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GEM MINERALS AND MATERIALS FROM THE NEOLITHIC AND CHALCOLITHIC PERIODS IN BULGARIA AND THEIR IMPACT ON THE HISTORY OF GEMMOLOGY

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Abstract: Studies of prehistoric (Neolithic to Chalcolithic period) artefacts from the territory of Bulgaria during the past decade revealed a lot of specific gem and decorative minerals and materials: nephrite, malachite, serpentinite, turquoise, jadeite, jet, carnelian, agate and jasper (including heliotrope). Nephrite artefacts in Bulgaria, as well as in some other countries on the Balkans, are widespread during the Neolithic and rare during the Chalcolithic – the nephrite sources are under discussion. A Balkan "nephrite culture" is introduced, which is supposed to be the earliest in the world, compared to the well known Chinese "nephrite cultures". The Varna Chalcolithic necropolis (middle of the V mill. BC) is known with the earliest and largest amount of gold artefacts in the world, including also some copper objects from the copper mines near Stara Zagora. A large amount of beads are also identified as made by malachite (in rare cases with azurite), serpentinite, carnelian, agate, coal (jet), marble and shells. Some of the carnelian beads from Varna display 16+16 facets along their elongation, which is the first record for a constant and complex faceting of hard mineral known so far. An early prehistoric weight system links mineral beads and gold artefacts (the weight unit "van" is introduced, 0.4 g = 2 carats). The first report of turquoise beads for SE Europe is related to the Orlovo prehistoric site (Haskovo district). The "Thracian stone" in ancient sources is identified also as heliotrope, which is known since the Chalcolithic in the Eastern Rhodopes. Some of the artefacts are masterpieces of art and as stage of perfection, thus pointing to the Balkans as a cradle of prehistoric gemmology.

Keywords: nephrite, malachite, serpentinite, jadeite, turquoise, carnelian.

1. Introduction

Archaeomineralogy (archaeological mineralogy; mineralogical archaeology) is an important interdisciplinary science, which is related to the study of archaeological objects of a natural mineral or rock composition, as well as of their sources (including in the broad sense the mineralogical, technological, economic and cultural approach). In certain cases archaeomineralogy is linked also to gemmology (archaeogemmology) as a science for the study of natural and artificial raw materials, which have been used for some sort of jewellery or decorative purposes since the dawn of humankind to modern times (mineral species and aggregates; rocks; bioobjects; glass and pottery, mineral pigments as well as metals and alloys) (Kostov, 2003). Archaeomineralogy can be viewed as part of geoarchaeology as a broader scientific field for application of all the different branches of Earth sciences during archaeological searching, prospecting and study of sites, monuments or artefacts.

A large amount of stone artefacts (lithic material from the Neolithic and Chalcolithic) from museum collections in Bulgaria are studied with respect to specific in colour or genesis minerals and mineral aggregates.

2. Materials and Methods

Among the Neolithic sites are found many nephrite objects, which are considered rare during the next Chalcolithic period. Beside the metal (suggested as the earliest and largest gold treasure; copper) and non-metal (minerals, rocks, pottery, pigments, bioobjects) artefacts in the Chalcolithic graves (V mill. BC) from Varna (as well as in the Durankulak necropolis to the north) in Bulgaria (The First Civilization, 1982; Ivanov and Avramova, 2000; Kostov, 2005b; Gergova and Kostov, 2008 and references therein) are found numerous beads of different shape and composition. The hardest beads are made of red chalcedony (carnelian) and agate. Other mineral beads are made or malachite, serpentinite, coal (jet) and carbonate material (marble or shells) of different shape. The largest amounts of shells used for decorative purpose are those made from *Dentalium spp*. Turquoise was identified to be used as beads for the first time in southern Bulgaria and heliotrope for small lithic tools in the Eastern Rhodopes – in both cases with local material. The origin of jadeite artefacts is under discussion.

The mineral beads or larger stone (mineral and rock) artefacts have been identified and studied by X-ray powder analysis (TUR-M62 and DRON-2: University of Mining and Geology "St. Ivan Rilski", Sofia), optical microscopy (stereomicroscope MBS-9 and Olympus CZ-61), and in some cases (nephrite) – by microprobe analysis (SEM JEOL-SUPERPROBE 733; Geological Institute, Bulgarian Academy of Sciences, Sofia) and electron paramagnetic resonance (EPR; JEOL FA100; Institute of Catalysis, Bulgarian Academy of Sciences, Sofia), as well as by their color. Their morphometrical characteristics and physical properties are compared to similar artefacts on the Balkan Peninsula and elsewhere (Kostov, 2007a). The mineral artefacts are compared also to the known gem and decorative minerals and aggregates, which are described on the territory of Bulgaria (Petrussenko and Kostov, 1992).

3. Results and Discussion

Nephrite. Nephrite $Ca_2(Fe,Mg)_5Si_8O_{22}(OH)_2$ is a Fe-Mg bearing silicate mineral with a double-chain structure, which is classified in the group of amphiboles (a massive fine fiber variety with an intermediate composition in the tremolite-ferroactinolite series). It is known mainly with a pale green or dark green colour. In some cases small black inclusions can be found in the green aggregates, which are attributed to magnetite and other spinel-type minerals. Nephrite and jadeite are both unified in gemmological literature, as well as to some old and popular science papers, as nephrite-jade and jadeite- jade respectively. However, the term jade is not a valid scientific name for nephrite.

According to archaeological data nephrite objects are spread throughout the Neolithic and Chalcolithic period (~VII-V mill. BC in Bulgaria). Recent observations and redeterminations in museum collections revealed a lot of nephrite samples which have been mislabeled or unidentified from a mineralogical point of view (Kostov, 2005a; 2007a and references therein). Small axes and chisels with a fine polish represent the dominant part of nephrite artefacts. Among the artefacts can be found ritual zoomorphic (frog-like) figurines or amulets, two of them with a 4-fold (swastika type) rotational symmetry from the Early Neolithic sites at Kurdjali and Kovachevo (Kostov, 2007a; Fig. 1). A unique and finely polished scepter, 36.4 cm long, found at the Early Neolithic site at Galabnic near Sofia is supposed to be nephritoid (serpentinite) or nephrite-bearing (Kostov, 2007a). Most of the nephrite artefacts are located in Southwest Bulgaria – they have been found mainly in archaeological sites

Fig. 1. Nephrite artefact (axe fragment, 4.6x3.6x0.9 cm) N30939; Kovachevo, Blagoevgrad region, Early Neolithic (left; photo R. Kostov) and zoomorphic nephrite amulet N4532; Regional Historical Museum Kurdjali, Early Neolithic, 4.2x3.7 cm (right; photo V. Alexeev).

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along the Struma valley with the largest number and variety of finds from the Kovachevo prehistoric site. Seventeen nephrite artefacts (represented mainly by small axes) are described throughout the Early Neolithic to Late Chalcolithic at the Karanovo prehistoric site in Central Bulgaria (Kostov and Lang, 2005). At the Varna II Chalcolithic necropolis, a fine nephrite "hair pin" is found with dimensions 12.1x0.7 cm (Kostov et al., 2003).

Electron paramagnetic resonance (EPR) spectroscopy of nephrite from Neolithic artefacts found at sites along the Struma valley revealed three main types of signals: a weak signal of Fe^{3+} (g~4.3; in supposed tetrahedral sites in the structure), a strong and broad signal due to iron-bearing phases and/or Fe^{3+}-Fe^{2+} clusters (g~2) and a 6-component signal from Mn^{2+} (g~2). The EPR data, as well as the microprobe data (including the inclusions), are considered as useful for determination of groups of nephrite samples from at least two possible deposits (occurrences) on the territory of Southern Bulgaria or some other Balkan countries (Kostov, 2007a; 2009).

Neolithic nephrite artefacts, including an amulet, are reported also from Serbia (Antonovic and Stojanovic, 2009), as well as from some other Balkan countries (Kostov, 2007a). No nephrite deposits are known from geological publications in the Balkan region so far, despite of the favorable geological setting with a lot of ultrabasic (serpentinite) exposures. One possible source has to be confirmed in Turkish Thrace, linked to the Yenikoy melange (Özbek, 2009). The well known European nephrite deposits in Poland, Switzerland and Italy have been (re)discovered in the late XIX and early XX centuries, and do not provide information or can not be related to trade routes on the Balkans in prehistoric times. As the Early Neolithic on the Balkans is dated about the VII-VI mill. BC, thus the observed nephrite objects as part of the Balkan prehistoric area are considered as representatives of the earliest "nephrite culture", long time before the well known "nephrite cultures" (Hemudu; Hongshan; Liangzhu; Longshan) in Neolithic China or in the Russian Federation (Kitoi; Glaskovo) (Kostov, 2005a; 2007a).

Jadeite. Jadeite $NaAlSi_2O_6$ belongs to the pyroxene group. In prehistory jadeite artefacts are spread during the Neolithic in Western Europe and on the British Isles (see D'Amico et al., 1995). Such artefacts are known from several archaeological sites in Bulgaria, but their origin as far as from the Alps (Errera et al., 2006; Pétrequin et al., 2009) is under discussion (Kostov, 2007a). The mineral is not found in Bulgaria, but is known from occurrences in Turkey and F.Y.R. Macedonia.

Malachite. Malachite $Cu_2(CO_3)(OH)_2$ is a wide spread copper carbonate mineral. Among the nonmetallic jewellery objects from grave N3 of the Chalcolithic necropolis Varna II is a necklace of short cylindrical mainly malachite beads (in a single case – a bead with admixture of azurite $Cu_3(CO_3)_2(OH)_2$, and in several other cases – with admixtures of cuprite Cu_2O) (Kostov et al., 2003). The study of the nonmetallic mineral artefacts of gemmological interest found in the prehistoric cemetery at Durankulak (NE Bulgaria) display similar beads and admixtures (Kostov and Dimov 2003; Fig. 2).The malachite short cylindrical beads have an average weight of 0.06 g and 0.20-0.25 g. Malachite beads are known since the VII mill. BC from Jericho in the Near East and from Ergani-Cayonu Tepesi and Catalhouk in Turkey (Savascin 1986). In Bulgaria malachite artefacts are reported since the Neolithic and related to malachite are the big copper deposits Mechi Kladenets (Ai Bunar) of Chalcolithic age near Stara Zagora – supposed to be the earliest for Europe.

Serpentinite. Serpentinite is a rock composed mainly by chrysotile $Mg_3Si_{21}O_5(OH)_4$, antigorite $(Mg,Fe)_3Si_2O_5(OH)_4$ and lizardite $Mg_3Si_2O_5(OH)_4$, formed after ultrabasic rocks. This rock is widely used as raw material in the Neolithic and Chalcolithic of Bulgaria. At the Kovachevo prehistoric site the serpentinite artefacts are ~50% among the lithic materials (Kostov, 2007a). The serpentinites

Fig. 2. Malachite (with azurite bead in the middle) beads in a necklace from the Durankulak necropolis, Historical Museum Dobrich NK1441-3, diameter of beads 0.2-0.5 cm (photo V. Alexeev).

are dark-coloured, almost black. They are represented mainly by antigorite, in some cases with talk, and small black oxide inclusions. A fine reel-holder (3.2x3.1 cm; 50.85 g) made of dark serpentinite, probably intended to be used in the drilling process, is known from the Chalcolithic necropolis Varna II (Kostov et al. 2003). Pale green serpentinite short cylindrical beads from the Varna Chalcolithic necropolis have an average weight of 0.025 g and 0.07 g and similar beads are described from the near-by Durankulak site (Kostov and Dimov, 2003).

Turquoise. Turquoise $CuAl_6(PO_4)_4(OH)_8.4H_2O$ is a copper-bearing phosphate mineral with a distinctive blue or bluish green colour. Turquoise was identified among beads from the prehistoric (Neolithic to Chalcolithic) Orlovo site near the town of Haskovo in southern Bulgaria (Kostov et al., 2007). The finds at Orlovo of polished beads and bored bead blanks of turquoise (Fig. 3) suggest a local prehistoric bead workshop for this prestigious material. The most reliable source for the raw material is the Spahievo Pb-Zn ore field south-west of Haskovo, where turquoise mineralization has been found both during underground mining and at the surface as small veinlets. The turquoise beads are considered as the earliest report of this gemmological material in southeast Europe.

Coal (Jet). The gemmological name jet is generally employed in most countries for a black hard, fossilized coniferous wood, which is capable of being carved and can be highly polished. Jet is an amorphous organic non transparent substance with a brown streak and 2.5-4 hardness on the Mohs scale. Coal beads are found both in the Durankulak (Kostov and Dimov, 2003) and Varna (Kostov, 2007a) necropolis. At the second site they are represented by short cylindrical (the smallest beads are with mean weight 0.003, 0.006 and 0.009 g) and barrel-shaped beads (mean weight 0.015, 0.022 and 0.06 g). In previous works they are published as made of lignite, but recent studies point to jet, due to the law refractive values and microscopic observation (Kostov et al., 2010).

Fig. 3. Turquoise bead (1.2x0.7x0.3 cm) from the Orlovo prehistoric site, Regional Historical Museum Haskovo (photo I. Petrov).

Carnelian and agate. Among the metallic (gold and copper) and non-metallic artefacts found in graves from the Chalcolithic sites at Durankulak and Varna are numerous beads of a carnelian and agate composition (Fig. 4). Three main morphological types of beads have been described: type 1 – elongated barrel-shaped; type 2 – elongated with trapezohedral facets; type 3 – short cylindrical. The carnelian and related beads of type 2 have a “constant” number of 32 facets, 16+16 on both sides on the elongation of the bead (the form is a truncated 16-fold trapezohedron), which is considered the earliest in Chalcolithic times complex type of faceting on a hard mineral as quartz (chalcedony is 6.5-7 on the Mohs scale). In the hole of a single carnelian bead a gold mini-cylinder (~2x2 mm) was found, probably with the purpose to tighten up some sort of strip.

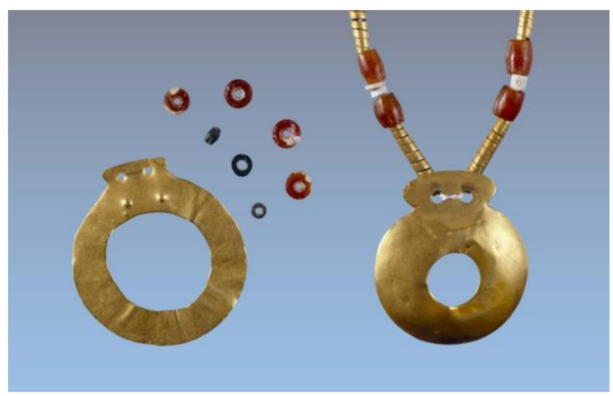

Fig. 4. Gold ring shaped (diameter 3.8 and 3.6 cm) and spiral artefacts, mineral (4 short cylindrical and 4 barrel and faceted red carnelian and 3 white carbonate) and 3 black jet beads from the Varna Chalcolithic necropolis, NI.2771, graves N97 and N254 (photo V. Alexeev).

The mean dimensions (length to width) for the types 1and 2 beads is 1.29 (weight 0.40 g) and for type 3 – 0.54 (weight 0.15 g) (Kostov and Dimov, 2003; Kostov and Pelevina, 2008). Three main types of colour have been distinguished visually among the carnelian beads: orange red, red and

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dark red. It has been suggested that the uniform dark red colour may have resulted from a process of heating of pale red or pinkish coloured carnelian beads (for Indian carnelian see Kenoyer, 1997). Such technique for improving the colour of gemstones is known even today for carnelian in Asia Minor, Iran, Pakistan and India ("mekke tasi", Savascin, 1986). Several operations of manufacture have been suggested for the described carnelian beads – pecking, shaping, faceting, polishing, drilling and tumbling.

The social and symbolic meaning of the chalcedony beads are studied – in the Durankulak necropolis they are distributed equally among male and female graves and in the Varna I necropolis they are typical for the symbolic graves (cenotaphs). In both cases the chalcedony beads are associated with gold and copper objects, as well as with pottery and bone or shell artefacts – they are considered as prestigious jewellery objects (Kostov and Pelevina 2008). The origin and trade routes of carnelian is not yet clarified.

The aesthetic value of brilliance and colour among the Varna mineral beads has been discussed (Chapman, 2007; Gaydarska and Chapman, 2008). Probably all of the beads are linked to a complex weight system among each material and between the different mineral and metal substances (Kostov, 2004; 2007a). The gold objects from the Chalcolithic necropolis at Varna are assumed to be the "oldest gold of mankind" according to their number and quantity. Analysis of the measured weight of the different types of gold artefacts (beads, appliqués, rings, bracelets, pectorals and diadems; Fig. 4) has revealed at least two weight units of 0.14-0.15 and 0.40-0.41 g in correspondence to the carnelian beads weight (Kostov, 2004; 2005b). The second weight is suggested as a basic "Chalcolithic unit" (=2 carats) with the name "van" (from the first letters of Varna necropolis). The weight units as applied to the golden objects are supposed to be the earliest in history and related in later times to other weight units in the Near East, Mesopotamia and Ancient Egypt.

Jasper (including heliotrope). Jasper of different colour is spread in Bulgaria mainly in the Eastern Rhodopes and West Srednogorie regions (Petrussenko and Kostov, 1992). Small red jasper of jasperoid beads are known from the prehistoric Durankulak necropolis (Kostov and Dimov, 2003). Heliotrope (bloodstone; dark green jasper with red spots) is another rare material, which has been found at the Chalcolithic workshop at Sedlare in the Eastern Rhodopes. The "Thracian stone" in ancient sources (Pliny the Elder) is identified also as heliotrope, probably traded in the past from the same area (Kostov, 2007b).

4. Conclusions

Archaeomineralogical studies of prehistoric (Neolithic to Chalcolithic period) artefacts from the territory of Bulgaria revealed a lot of specific (important to the history of gemmology) gem and decorative minerals and materials as nephrite, malachite, serpentinite, turquoise, jadeite, coal (jet), carnelian, agate and jasper (including heliotrope).

Nephrite artefacts are known from prehistoric sites since the Early Neolithic and they "disappear" at the end of the Chalcolithic period. For gemmologists the precision and symmetry of the objects as well as the perfection in the final polishing is a surprise. The possible nephrite sources in Bulgaria, as well as in some other countries on the Balkans are under discussion. A Balkan "nephrite culture" is introduced, which is supposed to be the earliest in the world, compared to the well known Asian "nephrite cultures".

The Varna Chalcolithic necropolis (middle of the V mill. BC) is known as the place with the earliest and largest amount of gold artefacts in the world, including also some copper objects from the copper mines near Stara Zagora. A large amount of mineral beads are also identified as made from malachite (in rare cases with azurite), serpentinite, carnelian, agate, coal (jet), marble and shells. Some of the carnelian beads from Varna display 32 (16+16) facets along their elongation, which is the first record for constant and complex faceting of hard mineral known so far. An early prehistoric weight system links mineral beads and gold artefacts and the weight unit "van" is introduced (0.4 g = 2 carats).

The first report of turquoise beads for SE Europe is related to the Orlovo prehistoric site (Haskovo district) with a local source. The "Thracian stone" in ancient sources has been identified also as heliotrope, which is known since the Chalcolithic in the Eastern Rhodopes.

The territory of contemporary Bulgaria and the Balkans in general can be suggested as a homeland of developing of prehistoric (Neolithic and Chalcolithic) jewellery craft and trade (different mineral beads and other artefacts, faceted beads, precious

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minerals and bioobjects, gold and copper), which is of importance both for the European and world history of gemmology.

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