the sample plane in reference to the primary beam shield, since it is a fixed and highly centred part of the apparatus (Fig. 1A and 1B).

Evaluation of recorded patterns was done in Bruker Diffrac*Plus* EVA software, applying Search/Match algorithm for ICDD PDF2 (2005) database, on Fourier-polynomial smoothed (Wells & Brown 2009) and background removed curves. In some cases $K_{\alpha 2}$ removal by Rachinger algorithm (Rachinger 1948, for an experimenttaly determined $K_{\alpha 1}/K_{\alpha 2}$ ratio) also proved useful, but it's not a necessary data reduction step for successful evaluation. For a more detailed evaluation, Rietveld refinement can be applied, however in lack of knowledge on chemical composition this step may become unreliable.

Prior to presenting the obtained mineralogical data, a summary of instrumental alignment, set-up and testing on standards is presented. With the use of NIST 1976b standard the nature of peak shifting on specimen misalignment can be tested. The observed effect is known as parallax effect and it is arising from detector interface construction, and if it is not digitally corrected (Guinebretière 2013), it can be eliminated by goniometer and slit alignment. Its effect is eliminated in parallel beam geometry by minimizing detector window and slit opening. In our case it was found that a detector opening of 5° and a 0.1 mm beam exit slit gives reliable results (Fig. 2.). This allows us to run an 11 hours measurement in 10 minutes, without significant peak broadening arising from parallax effect. Depending on surface topography, the use of \pm 0.01° to $\pm 0.06^{\circ}$ 2 θ -scale window could be required for Search/Match.



Fig. 2.: Patterns of NIST 1976b corundum calibration standard, 1 - pattern with powder stage and 0.6 mm exit slit with 1° Vantec window (15 minutes), 2 - pattern with stative used for ND-XRD, 0.6 mm exit slit with 1° Vantec window (5 minutes), 3 - pattern with stative used for ND-XRD, 0.1 mm exit slit with 5° Vantec window (5 minutes), 4,5 - patterns with ~0.5 mm misalignment (intensity differences are due to different recording times)

2. ábra: A NIST 1976b kalibrációs standardon mért görbék, 1 – gyári asztallal mért görbe, 0.6 mm kimeneti réssel és 1°Vantec ablakkal (15 perc), 2 – mikroszkópi állvánnyal mért görbe 0.6 mm kimeneti réssel és 1°Vantec ablakkal (5 perc), 3 - mikroszkópi állvánnyal mért görbe 0.1 mm kimeneti réssel és 5°Vantec ablakkal (5 perc), 4,5 - ~0.5 mm mintasík eltolódással mért görbe (az intenzitásbeli különbségek az eltérő mérési időkből adódnak

The mineralogical composition of samples is presented in tabulated form, with short mineral description, and also the evaluated patterns are shown, for graphical confirmation of results. The selecting of mineral species or groups is not arbitrary and is based on the matching precision of measured and theoretical peak positions on the first place. If the several first returned results do not fit the patterns, searching window \pm limits must be widened. This evaluation process have helped us in the processing of several complex materials. We were able to differentiate omphacite and jadeite in greenstone tools, XRD result being in high agreement with chemical investigations (Szakmány et al. 2013). In the case of blueschist tools, we were able to match the glaucophane - ferroglaucophane series, confirmed by energy dispersive spectrometry too (Kereskényi et al. 2015).

After evaluating of XRD patterns, the investigated samples can be grouped in several major classes: (1) omphacite-jadeite eclogite type rocks with omphacitite and jadeitite compositions also, (2) amphibole dominated schists with actinolite schist and greenschist examples, (3) chlorite schists and (4) hornfels type contact metamorphic siliceous rock.

Results

(1) Omphacite-jadeite eclogite type

Before the presentation of results, a short summary on pyroxene crystal structures and XRD peaks is needed, in order to understand the evaluation process and results. Pyroxene structure is built up by silicate tetrahedra aligned in single chains along the "c" crystallographic axes bonded by oxygens, laterally – on "a" and "b" crystallographic axes – bonded by cations.

2θ d(Å)											
h	k	1	Diopside	Diopside $+1/2I_{FWHM}$	Omphacite	Jadeite	Diopside	Omphacite	Jadeite		
1	1	0	13.70	13.77	14.16	14.28	6.458	6.196	6.248		
0	2	0	19.94	20.01	20.54	20.41	4.450	4.347	4.320		
1	-1	-1	20.18	20.25	20.71	20.73	4.397	4.281	4.285		
0	2	1	26.66	26.74	27.27	27.46	3.340	3.245	3.268		
2	2	0	27.60	27.68	28.55	28.79	3.229	3.098	3.124		
2	-2	-1	29.92	29.99	30.85	30.63	2.984	2.917	2.896		
1	-3	-1	35.04	35.12	35.51	35.56	2.559	2.523	2.526		
2	0	-2	35.47	35.55	35.87	35.96	2.528	2.495	2.501		
0	0	2	35.48	35.55	35.88	36.08	2.528	2.488	2.501		
1	-1	-2	35.62	35.70	36.18	36.13	2.518	2.484	2.481		
2	2	1	35.63	35.70	36.37	37.22	2.518	2.414	2.469		
3	1	1	39.02	39.09	39.72	40.90	2.307	2.205	2.268		
0	4	0	40.52	40.59	40.80	40.96	2.225	2.202	2.210		
3	-1	-2	40.63	40.70	41.45	41.51	2.219	2.173	2.177		
1	1	2	40.63	40.70	41.46	41.82	2.219	2.158	2.176		
2	-2	-2	41.02	41.10	41.78	41.97	2.198	2.151	2.160		
0	2	2	41.02	41.10	42.14	42.18	2.198	2.141	2.143		
3	3	0	41.94	42.01	43.42	43.78	2.153	2.066	2.083		
3	-3	-1	42.45	42.52	43.96	43.80	2.128	2.065	2.058		
4	-2	-1	42.95	43.02	44.42	44.28	2.104	2.044	2.038		
4	2	0	43.57	43.64	44.97	45.49	2.076	1.992	2.014		
0	4	1	44.45	44.53	45.27	45.60	2.036	1.988	2.001		
4	0	-2	45.04	45.11	45.97	46.13	2.011	1.966	1.972		
2	0	2	45.04	45.11	46.16	46.75	2.011	1.941	1.965		
2	4	0	45.06	45.13	46.80	46.99	2.010	1.932	1.939		
1	-3	-2	46.13	46.21	47.08	47.16	1.966	1.925	1.929		
2	-4	-1	46.62	46.69	48.34	48.22	1.947	1.886	1.881		
diopside, CaMgSi ₂ O ₆ a=9.746 Å, b=8.899 Å, c=5.251 Å, β=105.63°											
	omphacite (Ca,Na)(Mg,Fe ²⁺ ,Al)Si ₂ O ₆ a=9.54-9.68 Å, b=8.57-8.90 Å, c=5.23-5.28 Å, β=105-108°										
			a=0 /10	jadeit	e Na(Al,	Fe^{3+})Si ₂ =5 210	О ₆ Å в—107	7 580			

Table 1.: Theoretical peak position for several C2/c space group clinopyroxenes, showing differences in °(2 θ) and ångströms (1/2I_{FWHM} – half value of the instrumental peak full width at half maximum)

1. táblázat: Néhány *C2/c* tércsoportú klinopiroxén elméleti csúcspozíciói, a $^{\circ}(2\theta)$ szög- és ångströmbeli különbségei ($1/2I_{FWHM}$ – a műszer félérték szélességének fele)

Table 2.: The position of most important peaks for omphacitic pyroxenes, plotted on **Fig. 3**, °(20) regions are: 1: 10-11°; 2: 13-14°; 3: 19-21°; 4: 26-27°; 5: 27-28°; 6: 29-32°; 7: 35-37°

2. táblázat: Omfacitos összetételű piroxének legfontosabb csúcsainak helye, (ld. a 3. ábrán), a °(20) tartományok: 1: 10-11°; 2: 13-14°; 3: 19-21°; 4: 26-27°; 5: 27-28°; 6: 29-32°; 7: 35-37°

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$			4°				10	00	20		21	20	21	10	11	31	02	12)2	21	31
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$			3=106.3		լՙኣՙ୳		-	2(0		.0	2.	-2	3	ς.	-	-2	-	õ	2.	T
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	1827		=5.263 Å, [20Cr0.04Ti0.0 Si1.92O6)	(%) .I.A		0.40	0.40	5.20		9.5	22.4	100	31.8	32.3	25.9	4.4	39.8	39.8	39.7	0.8
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	PDF 85-	C2/4	b=8.874 Å, c	g _{0.89} Fe _{0.08} Al ₀ . Ca _{0.76} Na _{0.10} (əulaV b		6.426	1.658	1.437		3.333	3.213	.986	2.931	2.89	2.556	2.539	2.525	2.525	2.5	2.377
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$			a=9.709 Å,	M			.77	.036	266.		.72	.74	06.	.47	16.	.07	.31	.52	.52	.89	.81
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$					əlanA		13	19	19		26	27	29	30	30	35	35	35	35	35	37
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$			=106.598	ž	(լ'ϡ'ϥ)			200	-111	020	021	220	-221	310	-311	-202	-131		-112	002	
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$	-1069	c.	=5.270 Å, β	0.60Fe _{0.25} Al _{0.2}	(%) .I.A			0.30	9.00	9.00	7.3	17.6	100	34.9	26.8	25.6	25.6		40.3	40.3	
PDF 70-1874 PDF 70-1874 $P2/h$ $P2/h$ $a=9.622$ Å, $b=8.826$ Å, $c=5.279$ Å, $\beta=106.92^{\circ}$ $a=9.646$ Å, 1 $a=9.622$ Å, $b=8.826$ Å, $c=5.279$ Å, $\beta=106.92^{\circ}$ $a=9.646$ Å, 1 $a=9.622$ Å, $b=8.826$ Å, $c=5.279$ Å, $\beta=106.92^{\circ}$ $a=9.646$ Å, 1 $a=9.622$ Å, $b=8.826$ Å, $c=5.279$ Å, $\beta=106.92^{\circ}$ $a=9.646$ Å, 1 $a=9.622$ Å, $b=8.826$ Å, $c=5.279$ Å, $\beta=106.92^{\circ}$ $a=9.646$ Å, 1 $a=9.621$ Å, $a=105.252$ Å, $b=8.826$ Å, $c=2.279$ Å, $b=106.92^{\circ}$ $a=9.646$ Å, 1 $a=9.613$ 12.5 $a=10.10$ $a=9.646$ Å, 1 $a=10.010$ $a=10.020$ $a=9.646$ Å, 1 $a=10.010$ $a=10.020$ $a=10.111$ $a=9.646$ Å, 1 $a=10.010$ $a=10.010$ $a=10.010$ $a=10.010$ $a=10.010$ $a=20.100$ 13.5 1100 $a=20.011$ 4.413 17.3 -1111 20.111 4.4 $a=20.998$ 38.4 31.0 30.70 2.21 30.00 2.548 2.552 2.548 2.552 2.522 2.522 2.552 2.552 2.5	PDF 71	C2/	b=8.824 Å, с	0.32Ca0.59Mg0 (Si2C	2007 A.D.			622	412	412	323	191	976	606	879	546	546		525	525	
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$			a=9.646 Å,	Nŝ	onlo / p			0.18 4.	.11 4.	.11 4.	6.81 3.	7.93 3.	00 2	0.70 2.	.03 2	5.22 2.	5.22 2.		5.52 2.	5.52 2.	
$\begin{array}{ c c c c c c c c c c c c c c c c c c c$)6.92°		alonA	010	110	200 15	020 20	-111 20	021 26	220 27	-221 30	310 30	-311 31	-131 35	-202 35	031	002 35	-112 35	320
PDF 70-18 P2/h a=9.622 Å, b=8.826 Å, c=5. a=9.622 Å, b=8.826 Å, c=5. a=9.622 Å, b=8.826 Å, c=5. (SigO6) etcl T1.3.5 color T1.3.5	74		279 Å, β=10	32Na _{0.508}	Į'ϡ'ų)																
PDF a=9.622 Å, b=8.826 a=9.622 Å, b=8.826 (200) region Fe _{0.475} Al ₀ . (1001 8.826 19.26 19.26 19.26 20.10 19.26 19.26 20.10 19.26 33.23 20.10 19.26 35.19 25.48 35.19 25.48 35.52 25.55	70-187	P2/n	Å, c=5.	525Ca _{0.45} Si ₂ O ₆)	(%) .I.A	0.8	13.5	1.5	17.3	17.3	3.4	11.6	100	38.4	18.7	25.2	25.2	16.0	40.5	40.5	24.1
a=9.622 a=9.6222 a=9.622 a=9.622 a=9.622 a	PDF		Å, b=8.826	Fe _{0.475} Al _{0.5} (S	əulaV b	8.826	6.371	4.603	4.413	4.413	3.323	3.185	2.979	2.898	2.879	2.548	2.548	2.542	2.525	2.525	2.519
(20) Log (00) Log (00			a=9.622		อเชินพ	0.01	3.88	9.26	0.10	0.10	6.80	7.99	9.98	0.82	1.04	5.19	5.19	5.27	5.52	5.52	5.60
				u	oigər (θ2)°	-	2 1	-	10	0	4	5 2	1	5	m	0	C	m	10	(n)	n



Fig. 3.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: 99.3.1863.

3. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: 99.3.1863



Fig. 4.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: N1/81-1938

4. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: N1/81-1938



Fig. 5.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: N11/169-1938

5. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: N11/169-1938

As a consequence of cation arrangement along the chains, monoclinic and rhombic structural varieties are formed, known as clinopyroxenes and orthopyroxenes. The difference between the two types is readily deduced from diffraction data, but not for the identification of clinopyroxene species, their classification being done on chemical composition basis (Morimoto et al. 1988). The most clinopyroxenes have space group C2/c, and the differences in the end member phases XRD peaks positions are given by the distortions caused in the unit cell with the changing size and bond lengths of the cations. For instance, diopside and jadeite have similar structures (Prewitt & Burnham 1966), and if we calculate peak positions for their theoretical unit cells (Fehér 2009, a complete collection of valid mineral species description), we find some obvious, but very small differences in peak positions, more significant if we compare also with omphacite (Table 1.) Here, if we use the instrumental broadening (obtained on NIST 1976b corundum and verified with NIST 640a silicon) to extend the angular range for single peaks, we still may have enough differences in peak positions to delimit their maxima. Intensities for each individual peaks are influenced by chemical composition, but due to the large expected distortions arising from rock texture and surface morphology the relative intensities will become unreliable in mineral identification, thus the theoretical differences are not considered here. More important feature of these structures is their ability to form solid solutions, cation substitutions and cation ordering phenomena, which produce unit cell distortions (Nestola et al. 2007) and modulated structures expected on nanometric scale. These solid solution crystals are difficult to identify by XRD, but if there exists dominant end members, or dominant species in the samples, correct identification is possible. For instance, omphacite as a 1:1 solid solution of jadeite with diopside or hedenbergite will form with a different unit cell, P2/n (Nestola et al. 2007), which also will create additional differences in peak positions. The most important is the appearance of a peak at 10.22 \pm 0.05° (2 θ) (Cu-K α source), but this peak can also be of very low intensity. Unfortunately, this nature of omphacitic solid solutions also gives room for a larger variability of peak position, in a -0.25° (20) with regard to the values in Table 1.

99.3.1863 "Gorzsa 11" tool

For 3 recorded patterns the Search/Match ($\pm 0.05^{\circ}$ window on 20-scale) returned omphacite as best match, and observed peak positions match the general clinopyroxene structure (*C2/c*, **Table 2.**). Shifting of peak positions (Nestola et al. 2007) due to cation substitution is considered, most of the omphacitic species have major peaks falling inside a ~0.10 ° (20) range, closed to the instrumental

broadening range, complicating the evaluation. Several of first matches of evaluation are plotted on Fig. 3. to demonstrate the effect of chemical composition on peak positions (Giustetto et al. 2008), according to database entries. Since jadeite or jadeite – diopside solution type phases were not returned, they are not plotted on **Fig. 3**. We observe the domination of C2/c structure, while diopside or other alkaline clinopyroxens are not found, and the found omphacite structures tend to a jadeite >> diopside solid solution. The SEM+EDS investigation (Bendő et al. 2014) of this tool proved the coexistence of omphacite, jadeite, ferrous jadeite and aegirine-augite.

N.1/81-1938 "JPM 1 81-1938" tool

The rock from which this tool was made is an almost pure omphacitite (Fig. 4.), with some jadeite and Ca-bearing jadeite components, which is listed in the database as "jadeite - diopside" solution (Table 3.). The splitting of peaks at $\sim 20^{\circ}$ (20) angles is related to presence of both omphacite and C2/c phase(s) with different cations and narrow peaks indicate the well-developed crystalline nature of pyroxenes (crystallite sizes $> 1 \mu m$). Some white spots were distinguished in its texture, but measurement trials resulted in patterns with very few peaks besides those of omphacite, thus phase identification was not possible, the patterns are not included here. The SEM+EDS investigation (Bendő et al. 2014) of this tool proved the coexistence of omphacite and jadeite, while the white spots were mainly epidote/zoisite + albite

N.11/169-1938 "JPM 1 169-1938" tool

Due to its homogeneous texture, with no visible spots or grains, one measurement was run on a larger surface. The pattern shows peaks of omphacite and jadeite-diopside as main phases, while sanidine $(d_{(130)}=3.752 \text{ Å} - 23.69 \circ 2\theta)$, $d_{(220)}=3.273$ Å – 27,215 °20) and ilmenite $(d_{(104)}=2.754 \text{ Å} - 32.48 \text{ }^{\circ}2\theta)$ were identified based on several smaller peaks (Fig. 5.). The peaks with net height < 5 counts were not included in the evaluation, even if they belong to accessory phases, the uncertainty of identification is too high. The SEM+EDS investigation (Bendő et al. 2014) of this tool proved the coexistence of jadeite, ferrous jadeite and aegirine-augite. In this case, the observed omphacite peaks measured by XRD may be a consequence of aegirine-augite and ferrous jadeite presence, thus only jadeite is marked as certainly identified.

39/1903 "MNM 39-1903" tool

An omphacite – jadeite green stone (**Fig. 6.**), jadeite with larger crystallite sizes, thus better developed grains, according to minimal peak broadening.

	Р	DF 78-2 C2	2037 (A 2/c)	PDF 80-1867 (B) <i>C2/c</i>				
	a=9. c=5	.501 Å, .238 Å,	b=8.654 β=107.	4 Å, 23°	a=9 c=5	.561 Å, 5.249 Å	b=8.73 , β=107	0 Å, 03°	
	Angle (°20)	d (Å)	R.I. (%)	h,k,l	Angle (°20)	d (Å)	R.I. (%)	h,k,l	
1	14.13	6.263	4.6	1,1,0	14.02	6.314	2.7	1,1,0	
•	20.31	4.369	7.8	-1,1,1	20.33	4.365	12.0	0,2,0	
2	20.51	4.327	16.4	0,2,0					
2	27.23	3.273	10.5	0,2,1	27.05	3.294	13.0	0,2,1	
3	28.48	3.131	13.7	2,2,0	28.25	3.157	18.6	2,2,0	
	30.40	2.938	100.0	-2,2,1	30.22	2.955	100.0	-2,2,1	
4	31.30	2.855	42.3	3,1,0	31.06	2.877	39.6	3,1,0	
	31.41	2.846	44.8	-3,1,1	31.25	2.860	33.6	-3,1,1	
	35.87	2.501	50.4	0,0,2	35.54	2.524	24.9	-1,3,1	
5					35.75	2.510	42.9	0,0,2	
5					35.75	2.510	42.9	-1,1,2	
	36.81	2.440	35.4	2,2,1	36.53	2.458	36.5	2,2,1	
	40.48	2.226	13.2	3,1,1	40.16	2.243	14.8	3,1,1	
6	41.49	2.175	13.9	1,1,2	41.27	2.186	13.6	1,1,2	
					41.47	2.176	10.0	0,2,2	
7	43.39	2.084	16.7	-3,3,1	43.08	2.098	17.9	-3,3,1	
0	45.65	1.986	17.5	0,4,1	45.14	2.007	10.3	-4,0,2	
0					45.27	2.001	21.3	0,4,1	
	56.96	1.615	13.0	-2,2,3	56.79	1.620	13.6	-2,2,3	
					57.76	1.595	15.0	-5,3,1	
9					57.76	1.595	15.0	1,5,1	
	58.19	1.584	15.2	1,5,1					
	58.19	1.584	15.2	-5,3,1					
10	62.44	1.486	13.0	-5,3,2	62.15	1.492	13.3	-1,3,3	
10	62.44	1.486	13.0	-1,3,3	62.15	1.492	13.3	-5,3,2	
	67.83	1.381	10.6	-3,5,2	67.53	1.386	19.9	5,3,1	
11	68.14	1.375	14.7	2,6,0	67.53	1.386	19.9	2,6,0	
	68.14	1.375	14.7	5,3,1					
	A:	Ca _{0.29} N	a _{0.6} Al _{0.4}	76Mg _{0.21}	Fe _{0.08} (A	l _{0.01} Si _{1.9}	₉₉ O ₆)		
F	B: (Ca _{0.4}	7Na _{0.41} F	e _{0.04} Mg	_{0.02})(Mg	_{0.44} Fe _{0.0}	₃ Ti _{0.01} A	l _{0.52})(Si	$_2O_6)$	

Table 3.: Peak positions for omphacite (A) and jadeite-diopside solid solution phase (B), from ICDD PDF card3. táblázat: Omfacit (A) és diopszid-jadeit (B) elegy fázis csúcspozíciói az ICDD PDF kártyák alapján



Fig. 6.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: 39/1903

6. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: 39/1903



Fig. 7.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: Olad-329

7. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: Olad-329



Fig. 8.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: 81/W2.5

8. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: 81/W2.5 **Table 4.:** Main theoretical peak positions of several clinoamphiboles, calculated according to their unit cell in Fehér (2009), calculated for Cu-K α radiation

4.	táblázat:	Néhány	klinoamfibol	elméleti	csúcspozíciói,	Fehér	(2009)	által	megadott	elemi	cellából	Cu-Ka
su	gárzásra sz	zámolva										

	1	2	3	4	5
h k l	(°20)	(°20)	(°20)	(°20)	(°20)
0 2 0	9.71	9.79	9.79	9.72	9.78
1 1 0	10.44	10.48	10.46	10.39	10.34
		17.34	17.28	17.23	17.13
1 3 0	17.29	17.42	17.39	17.27	17.31
1 -1 -1	18.08	18.14	18.00	18.01	18.03
2 0 0	18.53	18.59	18.54	18.42	18.27
0 4 0	19.49	19.66	19.65	19.50	19.63
0 2 1	19.84	19.95	19.90	19.82	19.76
2 2 0	20.96	21.06	21.00	20.86	20.76
2 0 -1		21.99	21.78	21.80	21.83
1 1 1	22.15	22.27	22.26	22.12	21.89
1 -3 -1	22.78	22.91	22.78	22.73	22.80
2 -2 -1	24.05	24.13	23.93	23.91	23.97
0 4 1	26.15	26.33	26.28	26.14	26.00
1 3 1	26.16	26.33	26.32	26.14	26.17
1 5 0	26.17	26.38	26.35	26.16	26.28
2 4 0	27.02	27.19	27.14	26.95	26.94
3 1 0	28.38	28.49	28.40	28.22	28.00
2 0 1		28.66	28.67	28.44	28.09
3 -1 -1	29.38	29.45	29.20	29.17	29.17
0 6 0	29.42	29.67	29.49	29.41	29.52
2 -4 -1	29.52	29.68	29.66	29.44	29.63
1 -5 -1	30.16	30.35	30.26	30.12	29.81
2 2 1	30.19	30.36	30.37	30.12	30.26
3 3 0	31.66	31.81	31.73	31.52	31.36
3 -3 -1	32.57	32.69	32.45	32.38	32.42
1 5 1	32.84	33.07	33.06	32.82	32.78
1 -1 -2	34.14	34.27	34.05	34.04	33.97
0 6 1	34.31	34.57	34.53	34.31	34.42
2 4 1	34.78	35.00	34.97	34.73	34.50
0 0 2	34.93	35.09	35.00	34.87	34.66
2 6 0	35.00	35.25		34.95	35.03
2 0 -2	35.23	35.34		35.07	35.10
1 7 0	35.76	36.06		35.76	35.96
	1. Actine	olite Ca ₂ (M	Ig,Fe ²⁺) ₅ Si	$_{8}O_{22}(OH)_{2}$	
2. 7	Fremolite	[]Ca ₂ Mg ₅ S	5i ₈ O ₂₂ (OH)	₂ ([]=vacar	ncy)
3. Pota	sscipargas	site (Na,K)	Ca ₂ (Mg,Fe	$e^{2+})_5 Si_8 O_{22}($	OH,F) ₂
4. Ferr	ohornbeln	de []Ca ₂ [F	e^{2+}_4 (Al,Fe	³⁺)]Si ₇ AlO ₂	22(OH)2
5.	Arfvedso	nite NaNa	$(Fe^{2+}Fe^{3+$)Si ₈ O ₂₂ (OF	I)2

10

20



40

50

60

Fig. 9.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: KGY-1

2Theta degrees

30

9. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourierpolinommal simított és háttér illesztett). Minta: KGY-1



Fig. 10.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: SM-KE64

10. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourier-polinommal simított és háttér illesztett). Minta: SM-KE64



Fig. 11.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: WE 18 3.3

11. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourier-polinommal simított és háttér illesztett). Minta: WE 18 3.3

Also the preferred orientation of (002) and (022) peaks indicates an oriented texture in the measured surface, with possibly platy crystallites of jadeite. The SEM+EDS investigation (Bendő et al. 2014) of this tool proved the coexistence of jadeite and ferrous jadeite.

Olad-329 "SM 329" tool

This tool is also omphacite – jadeite rock, with some minor unsolved peaks, which may belong to accessory minerals (**Fig. 7.**). In contrast with previous omphacite – jadeite samples, the presence of magnetite ($d_{(311)}=3.531$ Å – 35.43 °2 θ) is highly possible in this sample. The SEM+EDS investigation (Bendő et al. 2014) of this tool proved the coexistence of omphacite and jadeite.

81/W2.5 "WE 81 W2-5" tool

The main phase returned by Search/Match is jadeite, with omphacite and uncertain diopside - according to (110) peak position - (Fig. 8.), a \pm 0.05° (20) window on 20-scale had to be applied. The SEM+EDS investigation (Bendő et al. 2014) of this tool proved the dominance of jadeite with minor omphacite.

(2) Amphibole dominated schists, amphibolites

A short summary of amphibole structures is needed here also, since the amphiboles, structurally related to pyroxenes, are also difficult to be identified on a species level by XRD. The $[SiO_4]^{2-}$ anions form double chains, bridged by cations, and additional anions, mainly OH⁻ but also F⁻ (and more or less Cl) are incorporated to obtain structure neutrality. Cation ordering and distribution also creates the clino- and ortho series, but substitution and solid solution forming possibilities are greater than for pyroxenes. The classification is based on chemical compositional fields (Hawthorne et al. 2012), with multiple solid solution series and joints. Position of major peaks, calculated based on the unit cell given in Fehér (2009), for common clinoamphiboles are listed in Table 4.

"KGY 1" tool

This is a nephrite (=ferroactinolite – tremolite made fine grained or fibrous amphibolite, Zhou & Feng 2010) tool, with well-polished smooth surfaces, characteristic well visible fibrous texture. The first matching result was actinolite, with all the peak positions in >95% accordance of measured pattern (Fig. 9.). Although clinoamphibole species are more complicated to identify by Search/Match, than clinopyroxenes, in this case the match was acceptable. Even the oriented, long fibrous texture of the material did not produce complex preferred orientation effects. Pattern with good statistics was obtained with measurement time as low as 1 minute (Fig. 9.).

"SM KE64" tool

This tool is made up by amphibole, best match is actinolite (**Fig. 10.**) with minor preferred orientation and good agreement between theoretical and measured peak positions.

"WE 18 3-3" tool

In this tool, clinoamphibole presence is exclusive, as rock forming phase, but the Search/Match returned acceptable hits by applying the $\pm 0.1^{\circ}$ (20) window on 2 θ -scale. The best match was tremolite, with actinolite (**Fig. 11**.) retrieved in the second Search/Match iteration. Peak intensities are severely distorted due to surface morphology effects, the lack of plane surfaces resulted in shielding effect, either at low or high angles. We opted to record the higher angle region, most of the amphibole peaks being observed there.

"WE 67_W2-6" tool

The material of tool is amphibole dominant, mainly pargasite, but the presence of actinolite and tremolite is also likely. In contrast with the other amphibole rocks, this contains also sanidine and magnetite as rock forming phases, possibly with albite and anatase as accessory phases (**Fig. 12**.). Several major peaks and more small peaks remain unsolved, as these probably belong to the amphibole structure(s). Speculating on accessory phases would be inadequate, since peak positions may also be shifted by stress and strain, which is likely to affect minerals of metamorphic rocks.

"WE 81 W1-58" tool

This tool is dominantly made up by alkaline amphiboles, mainly by gedrite (**Fig. 13.**). Several more phases were observed as rock forming minerals, such as anorthite, pyrope garnet and ilmenite with magnesium substitution.

(3) Chlorite schists

The mineralogy and XRD identification of chlorite species is as complex as of amphiboles and pyroxenes. However, given the phyllosilicate layered type structure, cation distribution and substitution has some characteristic effect on (001) type peaks position, although differences can properly be observed on the (0l0) peaks and their relative intensities (**Table 5.**). Even if polytypes cannot be identified with these peaks, the chlorite structure can be recognised.

"SM 9-14-10_1" tool

The material of this tool is hardly classified as a specific rock type, since it is made up by clinochlore alone, with some trace presence of biotite (**Fig. 14.**).



Fig. 12.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: 67/W2.6

12. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourier-polinommal simított és háttér illesztett). Minta: 67/W2.6



Fig. 13.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: 81/W1.58

13. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourier-polinommal simított és háttér illesztett). Minta: 81/W1.58



Fig. 14.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: SM 9-14-10

14. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourier-polinommal simított és háttér illesztett). Minta: SM 9-14-10

Table 5.: Variation of (001) peaks position inclinochlore polytypes with chemical composition

(h,k,l)	1 Clinochlore-1MIIb PDF 46-1322	2 Clinochlore-1MIIb ferroan PDF 29-0701	3 Clinochlore 2a* PDF 73-2376	4 Clinochlore 1Mia PDF 89-2972					
(0,0,1)	14.200	14.100	14.368	14.165					
(0,0,2)	7.100	7.070	7.184	7.082					
(0,0,3)	4.750	4.710	4.789	4.721					
(0,0,4)	3.554	3.540	3.592	3.541					
(0,0,5)	2.840	2.828	2.874	2.833					
	1 Mg ₅ Al(Si,Al) ₄ O ₁₀ (OH) ₈								
2 (Mg,Fe) ₆ (Si,Al) ₄ O ₁₀ (OH) ₈									
	3 Mg ₆ Si ₄ O ₁₀ (OH) ₈								
4	$Mg_{2.5}Fe_{1.65}$	$_{5}Al_{1.5}Si_{2.2}A$	$l_{1.8}O_{10}(\overline{OH})$	4) ₈					

5. táblázat: Klinoklór politípek (001) típusú csúcsainak helye, kémiai összetételtől függően

Even if the (005) and some other peaks of the best matching structure are displaced, the measured phase corresponds to a magnesian clinochlore (Zheng & Bailey 1989). The strong preferred orientation of (0,0,1) type peaks is due to the oriented texture of the rock material, signalling a dinamometamorphic origin. Also, given the coincidence of chlorite lamellae parallelisms to the plate tool surface, indicates that the objects were fabricated from cleaved blocks of schist. The presence of platy, greenish matrix (Fig. 15a) and green transparent grains were observed by stereomicroscopy (Fig. 15b). The black metallic fragments could be iron oxide (e.g. magnetite) but biotite is also possible, while the orange grainy masses are Fe oxy-hydroxide alterations.

(4) Hornfels type contact metamorphic siliceous rock

"HOM 53-66-10" tool

The tool has a texture with no visible patches, homogeneously green coloured, one pattern was recorded. The Search/Match with $\pm 0.05^{\circ}$ window on 20-scale returned quartz and plagioclase varieties as highly matching phases.





Fig. 15.: Stereomicroscopic images of chloritic rock (sample SM 9-14-10), a – texture in general, b – chlorite grains

15. ábra: Sztereomikroszkópos felvétel a kloritpaláról (SM 9-14-10 jelű minta), a – a kőzet szövete, b – klorit szemcse

On a second iteration, run for peaks other than quartz $(SiO_2, trigonal)$ and calcian albite $[(Na_{0.9}, Ca_{0.1})AlSi_3O_8]$, ferroan clinochlore 1MIIb $[(Mg,Fe)_6(Si,Al)_4O_{10}(OH)_8]$, epidote $[Ca_2Al_{2.4}Fe_{0.6}(SiO_4)_3(OH)]$, magnesian ilmenite $(Mg_{0.208}Fe_{0.955}Ti_{0.833}O_3)$ and muscovite $[(KAl_2(AlSi_3O_{10})(OH)_2]$ was retrieved – chemical compositions according to ICDD PDF files. As it is marked in **Fig. 16.** some K-feldspar would be expected, but since no clearly distinguished peaks are observed, it was omitted from the results.



Fig. 16.: XRD patterns and matched phases PDF files (gray: original raw pattern, black: Fourier filtered and background modelled). Sample: HOM 53.66.10

16. ábra: XRD görbék és a kiértékelés során kapott fázisok, PDF számmal (szürke: eredeti mért görbe, fekete: Fourier-polinommal simított és háttér illesztett). Minta: HOM 53.66.10

Discussion

Peak positions were accurate enough in all measurements to conclude in successful Search/Match evaluation of patterns. As a routinely used evaluation process in our lab, it's useful to run the Search/Match in several iterations, creating "diff patterns" as secondary raw data, from the unidentified peaks. This way, in each step we have a set of peaks which defines a new intensity position data range, maximizing exact phase matching even at accessory minerals level. However, when only a few peaks are left, usually no acceptable hits are returned by the algorithm, thus a limit of the method is met. A matched phase may be accepted as valid if all theoretical peaks appear in the measured data, or we can deduce the cause or peak shifts (e.g. chemical substitution, solid solution series or strained structure). If most of the peaks are overlapping with those of major phases, and intensity distributions cannot be solved, then again we cannot accept that phase as valid without validating with other methods.

The most attention demanding task is the alignment of specimen surface, which can be done with the given goniometer equipment, without the exclusive need of precision centring sample stage. Even if needed, currently available precision sample stage compatible with this type of equipment don't met the requirements, being built without tilting and oblique sample rotating possibilities. Without these options specimen surfaces cannot be optimally adjusted to the goniometer.

Measurement times down to 1 minute already enabled the correct identification of rock forming minerals at least on group level. On well-polished, plane surfaces counting time can be reduced, since most of the diffracted intensity reaches the detector, and is integrated in registered pattern. However, if specimen surface is scarred and bent or uneven, the beam optics setting will prevent X-ray photons diffracted at larger than $\pm 0.06^{\circ}$ (20) diffraction angle difference to reach the detector. This reduces recorded intensity and low counting statistics results in useless pattern. On the basis of our observations, the choice on counting time will be easily determined according to macro geometric properties of tools.

Comparing the patterns and selected peaks of omphacite – jadeite samples, we can observe several characteristic differences. Peak positions, broadening and degree of overlapping may be used to differentiate between samples. According to measured peak maxima and overlapping, we observe ompachite with variable Ca-Al content and also jadeite with variable Fe content, even in transition to diopside (**Table 6.**). Changes in peak intensity are not suitable to use, due to complex preferred orientations, improbable to determine and model correctly.

Discrimination of amphibole species on crystal structure criteria is generally not accepted in mineralogist communities, since influence of cations on diffraction patterns is minimal. However, a sample dominated by one species with few cation substitutions measured on a diffractometer with good alignment and set-up, could result in identification of that species. Colour, on the other hand, is not plausible to be used as identification criteria of amphiboles, but still it will give information on the Fe²⁺ content of the species.

<u>xxx=dominant</u> <u>xx=secondary</u> <u>x=accessory</u>	omphacite	jadeite-diopside	jadeite	diopside	other
99.3.1863 "Gorzsa 11"	XXX	?			
N.1/81-1938 "JPM 1_81-1938"	XXX	XX			
N.11/169-1938 "JPM 1_169-1938"	XX	XX	?		sanidine, ilmenite
39/1903 "MNM 39-1903"	XXX		XXX		
Olad-329 "SM 329"	XXX		Х		magnetite, ilmenite
81/W2.5 "WE 81_W2-5"	х		XXX	х	

Table 6.: Omphacite-jadeite eclogite type pyroxenite tools composition**6. táblázat:** Omfacit-jadeit eklogit típusú piroxenit eszközök összetétele

Table 7.: Amphibole schist,	amphibolite tools composition
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7. táblázat: Amfibol palá	k, amfibolit eszközök összetétele
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	Actinolite	Tremolite	Pargasite	Gedrite	other
KGY 1	XXX				
SM KE64	XXX				
WE 18_3-3	XX	XXX			
WE 67_W2-6	XX		XX		magnetite, anatase, sanidine, albite
WE 81_W1-58				xxx	anorthite, pyrope, ilmenite

That is, in the specimen where we identified tremolite and gedrite, the tool was made up by white to grey fibrous material. Strongly oriented textures are expected to be characteristic, for high pressure samples mainly. Accordingly, if preferred orientation is not observed, it may suggest that the specimen is not linked to high pressure metamorphism. This could be of interest in special in the cases of nephrite type tools, given their disputed petrogenesis (Liu et at. 2011) between serpentinite related metasomatic, dolomite related contact metamorphic or regional metamorphic origin. The later one could be separated from metasomatism related materials based on dominant preferred orientation of crystallites. Summary of investigated specimens is given in Table 7.

Conclusions

The results of our experiments can be summarized in several important conclusions. First of all, we managed to use a unique combination of X-ray optics and detectors for non-destructive investigation of larger than usual samples, without conventional sample stages. The results of Search/Match evaluation proved to be in accordance with chemical investigations. Given the short recording times, and minimal number of measurements, this positive feedback is significant. Of course, increasing recording times and measurement number will create a more useful data package, with possible application in the petrogenetical characterization of greenstone materials. But the rapid screening is essential and cannot be skipped in order to find the best specimens for detailed investigation.

Selection of rock materials improbable to differentiate by other methods is straightforward. Chlorite and chloritic schists, amphibole bearing schists or amphibolites and pyroxenites, even with mineralogic subtypes, are easily recognized.

According to observations of actinolite amphibolites, even semiquantitative textural information could be extracted, e.g. for the degree of amphibole orientations and crystallite shapes. This information could be useful to determine amphibolite origin.

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3D VISUALISATION AND MULTIDISCIPLINARY ANALYTICAL TECHNIQUES ON CULTURAL HERITAGE OBJECTS FROM THE COLLECTION OF THE HUNGARIAN NATIONAL MUSEUM 3D MEGJELENÍTÉSI TECHNIKÁK ÉS LEHETSÉGES ARCHEOMETRIAI ALKALMAZÁSUK A MAGYAR NEMZETI MÚZEUM MŰTÁRGYAIN

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Abstract

3D visualisation techniques can serve archaeometric provenance studies by supporting non-destructive characterisation methods. In this paper, specific gravity measurements supported by 3D imaging techniques will be considered. The subject of analysis is so-called "greenstone" axes, all of them items of high prestige transported by long distance trade networks in prehistory. The 3D images obtained by (1) laser scanning (2) image matching methods help us fully document the artefacts and furthermore calculate the volume exactly. The advantages and drawbacks of these imaging techniques will be considered in respect of "greenstone" characterisation.

Kivonat

A 3D leképezési technikák, mint roncsolásmentes vizsgálati lehetőségek segíthetik az archeometriai célú származási-hely meghatározást. Tanulmányunkban a háromdimenziós képalkotási módszereket sűrűség meghatározásra használtuk. A vizsgálat tárgya néhány "zöldkő" balta volt, amelyek igen nagy távolságból kerültek Magyarország területére, különlegesen hosszú távú kereskedelmi láncolat eredményeképpen. A tárgyak igen nagy presztízs-értéket képviselnek, és ma is jelentős kulturális értékkel bírnak, ezért vizsgálatuk kizárólag roncsolásmentes módszerekkel végezhető. A kőzetek fontos fizikai jellemzője a sűrűség (fajsúly), ami a jadeit esetében meglehetősen magas érték, ezért meghatározása hasznos lehet a kőzet azonosításában. A sűrűség meghatározásához a tömeg és a térfogat pontos ismerete szükséges. A térfogat mérését először hagyományos módon, a vízkiszorítás mérésével (mérőhengerben) kíséreltük meg, de úgy tapasztaltuk, hogy ez az eljárás nem eléggé pontos a balták térfogatának mérésére. Ezért a térfogat meghatározására 3D képalkotási technikákat használtunk, úgymint (1) lézer szkennelést és (2) képillesztést, melyek segítségével az eszközök pontos modellje előállítható és térfogatuk kiszámítható. Bemutatjuk az alkalmazott képalkotási technikák előnyeit és hátrányait, elsősorban a "zöldkő" eszközök szempontjából.

Keywords: 3D Scanning, Image matching, specific density, jadeite

KULCSSZAVAK: 3D SCANNING, KÉPILLESZTÉS, SŰRŰSÉG, JADEIT

Introduction

Various objects of cultural heritage relevance from the collection of the Hungarian National Museum (HNM) were subjected to non-destructive analysis by nuclear and 3D imaging techniques. The main objective was to gather more information on the objects for presentation and characterisation studies (Schulze et al. 2010, Biró et al. 2011). After initial trials on objects ranging from stone artefacts, pottery, metal and composite objects, ranging from Palaeolithic to Early

Mediaeval, the analyses concentrated on polished stone tools made of "greenstones". These items represent in Hungary prestigous objects of longdistance trade. It is imperative to preserve the objects' full integrity and at the same time gather all available information for characterisation. Analytical techniques applied comprise magnetic susceptibility (MS), prompt-gamma activatrion analysis (PGAA, for bulk chemical composition), electron microprobe analysis (EMPA) and Goebel-mirror X-ray diffraction for mineral phases (Szakmány et al. 2013).

Methods

One of the important and characteristic physical qualities of high-pressure metamorphites (jadeite, omphacite and eclogite) is their high specific density (Errera 2014). We were trying to measure this feature by traditional measuring cylinder, but the reproducibility of the measurement indicated that our accuracy is not adequate. With the availability of 3D imaging techniques, we were trying to calculate volume with great precision that helped us to definfe specific density of the pieces. The biggest obstacles in the way of the current project concerning data recording and 3D digital modelling: First of all, it is important to mention that we were short of both resources and time. On the other hand, it was difficult to gain access to the stone materials and the limited number of data recording occasions also caused problems. Besides, to protect the artefacts, a strict rule was laid down in the analysis of polished stone-axes: only nondestructive methods were allowed. Lastly, complying with the requirements of measurement, it was necessary for the outgoing formats of the models to be compatible with the CAD systems. As it was an important goal to work out a cost-efficient method that can later be applied to bigger quantities of items, in the project we have tried the laser scanning method, which can be considered traditional and is more and more available, and the more progressive matching method among the 3D object digitization methods. They must be, as a rule, non-destructive methods and must maintain compatibility with the CAD systems (Lin et al. 2010, Sumner & Riddle 2008, Grosman et al. 2008). The main components of the computer used in the project were an AMD Athlon II X 4 561 Quad-Core 3.00 Ghz processor, 8 GB RAM, ATI Radeon HD 5600 video card, 500 GB hard disc

drive, and a 64-bit Windows 7 Ultimate operation system. For the 3D scanning and partly for generating the models, a NextEngine laser scanner was used. The scanner has a built-in digital camera, automated platen, focus setting options and an integrated 3D modelling software (ScanStudio HD). The set exactness of the scanner: 0.12 mm, speed of recording points 60.000 point/second. It is an easily available, compact, table-top scanner (e.g., Hermon et al. 2011, Slizewskí & Semal 2009). A Canon EOS 70D type camera was used for matching. Its important characteristics are as follows: useful image points/complete number of image points: 20.20/20.90 Mpixel. Picture aspect ratio was 3:2. In this case two have been chosen from the picture element matching softwares based on digital photos: 123D Catch (Lo Brutto & Meli 2012) and 3DSOM Pro (Ciobanu et al. 2013).

123D Catch was chosen due to its compatibility with AutoCAD (which makes it possible for it to become widely available) and the fact that it was developed for Autodesk softwarebase. The software is currently in beta testing phase. 3DSOM Pro was chosen because it is easy to operate and it is the widely used digitizing software of the virtual store of the Hungarian National Museum. In the case of the matching method, although the base softwares (123D Catch, 3DSOM Pro) had different abilities, the editing work did not differ much. For the edition, a 3DsMax software was used in the matching method, whereas in the laser scanning method Mudbox and 3DsMax softwares were used. The detailed working process of the laser scanning method and that of the picture element matching method can be seen in Fig. 1.



Fig. 1.:

Work-flow chart of 3D laser scanning (1.) and 3D image matching (2.), respectively

1. ábra:

3D lézer szkennelés (1.) és a 3D képillesztés (2.) munkafolyamatának összehasonlítása



Fig. 2.: A tetrahedron, shown in black, formed by the a surface triangle (a-b-c, black crosshatch) and a reference point, e.g. the origin. Once its volume is calculated, the sum of these sub-volumes is a good approximation of the entire object volume

2. ábra: Egy tetraéder (feketével kiemelve), amelyet egy felületen lévő háromszög (a-b-c, feketével sraffozva) és egy referencia pont (pl.az origó) határoz meg. Az egyedi tetraéderek térfogatainak összegéből a tárgy teljes térfogata meghatározható.

The exact measurement of volume, on the basis of 3D imaging techniques, can be realised according to the following principles. The laser scanners result in a surface mesh of the scanned object. In this representation, the real surface of the object is approximated by a set of triangles (so called vertex). These are shown with cross-hatches in Fig. 2. If we select a reference point (usually the origin), and connect it with the corners of a triangle, we define a tetrahedron (Fig. 2.). For any complicated object, once approximated with such a triangular mesh, the volume is the sum of the subvolumes of all tetrahedra bounded by the origin and a mesh triangle (shown in black; Obádovics 1994, Schuster 2008). This calculation can be generalized to cases where the object is not convex or does not contain the origin .

The market-leading laser scanner devices can capture fine details down to about 100 micron sizes. The NextEngine 3D Laser Scanner used in the present study stores the data in STL or OBJ formats that can be readily loaded to a CAD software (**Fig. 3**.). The CAD programs offer functionality to measure the linear extents of the object, and have the surface and volume calculation algorithms built in, so the user can obtain the physical features of a laser-scanned object in a quick and convenient way.



Fig 3. A coarse 3D digital representation of an object (Csót, HNM 300/876.431) where the triangular mesh was made visible

3. ábra: A tárgy (Csót, HNM 300/876.431) egy kisfelbontású 3D digitális modellje, amelyen a felületi hálót láthatóvá tettük.

Similarly, the center of gravity can also be defined, supposing the density of the material is constant over the entire volume.

Once the mass of the object is measured, the digital volume is calculated, the average density of the object under study can be obtained with a few percent precision. In contrast to the Archimedes' principle (buoyancy method), which states that a body immersed in a fluid apparently loses weight by an amount equal to the weight of the fluid it displaces, the proposed method allows the unbiased density determination not only for solids, but also porous objects or wooden objects. In the latter cases they can take up water and bias the result. The reliable density information offers a new possibility to classify the stone tools.

Results

There was a clearcut difference between the different object-digitizing methods (laser scanning or matching). The number of triangles in a mesh 3D model made by laser scanning is 81.000, whereas that for the matching is only 8800. The same difference was experienced in working hours too: here, the time required to record the data, to create and to visualise a 3D model (from the time the artefact was delivered) was 11 hours in the case of laser scanning, whereas in the case of matching (also from the time the artefact was delivered) it took 5 working hours (**Fig. 4a, Fig. 4b**).

In the partial summary below, which is focussed on the scientific and practical experiences concerning the analysed artefacts, it was not our aim to give a general theoretical description of the proven methods of digitization (laser scanning, picture element matching) in terms of their methodologies. Advantages and drawbacks of each methods are summarised in **Table 1**.

Table 1.: Advantages / drawbacks of 3D laser scanning and 3D image matching, respectively

1. táblázat: A 3D lézer szkennelés és a 3D képillesztési technikák előnyei és hátrányai

Laser scanning method (NextEngine laser scanner).		Picture element matching method (123D Catch, 3DSOM Pro).			
ADVANTAGES	DRAWBACKS	ADVANTAGES	DRAWBACKS		
- The artefacts were suitable for the range of sensors and measure- ments and the automating possibilities of the scanner.	- Scanning reqired smooth and scattered lighting.	- The number of point data was enough, the 3D model could be handled easily with the software.	- There were 3D modelling errors due to the lighting phenomena connected to the exposure of the artefacts.		
- Some digital surface flaws occured due to the light phenomena caused by minerals.	- It was difficult to record the real colour of dark surfaces.	- Digital photos projected the texture of the surface in great details.	- It is difficult for the matching process to filter the shades.		
- The objects could be seen well: there are no holes, overlapping parts or bigger grooves.	- On the superficies of the artefacts there were only few identifying points The picture element matching process gave more real image of artefact.		- Good quality recording required elements with strong contours and contrasts.		
- The surface determiners of the polished stone-axes were easy to calculate with algorythms.	- When scanning, mistakes could be made with the correct recording sequence of the surfaces, the angle of exposure and the handling of covered parts.	- It was easy to make up for any lack of recorded data with photos of parts.	- In photos, only the focal point and its surrounding area were really clear.		
- The projected cloud of points was differentiated and rich in details.	- Many points were recorded, which slowed down the process. The software of the scanner tended to treat the surface parts separately.	- To bring the 3D model into a showable condition required less and more simple editing.	- Sometimes, there were not enough matching points for orienting in close-up and high resolution photos.		
- The surfaces of the objects had smooth texture.			- The background behind the objects considerably hindered the process of digitization.		
			- The parts standing out of the surface did not differ from the whole of the digital 3D model, blurring with it.		
			- Good texture covered the surface deficiencies of the 3D model.		
			- The process was made more difficult by the fact that basically, the photos had to be taken in a given round-sequence.		
			- Adding absolute sizes meant an extra process.		

Table 2.: Results of density measurements obtained on the models of 'greenstone' axes by 3D Scanning and 3D image matching

2. táblázat: Sűrűség mérési eredmények a "zöldkő" baltákon 3D lézer szkenneléssel és 3D képillesztési technikával felvett modelleken

Rock type	serpentinite?	jadeitite	jadeitite	iron-jadeitite	serpentinite	greenschist / metabasite	greenschist / metabasite??
3D DP photo	ΓI	P2	P3	P4	P5		
3D laser scan		G4	63	G2		GS	G1
Density (g/cm ³)	2.76	2.95 3.01	3.31 3.25	3.17 2.95	2.28	2.89	2.95
Volume (mm ³)	8115	51073 49898	9216 <i>9356</i>	8497 9122	7647	6744	77560
(g)	22.43	150.61	30.49	26.99	17.45	19.51	228.94
Dimensions (mm)	31*33*12.5	97*50*19.5	57*33*12	40*32*10	42*25.5*9.7	40*26*8	102*61*20
Locality	Belácz, Tolna megye	Veszprém megye	Bakonypéterd	Almásneszmély	MURSELLA beteen Árpád and Mórichida	Csót	Nyergesújfalu
Inv.Nr	MNM Ltsz: 51/1867.II.3	VMM Ltsz: MIH.1276	MNM Ltsz: 300/876.264	MNM Ltsz: 300.870.247	ELTE not yet inv.	MNM Ltsz: 300/876.431	MNM Ltsz: 300/876.83



Fig 4. 3D digital representation of a greenstone axe4. ábra: Zöldkő (jadeit) balta 3D modellje

Fig. 4a: by 3D laser scanning;4a: lézer szkenneléssel

Fig. 4b: 3D image matching method 4b: képillesztéssel

The actual results of our density-measurements on the 'greenstone' axes are summarised in **Table 2**.

In case of two items (Bakonypéterd and Almásneszmély, respectively), we had the possibility to compare results of 3D scanning and 3D image matching on the identical object. It was realised that for the images made by 3D matching we have to apply an internal scale (typically, the longest dimension of the tool) for getting the correct starting point for the calculation of volume. Moreover, the number of points recorded is much higher for 3D scanning, thus the volumetry is more direct and more reliable.

Densitometry by the help of 3D imaging has become one of the powerful arguments of identifying greenstone axes.

The current status of jadeitite/omphacite axes are presented, using the base map of Pétrequin et al. 2011 (Fig.1.), with the location of recently identified items, on **Fig. 5.** More on the problem is

presented in details by Bendő et al., this volume.

Conclusion

On the 3D object digitizing process based on the NextEngine laser scanner. the following conclusions can be drawn: The data of the points of the polished surfaces could easily be projected. At the same time, it was easy to spot any mistake made in recording the data. The 3D objects could be measured without texturing. The drawback of the method of digitizing artefacts with laser scanners is that it is slow and requires considerable resources and work. On the other hand, laser scanning is a proven, established process. In our case, it can be generally stated that their sizes, material characteristics and shapes made it easy to digitize the artefacts.

The possibilities of the used picture element matching process could be defined from the aspect of the current project in comparison with the laser scanning method.



Fig 5. Map of jadeite axe distribution after Petrequin et al. 2011 Fig.1. with new Hungarian sites known till March 2014 added in the little red rectangle. (see in details Bendő et al., this volume)

5. ábra: A jadeit balták elterjedése Petrequin et al. 2011 Fig.1. alapján, kiegészítve a 2014. márciusáig megismert új magyarországi darabokkal (ld. Bendő et al., ebben a kötetben)

Using this method, the sizes and shapes of the artefacts were advantageous in their digitizing, however, the blurred contours, the homogeneity and the lack of contrasts of their surfaces were disadvantages. In this case, the surface flaws of the 3D models were more difficult to spot, but they were easier to correct. On the other hand, the 3D models were suitable for measurement only with software correction. Generally speaking, this process is cheap, fast, it reqires fewer tools and less workforce, the tools are used daily in most scientific institutes and museums. However, picture element matching is a less proven method, however, the prospects of its applications are improving.

Taking everything into consideration, the process based on picture element matching is suitable for the non-destructive analysis and the visualisation of these artefacts.

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PRELIMINARY RESULTS OF AN ARCHAEOMETRICAL STUDY OF THE ECSE-HALOM (KURGAN) IN HORTOBÁGY, HUNGARY A HORTOBÁGYI ECSE-HALOM ARCHEOMETRIAI VIZSGÁLATÁNAK ELŐZETES EREDMÉNYEI

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Abstract

Ecse-halom is a kurgan in the Hortobágy region in Hungary that was built during the Late Copper Age/Early Bronze Age by eastern nomadic communities. It is located on the border between two modern settlements. A road of medieval origin runs along the body of the mound and separates it into two parts. Its southern half was ploughed and used as a rice field; later a military observation tower was built on the top of it. Despite of all the surface of the mound is in a fairly good condition and provides a home for regionally significant, species-rich loess steppe vegetation. During the winter of 2011 the research team of Professor Pál Sümegi conducted an undisturbed core in Ecse-halom and complex archaeometrical analyses were carried out on the profile of the mound. The mound comprises two construction layers as indicated by the decrease of magnetic susceptibility. The examination of organic compounds and carbonate content at various levels showed different values. The distribution of grain size within the section is characterized by mid-sized silt fraction.

Kivonat

Az Ecse-halom egy a késő rézkor és kora bronzkor során, keleti nagyállattartó népek által felépített kurgán a Hortobágy területén. Maga a halom két modern település határvonalán található, amely mentén egy, már a középkor során kialakult földút mélyül a kurgán központi részébe. A halom déli részét a 20. század közepén rizsföldként hasznosították, később egy szovjet katonai megfigyelő pontot is építettek a halomra. Ennek ellenére a halom felszíne helyenként viszonylag jól megőrződött, ezeken a részeken fajgazdag löszsztyepp vegetáció maradt fenn. 2011 telén Sümegi Pál professzor vezetésével egy zavartalan magfúrást mélyítettünk az őskori népek által kialakított halomba és a béléscsöves fúrás által feltárt üledékszelvényen komplex archeometriai vizsgálatot végeztünk el. Az egyes rétegződések eltérő mágneses szuszceptibilitás-értékei is megerősítik, hogy a halmot két fázisban hordták fel, míg a karbonát- és szervesanyag-tartalom arról tanúskodik, hogy igen változatos talajtani környezetből származott a halom üledékanyagban.

KEYWORDS: GREAT HUNGARIAN PLAIN, HORTOBÁGY, KURGAN, COPPER/BRONZE AGE, ARCHAEOMETRY

KULCSSZAVAK: ALFÖLD, HORTOBÁGY, HALOM, RÉZKOR/BRONZKOR, ARCHEOMETRIA

Introduction

Due to the constant agricultural cultivations in the eastern areas of the Great Hungarian Plain, most of the mounds are endangered. Many have permanently disappeared; the remaining is struggling from being destroyed. Therefore, research of mounds in Hungary is highly important and indispensable. We need to act now to perform scientific data collections, surveys and cadasters to protect the cultural heritage of these earth monuments. Nowadays, mounds are vanishing monuments of the landscape of the Great Hungarian Plain. Surveying and protection is our common goal, which needs precise, reliable scientific research work. Our research plan is the thorough surveying of mounds in the Tiszántúl and Hortobágy region, and to establish a strategy for their protection.

The kurgans are mound-graves in the steppe zone of Eurasia and nomad people built these mounds during the prehistoric ages (Late Copper/Early Bronze Age). Hungary is the westernmost point of
the Yamnaya culture (Anthony 2007; Ecsedy 1979). Mounds can be found at the banks of no longer existing rivers and at some points of higher altitude areas. According to their origin, mounds can be classified as burial sites and sacred points of ancient nomad people (Horváth 2011; Dani & Horváth 2012).

These mound-graves are highly important from archaeological and other points of view in the Carpathian Basin. In the 18th century the number of kurgans was estimated to be around 25,000 in the Great Hungarian Plain, but lot of them vanished in the past two centuries. Only a few hundred is still in good condition (Tóth 1988).

Mounds are salient cultural elements of the landscape of the Hungarian Plain. Through detailed and complex study they provide information not only on the history of millennia, the life, archaeological heritage and customs of the people buried inside them, but also on the occupied environment, the ancient flora and fauna, and the geological formations on the surface (Barczi et al. 2011; Pető & Barczi 2011; Szilágyi et al. 2013).

In order to protect all the remaining mounds, we need to perform a thorough archaeological, environmental historical, topographical, and morphological survey in these areas. This work needs a lot of field studies, and collections of data from archives and old maps.

In 2012 it became possible to launch the scientific analysis of the Ecse-halom, located in the area of the Hortobágy National Park (Sümegi 2012). The aim of this article is to present the preliminary results of the research from a geomorphological, landscape historical, botanical, sedimentological and micro-morphological point of view (Bede et al. 2014).

Methods

The handmade maps from the 18th and 19th century and later the printed maps broadly supported geomorphological and landscape historical researches, which were able to follow up the landscape changes of the last two and a half centuries. Since the Ecse-halom has stood on the borderline of settlements already in earlier ages, there are available documents available of borderpassing charters from the Middle Ages and the Early Modern Period, which retained very valuable records not only about the kurgan as a border-point, but the surrounding landscape and its usage as well.

We used a Topcon (Hiper SR GNSS, FC336 type) high accuracy satellite positioning equipment (RTK) for the preparation of the Ecse-halom's contour-map and for the field-modelling. With the help of this tool we were able to assess the entire superficies of the kurgan and its immediate surroundings (buffer zone) in great details as well. We evaluated the data and edited the geomorphological two and three dimensional field models with the help of ArcGIS 10 and AutoCAD Map 3D 2010 programs. With the help of these softwares we have also reconstructed the status of the kurgan before the disruption.

During the botanical investigation of the Ecsehalom we prepared a complete list of vascular plants, with attributes of frequency and coating. We made a 3D vegetation map, too. Each vegetational patch was documented by seven phytocoenological samples covering 2×2 m areas, and a 3D vegetation map was made.

For sedimentological researches machine drilling was carried out at the highest point of the mound, and samples were lifted from a double drill pipe 10 cm in diameter. The complete length of the core was 10 m; 99.7% of the core was removed. Samples were taken from the 1000 cm long core at 8 cm intervals on average; altogether 116 samples were included in the sedimentological, magnetic susceptibility, organic material, and carbonate analyses, furthermore for micromorphological survev (Dean 1974; Sümegi 2012). Micromorphological analysis was carried out according to the work of Murphy (1985).

Particle size measurements were conducted using an OMEC Easysizer 2.0 laser granulometer. Sediment samples were treated with hydrogen peroxide to disperse clay aggregates prior to analysis. Magnetic susceptibility measurements were taken with a Bartington MS2 meter. Graphical representation of the results was produced using Psimpoll software. For the element analyses the five-step extraction technique of Dániel (2004a; 2004b) was used. This involved extraction in distilled water as a first step to measure water soluble ions and ions bound weakly onto the mineral surface. In this paper the changes of the water soluble Fe content is shown because a contex was found between MS and water soluble Fe data.

Geomorphological results

The Ecse-halom kurgan is located in the Great Hungarian Plain, in Jász-Nagykun-Szolnok County, in the area of the historical Nagykunság (Greater Cumania), in the Hortobágy region, within the Hortobágy National Park, 12 km north-northeast of Karcag. It is a border point between the administrative areas of the settlements of Karcag and Kunmadaras (Fig. 1–2).



Fig. 1.: Ecse-halom on the Great Hungarian Plain

1. ábra: Az Ecse-halom az Alföldön



Fig. 2.: The Ecse-halom from the south, spring of 2014 (photo by Á. Bede)

2. ábra: Az Ecse-halom déli irányból, 2014 tavaszán (Bede Á. felvétele)

The Ecse-halom itself is located on an elevated point in the landscape, on a remnant surface covered by Pleistocene infusion loess. It shows connections with the loess landscape of the Nagykunság area, basically it is its northeastern protrusion that wedges into the Holocene alluvium of the Hortobágy. The mound rises on the eastern end of a slightly elevated, elongated loess ridge that is clearly separable from its surroundings on the basis of its vegetation and geomorphology. This remnant surface is surrounded (primarily in the north and east) by floodplain marshes and creek beds that are an organic part of, or geohydrologically connected to, the rather complex Kunkápolnás marsh system (Fig. 3).



Fig. 3.:

The Ecse-halom and its vicinity (based on M.5 and M.12). Dark blue: deep floodplain; light blue: shallow flood plain; dark green: high floodplain; light green: unflooded area; yellow: loess ridge; brown: Ecse-halom

3. ábra:

Az Ecse-halom és környezete (M.5 és M.12 nyomán). Sötétkék: mélyártér; világoskék: alacsonyártér; sötétzöld: magasártér; világoszöld: ármentes terület; sárga: löszhát; barna: Ecse-halom





The roughly round mound, slightly elongated along its west-east axis, has been deformed considerably during the past centuries (**Fig. 4**).

The most apparent is the deep road cutting the centre of the mound in an east-west direction, which has served as a road of local significance since medieval times and due to continuous abrasion and erosion cuts now many meters deep into the body of the mound (Bukovszki & Tóth 2008). A border line of medieval origin, which is still visible immediately north of the road in the form of a border ditch, was established along it.

The mound is topped by a triangulation point.

The effects of the 20th century are visible on the southern periphery of the mound, in the form of parallel dams and embankments of the rice field established here. A larger amount of soil was removed from the southern side of the mound, but the same can be said about the highest part of the northern side as well. Based on the environmental reconstruction it could be established that the body of the mound had two layers: the first, earlier was 1.3 m thick, the second was 2.9 m (**Fig. 5**). The traces of a ditch that was created when the earth was piled up on the mound are barely perceivable around the mound (Sümegi 2012). This filled up, geomorphologically hardly detectable ditch is more visible on the northwestern and northern edges.

The central coordinates of the Ecse-halom are N47°25'31.11", E20°57'47.71"; its absolute height is 93.5 m asl, its relative height is 5.5 m, its length is 75.5 m and width is 67.5 m.

Its geomorphological and geological characteristics are similar to the Csípő-halom mound, also located in the Hortobágy region, near the village of Egyek (Barczi & Joó 2003; Barczi et al. 2006).



Fig. 5.: 3D detail section of the reconstructed Ecsehalom. 1: the second construction layer of the mound with the recent soils; 2: the first construction layer of the mound; 3: the surface of the palaeosoil under the mound

5. ábra: A rekonstruált Ecse-halom háromdimenziós metszete. 1: a halom második felhordási rétege a mai talajfelszínnel; 2: a halom első felhordási rétege; 3: a halom alatti paleotalaj felszíne

Its best parallel, however, is 3 km to the west, also located in the Kunmadaras plain: the slightly lower and smaller Nagy-Füves-halom mound.

Landscape historical results

The mound had been built by nomadic people of the pit grave kurgans of eastern origins (Yamnaya culture), and can be dated to the Late Copper Age/Early Bronze Age (3300-2500 B.C.) (Dani & Horváth).

The first part of the name of the Ecse-halom (eče \sim äčä) is a Cumanian word meaning "sister (mother, woman)" (Baski 2007). Local tradition considers it as a personal name and connects it to a Cumanian warrior called Ecse, who was the owner of the mound (Pesty 1978; Győrffy 1921). The Ecsehalom is such a salient element of the landscape and a series of place names had been formed from its name in the vicinity during the past centuries: Ecse-rét (Ecse meadow) (M.3, M.6, M.7), Ecseróna (Ecse port) (M.3), Ecse-zug (Ecse corner) (M.5, M.7, M.10), Ecse-fenék (Ecse depth) (M.9), Ecse-kút (Ecse fount) (M.9), Ecse-háti-tanya (Farm of Ecse beck) (M.11), Ecse-halmi-major (Grange of Ecse mound) (M.12) and Ecse-gát (Ecse barrage) (M.12).

The topographical significance of the mound is also shown by the fact that since the 18th century until recent times, manuscript and later printed maps all indicated its position and name: "Ecze halom" (M.1), "Etse halma" (M.2), "Etse Halom" (M.3), "Etse halom" (M.4), "Ecse halom." (M.5), "Ecse halom" (M.6, M.7), "Ecse-hlm." (M.8, M.9, M.10, M.11, M.13, M.14, M.15), "Ecsehalom" (M.12).

The Ottoman Period treasure tale connected to the mound indicates its disturbance in the past.



Fig. 6.: The Ecse-halom and other mounds in the vicinity on the Second Military Ordnance Map of the Habsburg Empire (M.6)

6. ábra. Az Ecse-halom és a környező halmok a második katonai felmérésen (M.6)

A miserly man from Kápolnás, when running from the Turks, dug a large pit into the side of the mound with his two servants. They lowered a punt full of money here, and then the envious master pushed in the two servants as well, buried them and smoothed the ground. He was, however, cursed and sunk knee deep into the ground, and could never again lift his legs from the earth of the mound. He went mad and stayed like that until he died (Kimnach 1903).

Based on the environment and old maps we may assume that until the 20th century the Ecse-halom was primarily used for animal husbandry (pasture, mowing), and no arable farming was carried out in the immediate vicinity (Fig. 6).



The dirt road (practically a road cut deep in the soil) running across the Ecse-halom was shaped by hundreds of years of use and the consequent erosion. It was used already in the Middle Ages (16th century), since a road of local interest ran this way, connecting Kunkápolnás and Nádudvar (Elek 2008).

Later on (after the destructions of the Late Ottoman Period) it lost its significance, although locals still use it until this day. The continuation of the road to the east is the Ecse-gát (Ecse barrage), which enables the crossing of the deeper parts of the Kunkápolnás marsh system.

The Ecse-halom is mentioned first in a charter describing village borders from 1521 (in the form "Echehalma") (Benedek & Zádorné 1998; Gyárfás 1883). In the Early Modern Era it was the border point between the villages of Asszonyszállás and Kápolnás. Today it lies on the administrative border between Karcag and Kunmadaras; the borderline breaks in an angle on the tip of the mound.

Manuscript maps from the 18th 19th centuries (M.1, M.2, M.5, M.6) and later printed maps (M.7) consistently represent the whole area of the mound as pasture. In the beginning of the 20th century, however, its southern half was ploughed due to the increased demand for arable land, such as in 1943 (M.8). Socialist large-scale agriculture and the consequent large-scale landscape transformations did not spare the Ecse-halom either: in the 1950s rice parcels were established on its southern side (M.10), that traces are still visible. In the 1960s the area served again as pasture (M.11), and is used like that till today. In the wider vicinity of the mound farmsteads, dirt roads, ditches, embankments, grass fields and lower lying swamps can be found.

Fig. 7.:

Vegetation map of the Ecse-halom: 1: loess steppe; 2: uncharacteristic dry grassland with loess steppe elements; 3: population of Agropyron cristatum; 4: very dry ruderalia; 5: dirt road with trampled weeds; 6: triangulation point; 7: concrete elements of the foundation of the military observance tower on the surface; C1-7: coenological samples

7. ábra:

Az Ecse-halom vegetációtérképe. 1: löszpusztagyep; 2: jellegtelen száraz gyep, löszpusztagyep-elemekkel; 3: taréjosbúzafű (Agropyron cristatum)-állományok; 4: igen száraz, ruderális gyep; 5: taposott gyomnövényzet (földút); 6: háromszögelési pont; 7: katonai megfigyelő torony alapozásának felszíni betonelemei; C1-7: cönológiai felvételek



Fig. 8.: MS, water-solution iron content and sedimentological data from the core sequence of the Ecse-halom **8. ábra:** Az Ecse-halomba mélyített fúrás rétegsorának MS-értékei, vízoldható vastartalma és üledékföldtani adatai

The mound was used as an observation point of the Soviet military shooting-range of Kunmadaras, from where the bombings and shooting practices were controlled. As a consequence, a small sentry-box was set up on the northern edge of the mound (M.12), and when it was demolished in the second half of the 1980s a multi-storied, steel-framed observation tower was built in the southern side (Tóth 1988). The tower, which significantly diminished the landscape value of the mound, was pulled down by the national park after the pullout of the Soviet military troops, but its concrete base elements sunk into the mound are still there (**Fig. 7**); their removal and dispatch is an urgent task.

Botanical results

The vegetation of the Ecse-halom is in fairly good condition, partly due to its maintenance and the regular but not excessive grazing and mowing.

The kurgan rises above its marshy, alkaline environment, thus most of its surface is covered by a loess steppe association (*Salvio nemorosae-Festucetum rupicolae*) and its derivatives (**Fig. 7**). The mounds are characteristic places of survival of these, from a conservationist point of view, significant habitats and the association itself (Illyés & Bölöni 2007; Horváth et al. 2011; Joó 2003). In the northern half of the Ecse-halom, loess steppe in a fairly good condition can be found. Crested wheatgrass (Agropyron cristatum), characteristic for the dry vegetation of loess bluffs, forms only a few smaller patches beside the top and on the northern side. In the southern half of the mound, vegetation is secondary, uncharacteristic dry grassland, a fallow unploughed for decades. But even this area contains already a few loess steppe species. The steep, south-looking side immediately to the south of the top is covered by dry ruderal species, and is separated by a fairly sharp border from other vegetation zones. On the road cutting through the mound in an east-west direction the tracks are flanked by trampled weed associations. Arboreal vegetation is only very sparsely present in the area.

We have detected the occurrence of about 90 vascular plant species until now from the kurgan. It could be established that the flora of the mound is rich in species in a countrywide comparison. Among the attested species we may mention *Aegilops cylindrica*, *Agropyron cristatum*, *Androsace elongata*, *Bassia sedoides*, *Carthamus lanatus*, *Linaria biebersteinii*, *Muscari comosum*, *Ranunculus pedatus*, *Salvia nemorosa* and

Verbascum phoeniceum. Although the Ecse-halom is not among the most valuable mounds in terms of plant species composition, regionally it certainly represents significant natural value, especially thanks to the presence of species characteristic of loess steppe.

Sedimentological results

The vegetation and soil characteristics of the mound and its immediate environment (extralocal level) are different on local and regional levels. At a regional level, hydromorphic soils are typical, while at a local level hydromorphic and alkaline soils dominate. The same can be observed in terms of vegetation as well: marshes, alkaline marshes and wet alkaline meadows dominate at a regional and local level in the wider area. Extralocally, however, drier type of alkaline meadows and the vegetal elements of loess grassland dominate (Sümegi et al. 2013).

The MS values were at a maximum in the lower part of the core, where in some cases they exceeded the value of $100*10-6 \text{ m}^3\text{kg}^{-1}$; in contrast, they diminished drastically in the upper layer, between 150 cm and the surface. The MS minimum observed in the material of the mound (420–0 cm) indicates that the mound had been built in two phases. The decrease of magnetic susceptibility values also indicates solution and migration processes; these were observed both in the paleosol and in the 'B' level of the higher, recent soil layer (**Fig. 8–9**).

The maximum of organic material was recorded in the sole of the core. The organic content of one sample exceeded 10%, in others it was around 6-7%. The carbonate content at the same levels, however, was lower. In the lowermost layer, above the sole, we observed diminished organic material content and increased carbonate content.



Fig. 9.: The correlation between water-solution iron content and MS data from the core sequence of the Ecse-halom

9. ábra: A vízoldható vastartalom és az MS-értékek összefüggése az Ecse-halom rétegsorában

This trend, however, reversed in the layer at 800–780 cm depth. From 800 cm both carbonate and

organic content values fluctuate. In the zone between 150 and 80 cm we observed a significant decrease in organic material and an increase in carbonate content. The same anomaly was detected on a lower horizon of the core (570–530 cm). In these levels carbonate values reach their maximum (**Fig. 8**).

The distribution of grain size in the whole core shows uniformity. Mid-grained silt fraction, 0.016-0.031 mm in size, appears in the largest quantities. We detected outstanding values in only a few points of the core (**Fig. 8**).

Based on the macroscopic description, sedimentary facies features, geoarchaeological characteristics and magnetic susceptibility data of the core, the following layers, genetic levels and consequently natural and anthropogenic processes could be identified (**Fig. 8**).

The sole of the core at 1006-800 cm depth is composed of unsorted, clayey silt and floodplain sediment. Based on its carbonate content the sediment is the result of the work of the Sajó and Hernád rivers that played a primary role in the formation of the terrain of the area in the Pleistocene. As the results of Franvó (1966). Rónai (1985), Sümegi (1989; 1997), Nyilas & Sümegi (1991), Szöőr et al. (1991; 1992) and their investigations was reconstructed - and after then many others have pointed out - that most part of the near-surface sediments of the Hortobágy is not the poor carbonate sediment Tisza/Old Tisza river, but the carbonate-rich sedimentary Old Sajó-Hernád rivers accumulated. As a result, it was developed loess similar, but basically alluvial sediment (Földvári 1958; Sümegi 1989; Szöőr et al. 1991; Szöőr et al. 1992), which constitutes one of the basic conditions of alkalization and alkalization of bedrock (Scherf 1935).

At 800–780 cm depth a thin layer immediately above the sole can be found, also deriving from the alluvium of the Sajó-Hernád river system, consisting of well-sorted riverine sand. This is indicated by the data on grain composition and carbonate content, and the shell fragments of certain Mollusca species.

The layer between 780 and 570 cm is composed of infused loess formed in the second half of the Pleistocene, consisting of fine and coarse silt fraction, which corresponds to loess sediments found in other areas of Hortobágy. Its age can be placed between 25,000 and 12,000 years, and it forms part of the Holocene base rock.

The 'B' level of the paleosol (570–530 cm) is characterized by high carbonate content and low MS values. Shells of snail species indicate steppe and forest steppe environment, but based on the macroscopically observable traits the paleosol was categorized as meadow chernozem. The 'A' level of the paleosol can be found in the section between 530-420 cm.

Magnetic susceptibility values are almost completely identical to those of the 'A' level of the Early Holocene soil, rich in organic material. At the same time we could detect a change in the MS value at 290 cm depth, indicating the movement of elements and solution, consequently a long-term open surface. Based on this the Late Copper Age-Early Bronze Age Yamnaya community probably raised the mound in two phases (Fig. 5, 8). The first layer was found between the depth of 420 and 290 cm. The two mound building horizons are separated by a short period of time not longer than one or two generations (30-50 years), since no carbonate content could be observed in the lower layer, indicating that carbonate movement had not started and become intensive on the surface of the lower kurgan surface. It cannot be excluded that the two kurgan building phases are connected to two major burials, but this could be confirmed only through the complete excavation of the mound.

In the soil of the kurgan we managed to demonstrate the presence of carbonate, ferrous and alkaline patches, which indicate that the kurgan was formed from three types of hydromorphic, alkaline and prairie soils that could have formed a hydroseries in the region. Nevertheless, the majority of the kurgan is made up of prairie soils, thus in the Early Holocene it must have dominated in the vicinity of the kurgan. Consequently, although alkaline and hydromorphic soils must have occurred in the environment of the Ecsehalom, prairie soils must have dominated. The shallow hollow observed in a 100 m radius around the mound must have formed during the collection of the soil for the mound.

The part of the mound close to the surface is covered by a blackish brown chernozem formed during the past 4000 years, which is the pedogenic version of the material of the kurgan, but due to the protrusion of the anthropogenic surface it was formed in a stably drier steppe environment compared to the Early Holocene soil. Significant change in carbonate and organic material content was detected only on the surface of the kurgan: the significant organic material content in the 'A' level of the chernozem soil was formed on the island-like protrusion of the surface of the kurgan, while in the 'B' level, a horizon rich in well-developed carbonates was formed. The formation of both horizons indicates an intensive formation of prairie soil following the creation of the kurgan. The raised

geomorphological position, the relatively dry relief and the climatic conditions of the past 4000 years could have played a significant role in the formation of the recent prairie soil covering the surface of the kurgan (Sümegi 2012).

Micromorphological results

Based on the micromorphological research (Bullock et al. 1985; Stoops et al. 2010), three larger layers could be differentiated in the body of the kurgan, which formed on the top of a layer rich in carbonates. The sole, on top of which the whole formation was built, is a sediment horizon containing larger grains, and has low organic material content (**Fig. 10**).

The uppermost part of the mound contains recent soil levels that reflect the current state of the local area. In comparison, the middle level that had been created in its current location due to anthropogenic impact shows re-sedimentation, and occasionally loess-like characteristics. This is why the carbonate content is sometimes elevated in the sediments in the immediate environment of the mound, and occasionally calcareous shells could be detected in the sections. The snail shells found in the midsection of the profile indicate a formative environment different from the present conditions. Above the sole with low organic material content (in the inner part of the kurgan) a layer with low carbonate content can be found, whose grain composition is different from that of both its covering and underlying layers. This layer is contemporaneous with the construction of the kurgan, and is covered by a re-deposited, loess-like sediment. The micromorphological features and shells in the soil layer provide information on the coeval environment and external impacts (Fig. 11).

The quantity of charred wood remains in the area of the sections is high, which occur there as a result of burial activities preceding the construction of the kurgan. The layer, however, is exempt of recent impacts. This is confirmed by the fill of the voids found in the thin-sections, whose material is calcareous, and were filled after small-scale biological activities (Becze-Deák et al. 2007). Based on the current position of calcareous concentrations and the character of the fill of the voids, the soil buried due to anthropogenic impact must have been meadow chernozem. The number of ferrous inclusions indicating inactive impacts is higher, which characterizes the earlier environment of the area, and shows the duration of water coverage (Páll 2012).



Fig. 10.: Organic material and carbonate content (%) of the sediments from the mound and the comparison of the results of the sedimentological and micromorphological analyses (Páll 2012)

10. ábra: A halomtestből kiemelt üledékek szervesanyag- és karbonáttartalma (%), illetve a szedimentológiai és mikromorfológiai eredmények összehasonlítása (Páll 2012)



Fig. 11.: Micromorphological features. 1a–1b: ferrous concretions in the sections; 2: void; 3a–3e: calcareous concretions and skeletal particles of various size and appearance; 4a–4g: skeletal particles of various size and form in the thin sections (excretion, root remains, snail shells, charred wood remains) (Páll 2012)

11. ábra: Mikromorfológiai jellegzetességek. 1a–1b: vasas szeparálódások a metszetekben; 2: üreg; 3a–3e: különböző méretű és megjelenésű meszes göbecsek és vázrészek; 4a–4g: különböző méretű és alakú vázrészek a vékonycsiszolatokban (ürülék, gyökérmaradvány, csigahéjak, szenült famaradványok) (Páll 2012)

Conclusions

Although, these kurgans are important features of the Hungarian landscape, we still do not have enough information of them. Many hypotheses exist that blurs the current point of views. But mounds have high importance, as they hide numerous information dating back to the past thousand years. Each mound keeps valuable information from geoarchaeological, ethnological, onomastical, local historical, botanical, zoological, hydrological, and of course from archaeological point of view.

The geomorphological and landscape historical results show the Ecse-halom is surrounded primarily with alkaline marshes and meadows. Presently it stands on the border between two modern settlements, Karcag and Kunmadaras, along which runs a road of medieval origin, cutting deep into the centre of the body of the mound. It was further distorted during the 20th century, as its southern half was ploughed and used as a rice field. Later, a military observation tower was built on the top of it. Despite all this the surface of the mound is in a fairly good condition and provides a home for a regionally significant, species-rich loess steppe grass.

The kurgan, built in the Late Copper Age/Early Bronze Age by eastern nomadic peoples, comprises two construction layers as indicated by the decrease of magnetic susceptibility results. The examination of organic compounds and carbonate content at various levels showed different values. The distribution of grain size within the section is characterized by mid-sized silt fraction.

The micromorphological studies of Ecse-halom could clearly demonstrate that the levels of accumulation of the original habitat are not present. It deposited during different conditions by anthropogenic effects. The layers originate from the immediate vicinity of the mound, but due to local circumstances they have different characteristics than present-day soils. Micromorphological studies supported the results of sedimentological analysis indicating that a diverse, mosaic meadow chernozem soil with saliferous soil patches run in the study site during the formation of the kurgan at the end of the Copper Age and beginning of the Bronze Age. As a result of mound-grave creation, soil formation transformed. As a result, typical chernozem soil developed on the surface of the mound, forming an island surrounding by solonetz and meadow soils in the study area.

Maps

M.1: First Military Ordnance Map of the Habsburg Empire, 1783, 1:28.800, C.XXII. S.XX. Institute and Museum of Military History, Budapest.

M.2: "Geometrica delineatio totius terreni privilegiati oppidi Cumanicalis Kartzag Uj Szállás", 1784-1787, 1:40,000, Kováts, Gy. Archives of Jász-Nagykun-Szolnok County, T.300.

M.3: Territory of Kunmadaras (Ecse-rét), 1788, 1:14.400. Archives of Jász-Nagykun-Szolnok County, T.95.

M.4: Regulational plan of Berettyó river and Nagy-Sárrét meadow, 1794, 1:86.400, Gasner, L. National Archives of Hungary, S12. XI.132.

M.5: "A' Nagy Kun Karczagi Határ Átnézeti Térképe" (Overview Map of Karcag in the Greater Cumania), 1859, 1:28.800. Archives of Jász-Nagykun-Szolnok County, T.166.

M.6. Second Military Ordnance Map of the Habsburg Empire, 1861, 1:28.800, S.50. C. XL. Institute and Museum of Military History, Budapest.

M.7: Third Military Ordnance Map of the Habsburg Empire, 1883, 1:25,000, 5066/1. Institute and Museum of Military History, Budapest.

M.8: Military ordnance map of the Hungarian Kingdom, 1943, 1:50,000, 5066 NY. Institute and Museum of Military History, Budapest.

M.9: Military ordnance map of Hungary, 1952, 1:25,000, L-34-18-D-b. Institute and Museum of Military History, Budapest.

M.10: Military ordnance map of Hungary, 1956, 1:25,000, L-34-18-D-b. Institute and Museum of Military History, Budapest.

M.11: Military ordnance map of Hungary, 1966-1967, 1:10,000, 409-424. Institute and Museum of Military History, Budapest.

M.12: Topographical map (Hungarian uniform map system), 1977, 1:10,000, 68-413.

M.13: Military ordnance map of Hungary, 1980, 1:25,000, L-34-18-D-b. Institute and Museum of Military History, Budapest.

M.14: Military ordnance map of Hungary, 1991, 1:25,000, L-34-18-D-b. Institute and Museum of Military History, Budapest.

M.15: Military ordnance map of Hungary, 2003, 1:50,000, L-34-18-D. Institute and Museum of Military History, Budapest.

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