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# Thracian Golden Wreath from Kabyle, Bulgaria: Chemical Composition

## *Couronne d'or thrace de Kabyle, Bulgarie : composition chimique*

Deyan Lesigyarski[a], Boika Zlateva[a], Valentina Lyubomirova[a], Totko Stoyanov[b] and Ivelin Kuleff[a]

**Abstract:** The chemical composition of a Thracian gold wreath from Kabyle dated to 4th century BC (Thracian age in Bulgaria) was analyzed. Au, Ag and Cu concentrations were determined by inductively coupled plasma-atomic emission spectrometry (ICP-AES) while As, Bi, Cd, Co, Fe, Zn, Sn, Sb, Ga, In, Mn, Mo, Ni, Pb, PGEs (Pt, Pd, Ir, Re, Rh, Ru), Se, Te, Ti, U, W, Zn, and Zr concentrations – by inductively coupled plasma-mass spectrometry (ICP-MS). Data showed that the wreath was made of very well purified gold alloy (Au content between 97.1 and 99.9%). The bivariate plot of the Pd and Pt concentrations showed that the gold used for these artefacts came from four different sources. Due to the lack of analytical data about the platinum group elemental (PGEs) concentrations from different gold sources in Bulgaria it was impossible at this stage to identify the geographical location of the four sources for the gold wreath.

**Résumé :** *La composition chimique de la couronne d'or thrace provenant de Kabyle, daté du IVe siècle av. J.-C.(second Âge du Fer) a été déterminée. Les concentrations en Au, Ag et Cu a été établit par spectrométrie d'émission atomique couplée à une torche plasma (ICP-AES) et les teneurs en As, Bi, Cd,CO, Fe, Zn, Sn, Sb, Ga, In, Mn, Mo, Ni, Pb, PGEs (Pt, Pd, Ir, Re, Rh, Ru), Se, Te, Ti, U, W, Zn et Zr par spectrométrie de masse couplée à un plasma inductif (ICP-MS). Les données ont montré que la couronne a été réalisée avec un alliage d'or assez pure (Au entre 97,1 et 99,9 %). Le rapport entre les concentrations en Pd et en Pt a montré que l'or utilisé pour la fabrication de cet objet provient de quatre sources différentes. À cause du manque de données analitiques disponibles sur les concentration en PGEs dans les différents sources d'or de Bulgarie, il a été impossible à ce stade d'identifier leur location géographique.*

**Keywords:** Thracian gold wreath, Kabyle, Bulgaria, ICP-AES, ICP-MS.

**Mots clés:** *Couronne d'or de Thrace, Kabyle, Bulgarie, ICP-AES, ICP-MS.*

## 1. Introduction

The remains of the ancient town of Kabyle are situated on the eastern edge of the Zaychi vrah hill, near the big elbow of Tundzha River, ancient Tonzos River. Map of Bulgaria with the position of the ancient (respectively modern) settlement of Kabyle is presented in Figure 1. This settlement, which was one of the most important Thracian towns, was reorganized by Philip II of Macedonia becoming a strategic and economic center of that area after his conquest of South-Eastern Thrace in 342-341 BC (Domaradski and Taneva, 1998; Popov, 2002). Later, in 72 BC Kabyle was conquered by the Roman general Marcus Lucullus (Velkov, 1990). During the 2nd century AD the Kabyle fortress served as residence of the *Cohors II Lucensium*, and in the next century of the *Cohors I Athoitorum*. Therefore Kabyle was one of the two main Roman camps in the province of Thracia (Getov, 2003).

The gold samples from Kabyle were collected from the "Big tumulus", located between the modern village of Kabyle and the remains of the ancient settlement. The "Big

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Figure 1: Map of Bulgaria where the position of the ancient settlement of Kabyle is shown.
*Figure 1 : Carte de la Bulgarie montrant la localisation de la ville ancienne de Kabyle.*

tumulus" was about 8 m in height and 40-45 m in diameter. In the north-eastern part of the tumulus a pyre-grave was excavated. The body laid along with his personal belongings, arms and imported vessels (Stoyanov *et al.*, 2010). The funeral could be dated to the last third of the 4th century BC. It was assumed that a military officer of the ancient fortress was buried there (Stoyanov *et al.*, 2010; 2013).

The arms, found in the grave, consist of a sword, many iron spear heads and butts intended for long thrusting spears and javelins. Besides, a bronze horse-bit, which was torn to pieces before being placed under the funeral pyre, was found (Stoyanov *et al.*, 2010; 2013). After Donder, the horse-bit belongs to type VII (cat. Nr. 74-78, Taf. 9) (Donder, 1980), known from excavated finds in Olynthus, Thebes, Dodona and Peloponnesus. Two finds were discovered at Starosel and Dolna Koznitsa in Southern Thrace (modern South-Eastern Bulgaria). Among the remains of the funeral pyre many parts of a bronze wreath, cut into pieces and dispersed all over the fireplace were discovered. The wreath consisted of gilded bronze leafs and gilded clay beads of different forms, as well as large quantities of bronze beads (Stoyanov *et al.*, 2013). Among the remains of the funeral pyre also a twig from a second wreath made of gold, as well as small thin sheets or other gold artifacts were discovered (Stoyanov *et al.*, 2010; 2013). The gold samples investigated in the present study come from the above group of finds.

Despite the relatively large number in Bulgaria of golden artifacts dated to the Thracian period, their analytical study are very rare (e.g. Kuleff *et al.*, 2009). Archaeometric investigations of this period are also scarce in other contries (see e.g. Gondonneau *et al.*, 2001; Guerra, 2004; 2005, 2008; Guerra and Calligaro, 2007; Iliev, 2006; Junk and Pernicka, 2003, Schlosser *et al.*, 2009).

In the present work the analytical data obtained for the gold finds from the pyre-grave in the "Big tumulus" near Kabyle are reported. The analyses were carried out by ICP-AES (inductively coupled plasma-atomic emission spectroscopy) and ICP-MS (inductively coupled plasma-mass spectrometry). The aim of this investigation is to identify the gold sources by determination of large number of elements, including microcomponents. Moreover, due to the lack of literature data, the results obtained in this study will be a good base for comparison in future investigations.

## 2. Materials and methods

### Materials

For the purpose of this study ten gold samples were analyzed. Their brief description with respect to the archaeological context is reported in Table 1 while in Figure 2 pictures of several samples are shown. Since the buried man was initially cremated on a wooden construction of 4x4 m, made in several levels, the grave inventory was exposed to very high temperatures, traceable on some of the gold pieces (Stoyanov *et al.*, 2013).

| Nr. | Description |
|---|---|
| 3000.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil; weight 0.302 g |
| 3001a.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; smelted leaf and fruits from golden wreath; weight 0.333 g (sample from the leaf) |
| 3001b.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; smelted leaf and fruits from golden wreath; weight 0.333 g (sample from the stalk) |
| 3002a.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden beads from wreath; weight 0.422 g (sample from stalk) |
| 3002b.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden beads from wreath; weight 0.422 g (sample from acorn) |
| 3003.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil; weight 0.189 g |
| 3004.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; smelted fruit from golden wreath; weight 0.473 g |
| 3005.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil; weight 0.402 g |
| 3006.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil; weight 0.135 g |
| 3007.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil and covered with golden foil clay bead; weight 0.272 g |
| 3008.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil – smelted in the periphery leaf; weight 0.200 g |
| 3009.KAB | Village of Kabyle, Big tumulus, north-eastern part – funeral-pile; golden foil; weight 0.533 g |

Table 1: Description of the analyzed gold samples from the grave-tumulus near the ancient town of Kabyle.
*Tableau 1 : Description des échantillons d'or analysés provenant des tumulus situés autour de la ville ancienne de Kabyle.*

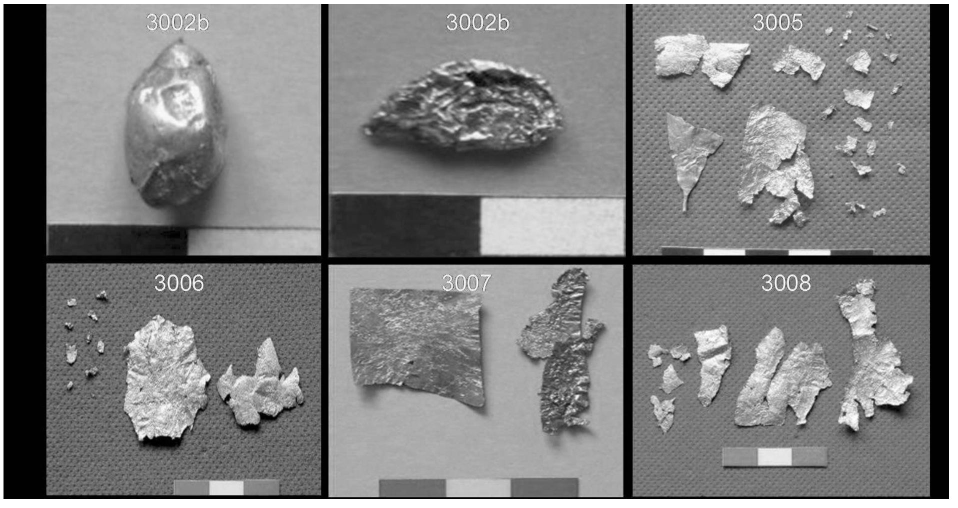


Figure 2: (see colour plate X) Pictures of some of the analysed gold-samples.
*Figure 2 : (voir planche couleur X) Photographies de quelques échantillons d'or étudiés.*

## Methods of analysis

For the determination of the major elements Au, Ag and Cu ICP-AES (Perkin Elmer's Optima 7000 DV) was used. The determination of microcomponents was performed by ICP-MS (Perkin Elmer SCIEX DRC-e system). The concentrations of the following 27 elements were determined (in mg $kg^{-1}$): As, Bi, Cd, Co, Fe, Ga, Hg, In, Ir, Mn, Mo, Ni, Pb, Pd, Pt, Re, Rh, Ru, Sb, Se, Sn, Te, Ti, U, W, Zn, Zr. The weight of each sample was about 5 mg, samples were digested in 3 mL aqua regia ($HNO_3$:HCl = 1:3) and after that diluted to volume of 20 mL with double deionized water. For ICP-MS determination additionally 1 mL was diluted to 10 mL with double deionized water.

The formation of AgCl deposit was carefully controlled. In this case additionally 1 mL concentrated HCl was added to the digested samples.

Suprapur chemicals and double deionized water (MilliQ) were used for preparing all solutions. Working matrix-matched standard solutions were prepared from single standard solutions (Merck GmbH, Germany) with initial concentration 1000 mg $mL^{-1}$ by appropriate dilution.

The validation of the methods was performed analyzing the standard reference materials Gold NA-Au-30 and NA-Au-31 (Aurubis AG, Germany). The results are presented in Table 2.

| | NA-Au-30 | | NA-Au-31 | |
|---|---|---|---|---|
| Element | Experimental value | Certified value | Experimental value | Certified value |
| Ag | 1.01±0.07 | 1.00 | 5.49±0.44 | 5.45 |
| Cu | 102±5 | 99 | 1075±68 | 1062 |
| As | 41±4 | 43 | 108±10 | 112 |
| Bi | 10.0±0.6 | 9 | 106±7 | 100 |
| Cd | 8.0±0.4 | 10 | 79±4 | 82 |
| Co | 9±2 | 10 | 118±20 | 124 |
| Fe | 32±2 | 34 | 796±76 | 806 |
| Mn | 8.0±0.2 | 7 | 60±5 | 62 |
| Ni | 51±2 | 48 | 1100±45 | 1092 |
| Pb | 10.0±0.4 | 9 | 94±5 | 90 |
| Pd | 58±2 | 55 | 1103±36 | 1112 |
| Pt | 56±1 | 58 | 1140±29 | 1152 |
| Sb | 10.0±0.5 | 9 | 110±6 | 102 |
| Se | 12±1 | 10 | 125±16 | 114 |
| Sn | 47±2 | 50 | 760±32 | 772 |
| Te | 11±1 | 10 | 120±11 | 112 |
| Zn | 10.0±0.3 | 11 | 110±5 | 114 |

Table 2: Experimental and certified values for Ag (%) and other 16 elements (mg $kg^{-1}$) in SRMs NA-Au-30 and NA-Au-31.
*Tableau 2 : Les valeurs expérimentales et certifiés pour Ag (%) et 16 autres éléments (mg $kg^{-1}$) dans SRM NA-Au-30 et NA-Au-31.*

## 3. Results and discussions

The results obtained by the analysis of gold samples from the pyre-grave in the "Big tumulus" near Kabyle are reported in Table 3, where the concentrations of the major components (Au, Ag, and Cu) are shown in weight %, while those of the micro components are in mg $kg^{-1}$.

Copper and silver are the major metallic impurities in native gold and most gold ores (Boyle, 1987; Hall *et al.*, 1998; Pohl, 1992). In general, the silver concentration in ancient gold objects from Europe is within 4 to 45% (Hauptmann *et al.*, 2010; Kuleff *et al.*, 2009; Pantazis *et al.*, 2003) with the most common concentrations of about 12-14% Ag (Hauptmann *et al.*, 2010; Pantazis *et al.*, 2003). The values for copper are within 0.5 and 15% (Hauptmann *et al.*, 2010; Pantazis *et al.*, 2003). Such chemical composition indicates the presence of a gold alloy containing a significant part of impurities. The results presented in Table III show different concentration intervals of the macro-components in the studied samples. Gold concentration between 97.1 and 99.9% is an indication that the analyzed samples were made of very pure gold alloy which could be obtained after careful purification of natural gold, most probably through cementation process (Ramage and Craddock, 2000). The relatively low silver and copper concentrations (between 0.07-2.3% and 0.005-0.8% respectively) support this conclusion. Considering that a carat is a 1/24 part of pure metal content in the alloy (when talking about gold and platinum alloys), it follows that the analyzed gold from Kabyle has purity of 23.31 to 23.98 carats (respectively from 970 to 998 in fineness). In comparison, the most common commercial forms of gold nowadays – 14 and 18 carats, are characterized by gold percentage between 58 and 75%. The low levels of the other measured elements (Table 3) once again lead to the assumption that the gold from Kabyle was carefully purified during the manufacturing process (Ramage and Craddock, 2000). However, the uncertainties of some of the elemental concentrations (see Table 3) indicate inhomogeneity of the material. For example, the iron concentration in the samples varied from 256 to 3396 mg $kg^{-1}$, that of platinum – 43 to 129 mg $kg^{-1}$ and of palladium is in the range from 15 to 109 mg $kg^{-1}$, etc. Such results can be explained with the very likely partial smelting of the gold pieces from the wreath during the incineration.

It is believed that concentrations of copper in ancient gold artifacts exceeding 3% are clear indication for alloying gold, possibly aiming to reduce its cost, while alloys with less than 1% copper are naturally occuring. Alloys with 1-3% copper could be natural or human made (Hall *et al.*, 1998). The Cu concentrations in the analyzed findings are, however, too low – less than 0.8%, so most probably a very pure gold alloy was used, most likely obtained after careful purification of natural gold (Ramage and Craddock, 2000).

It is well known that in alluvial deposits, gold can be associated with cassiterite ($SnO_2$), iron hydroxide ($Fe(OH)_3$) and the platinum group elements (PGEs). Gold ores in reef or lode deposits are often associated with quartzite and other metallic minerals such as arsenopyrite (FeAsS), chalcopyrite ($CuFeS_2$), galena (PbS), sphalerite ((Zn,Fe)S), tellurides and tetrahedrite (Hall *et al.,* 1998). The presence of traces of Sn in the studied objects is characteristic of alluvial gold, probably due to riverbed cassiterite and the absence of Sn is a sign for other gold sources: mine or refined gold (Kuleff, 2012; Ramage and Craddock, 2000; Vasilescu *et al.*, 2011). Extremely low Te and Sb were found, but being volatile elements, the refining technology might eliminate them (Ramage and Craddock, 2000). High concentration of iron was also measured in the gold samples. Considering that iron objects were found in the grave (Stoyanov *et al.*, 2013) which might have been in contact with the gold samples could be accepted as a reason for its high concentration and its difference between the samples. It can also be due to the long stay of the samples in the soil. Since iron is a component of common gangue minerals, it does not reflect the geographic provenance of the gold (Vasilescu *et al.*, 2011).

Regarding the small number of analyses of gold artifacts from the same historical period in Bulgaria, comparison with other results appears to be difficult. The wreath from Kabyle is dated to the same period with several golden breastplates from Southern Thrace (see Kuleff *et al.*, 2009) – about 4th century BC. However, in contrast to the Kabyle wreath, the breastplates were most likely produced of natural (not purified) gold. For example, the results obtained by analyses of natural (alluvial) gold from the region (Cojocaru *et al.*, 2003; Kovachev *et al.*, 2007; Stefanova *et al.*, 2007) are in very good accordance with those from the breastplates and they show relatively high silver and copper concentrations, higher than those measured in the samples from Kabyle.

Usually concentration ratios between elements of the platinum group can represent the similarity between the sources of natural gold and the gold that was used for production of the studied artifacts. It is very obvious to assume that the ancient goldsmiths have used only one source of natural gold to make an object. In the Classical and Hellenistic periods when gold was purified probably different sources might have been used.

In Figure 3 the distribution of the analyzed gold samples according to the concentrations of platinum and palladium is presented. The grouping of the samples shows that at least four sources of gold were used by the ancient goldsmiths. The scatterplot in Figure 4 presents the distribution of the samples according the ratios of Ag/Au and Pd/Pt. Considering the grouping of the samples in both bivariate plots the assumption of four sources of gold could be accepted. Samples 3006.KAB, 3007.KAB and 3008.KAB are grouped together in both scatterplots.

Additionally the samples 3003.KAB and 3005.KAB forming a group in Figure 3 are separated from the other samples in Figure 4. Very close concentration values of other trace elements as Sn, Te, Ti, Zn and Zr are also observed.

Furthermore, the similar concentrations of Zn and Zr (see Table 3) of the samples in the respective groups in Figure 3 could also be accepted as a confirmation of the above statement.

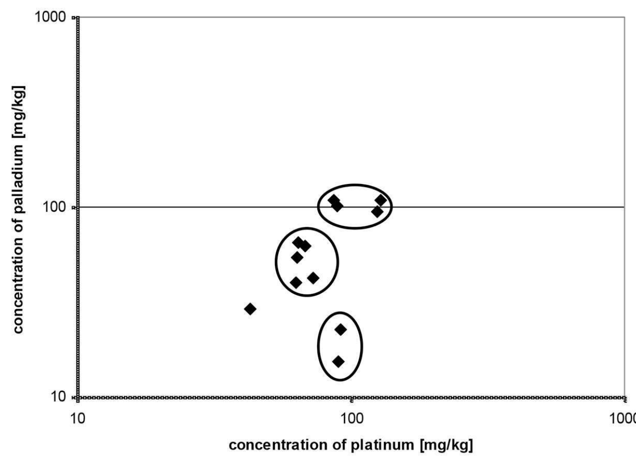


Figure 3: Correlation between the concentrations of platinum and palladium (mg kg$^{-1}$).
*Figure 3 : Correlation entre des concentrations en platine et en palladium (mg kg$^{-1}$).*

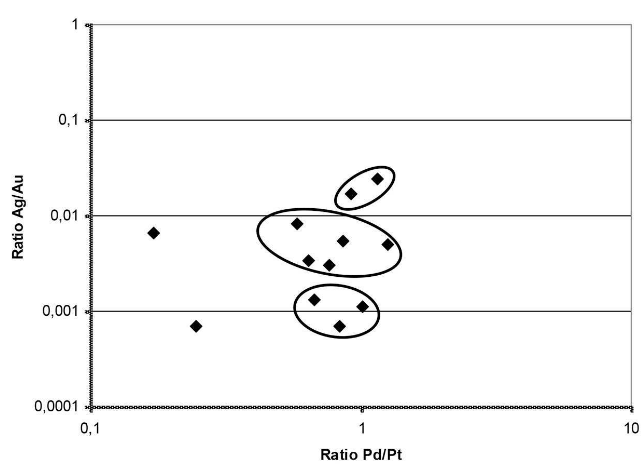


Figure 4: Distribution of the analyzed gold-samples according the Pd/Pt and Ag/Au ratios.
*Figure 4 : Distribution des échantillons d'or analysés en fonction des raports Pd/Pt et Ag/Au.*

| | Sample | | | | | |
|---|---|---|---|---|---|---|
| El. | 3000.KAB | 3001a.KAB | 3001b.KAB | 3002a.KAB | 3002b.KAB | 3003.KAB |
| Au[%] | 98.16±0.73 | 98.88±0.59 | 97.11±0.73 | 99.76±0.80 | 99.67±0.70 | 99.87±0.14 |
| Ag [%] | 1.64±0.48 | 0.49±0.10 | 2.33±0.48 | 0.11±0.03 | 0.070±0.016 | 0.070±0.018 |
| Cu [%] | 0.15±0.05 | 0.09±0.03 | 0.79±0.04 | 0.20±0.04 | 0.005±0.002 | 0.029±0.009 |
| As | 89.30±20.2 | 48.3±8.7 | 60.5±20.2 | 84.3±8.7 | 218±27 | 147±29 |
| Bi | 0.67±0.24 | <0.003 | <0.003 | 6.2±1.7 | 8.9±1.6 | <0.003 |
| Cd | 2.93±0.45 | <0.01 | <0.01 | 10.3±2.6 | 23.3±6.5 | 0.046±0.019 |
| Co | 0.35±0.11 | 0.60±0.16 | 0.6±0.1 | 0.60±0.16 | 0.50±0.04 | 9.4±1.4 |
| Fe | 256.3±46.1 | 403±55 | 458±46 | 826±55 | 772±27 | 3396±183 |
| Ga | <0.004 | <0.004 | <0.004 | 0.4±0.1 | 0.2±0.04 | 3.0±0.5 |
| In | 0.22±0.08 | <0.003 | <0.003 | 0.14±0.04 | 0.3±0.1 | 0.013±0.006 |
| Ir | 0.067±0.013 | 0.50±0.08 | 0.4±0.01 | 0.30±0.08 | 0.35±0.06 | 1.2±0.3 |
| Mn | 15.2±5.0 | 15.0±4.5 | 25.4±5.0 | 23.6±4.5 | 43.0±8.3 | 156±23 |
| Mo | 0.050±0.018 | 0.15±0.05 | 0.2±0.02 | <0.01 | 0.14±0.06 | 0.22±0.08 |
| Ni | 0.63±0.19 | 1.5±0.3 | 1.3±0.2 | <0.06 | <0.06 | 2.3±0.7 |
| Pb | 759±114 | 6.5±0.6 | 5.7±0.9 | 20.7±1.8 | 23.2±4.8 | 110±24.3 |
| Pd | 62.7±11.5 | 109±7 | 102±12 | 65±6 | 108±30 | 22.7±4.5 |
| Pt | 67.9±6.7 | 86.3±9.2 | 88.8±6.7 | 64±9 | 129±30 | 92±29 |
| Re | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 | <0.001 |
| Rh | 0.092±0.037 | 0.19±0.02 | 0.19±0.04 | 0.028±0.008 | 0.07±0.03 | 0.30±0.09 |
| Ru | <0.001 | <0.001 | 0.04±0.01 | <0.001 | <0.001 | 0.048±0.019 |
| Sb | 0.067±0.032 | <0.003 | <0.003 | <0.003 | <0.003 | 1.5±0.5 |
| Se | <0.3 | <0.3 | <0.3 | <0.3 | <0.3 | <0.3 |
| Sn | 6.0±2.1 | 8.2±3.0 | 14.4±2.1 | 2.0±0.7 | 2.9±0.6 | 48.3±7.2 |
| Te | 0.33±0.09 | 0.17±0.04 | 0.20±0.09 | 0.05±0.02 | 1.2±0.2 | 0.40±0.12 |
| Ti | 9.9±2.1 | 3.5±0.9 | 3.6±2.1 | 35.6±1.9 | 37.1±0.7 | 105±11 |
| U | <0.005 | <0.005 | <0.005 | <0.005 | <0.005 | 0.79±0.12 |
| W | <0.005 | <0.005 | <0.005 | 0.07±0.03 | <0.005 | <0.005 |
| Zn | 120±58 | 21±10 | 16.5±1.8 | 108±10 | 235±32 | 205±31 |
| Zr | 4.7±1.7 | 2.5±0.6 | 2.4±0.7 | 5.5±1.1 | 17.6±3.6 | 10.8±2.2 |
| El. | 3004.KAB | 3005.KAB | 3006.KAB | 3007.KAB | 3008.KAB | 3009.KAB |
| Au[%] | 98.86±0.14 | 98.84±0.09 | 99.05±0.07 | 99.19±0.50 | 98.85±0.88 | 99.42±0.40 |
| Ag [%] | 0.30±0.06 | 0.65±0.12 | 0.34±0.05 | 0.54±0.05 | 0.82±0.19 | 0.13±0.04 |
| Cu [%] | 0.14±0.02 | 0.046±0.005 | 0.04±0.01 | 0.025±0.003 | 0.23±0.04 | 0.030±0.008 |
| As | 32.9±4.9 | 113±11.3 | 62.6±24.3 | 36.9±8.9 | 36±14 | 55±15 |
| Bi | <0.003 | 0.69±0.21 | <0.003 | <0.003 | <0.003 | 0.7±0.3 |
| Cd | 0.073±0.038 | 0.74±0.18 | 0.10±0.04 | 1.02±0.19 | 1.0±0.4 | 0.8±0.3 |
| Co | 0.30±0.08 | 1.90±0.48 | 0.8±0.1 | 0.63±0.09 | 0.35±0.12 | 0.8±0.2 |
| Fe | 343±37 | 2120±680 | 1084±68 | 1041±98 | 280±123 | 1790±226 |
| Ga | <0.004 | 2.3±0.6 | 0.40±0.05 | <0.004 | <0.004 | 1.1±0.5 |
| In | 0.60±0.25 | 0.033±0.015 | <0.003 | <0.003 | 0.10±0.04 | 0.20±0.08 |
| Ir | 0.70±0.08 | 0.7±0.2 | 0.87±0.17 | 0.40±0.12 | 0.23±0.08 | 0.19±0.08 |
| Mn | 23.4±4.4 | 508±53 | 99±29 | 47±20 | 37±13 | 25±7 |
| Mo | 0.037±0.017 | 0.29±0.11 | 0.020±0.008 | 0.040±0.008 | 0.45±0.08 | 0.09±0.03 |
| Ni | 2.5±0.9 | 3.8±1.2 | 6.5±2.5 | 1.1±0.3 | 1.37±0.42 | 3.2±0.9 |
| Pb | 123±22 | 48.1±10.6 | 15.7±6.2 | 1.37±0.18 | 4.2±1.1 | 13.9±2.6 |
| Pd | 95.2±6.4 | 15.4±3.1 | 40±17 | 54.6±4.6 | 42.3±20.9 | 29±12 |
| Pt | 125±16 | 90±16 | 62.7±12.4 | 63.4±9.5 | 73±29 | 43±21 |

| | | | | | | |
|---|---|---|---|---|---|---|
| Re | (17±7).10$^{-4}$ | <0.001 | (25±12).10$^{-4}$ | <0.001 | <0.001 | <0.001 |
| Rh | 0.10±0.01 | 0.27±0.03 | 0.18±0.06 | 0.14±0.05 | 0.065±0.027 | 0.08±0.03 |
| Ru | 0.043±0.009 | 0.14±0.03 | 0.13±0.02 | 0.022±0.003 | 0.09±0.04 | 0.003±0.002 |
| Sb | 0.17±0.06 | 0.42±0.09 | <0.003 | <0.003 | 0.023±0.007 | 0.20±0.05 |
| Se | <0.3 | <0.3 | <0.3 | <0.3 | <0.3 | <0.3 |
| Sn | 14.6±2.7 | 41.5±6.2 | 11.2±1.2 | 3.2±0.5 | 264±88 | 18.8±5.5 |
| Te | 0.16±0.05 | 0.36±0.11 | 0.57±0.18 | 0.17±0.06 | 0.093±0.025 | 0.42±0.18 |
| Ti | 0.9±0.3 | 156±16 | 36±14 | 6.9±2.9 | 4.9±2.4 | 17.5±3.7 |
| U | <0.005 | 5.1±0.5 | 0.097±0.015 | 0.037±0.005 | <0.005 | 1.8±0.6 |
| W | <0.005 | 0.007±0.003 | 0.057±0.022 | <0.005 | 0.013±0.006 | 0.010±0.007 |
| Zn | 93.9±16.3 | 217±22 | 97±39 | 39.8±8.0 | 248±109 | 171±72 |
| Zr | 1.5±0.5 | 10.1±2.1 | 5.3±1.7 | 2.1±0.9 | 2.7±2.5 | 5.1±5.9 |

Table 3: Elemental composition of the analyzed gold samples (mg kg$^{-1}$) from the grave funeral-pile from the ancient town of Kabyle
*Tableau 3 : Teneurs mesurées pour les éléments présents dans les échantillons d'or analysés (en mg kg$^{-1}$), provenant des tumulus situés autour de la ville ancienne de Kabyle.*

Unfortunately, at this stage of investigation it is impossible to identify the sources of gold used for the preparation of the golden wreath and the other golden objects found in the pyre-grave in the tumulus of Kabyle due to the lack of analytical data for the concentrations of platinum group elements in different gold sources in Bulgaria. Using only this list of elements it is very difficult to determine the source of gold used be ancient goldsmiths (see also Pernicka, 2013). The possible origin of the golden wreath from North Aegean might be presumed having in mind that the deceased was an officer of the Macedon garrison of Kabyle, and considering the discovered golden stater of Philipp II given to the dead man as a *Charon's obolus*, as well as the imported pottery from South and the Thracian amphorae (for details see Stoyanov *et al.*, 2013).

## 5. Conclusion

Using ICP-AES and ICP-MS samples from the golden wreath found in the tumulus of Kabyle were analyzed. Results indicate that the wreath was produced using very high refined gold alloy (Au between 97.1 and 99.9%).

Comparosin to other gold finds is difficult, because of the lack of literature data. In most of the investigations from the same period the gold objects are found in differerent parts of Europe and Asia (see Gondonneau *et al.*, 2001; Guerra, 2004; 2005; Guerra and Calligaro, 2007; Guerra *et al.*, 2008; Hall *et al.*, 1998; Hauptmann *et al.*, 2010; Iliev, 2006; Kuleff *et al.*, 2009; Pantazis *et al.*, 2003). Recently, archaeometric investigation of gold objects from the same period found in Bulgaria is described in Todorov *et al.*, 2015.

However it could be assumed that the gold objects investigated in this study are produced from very pure gold, obtain through the process of purification. Alas, it is currently not possible to identify the gold sources used by the ancient goldsmiths. The study provides analytical data to be used for comparison with other archaeological finds and is a good basis for future research.

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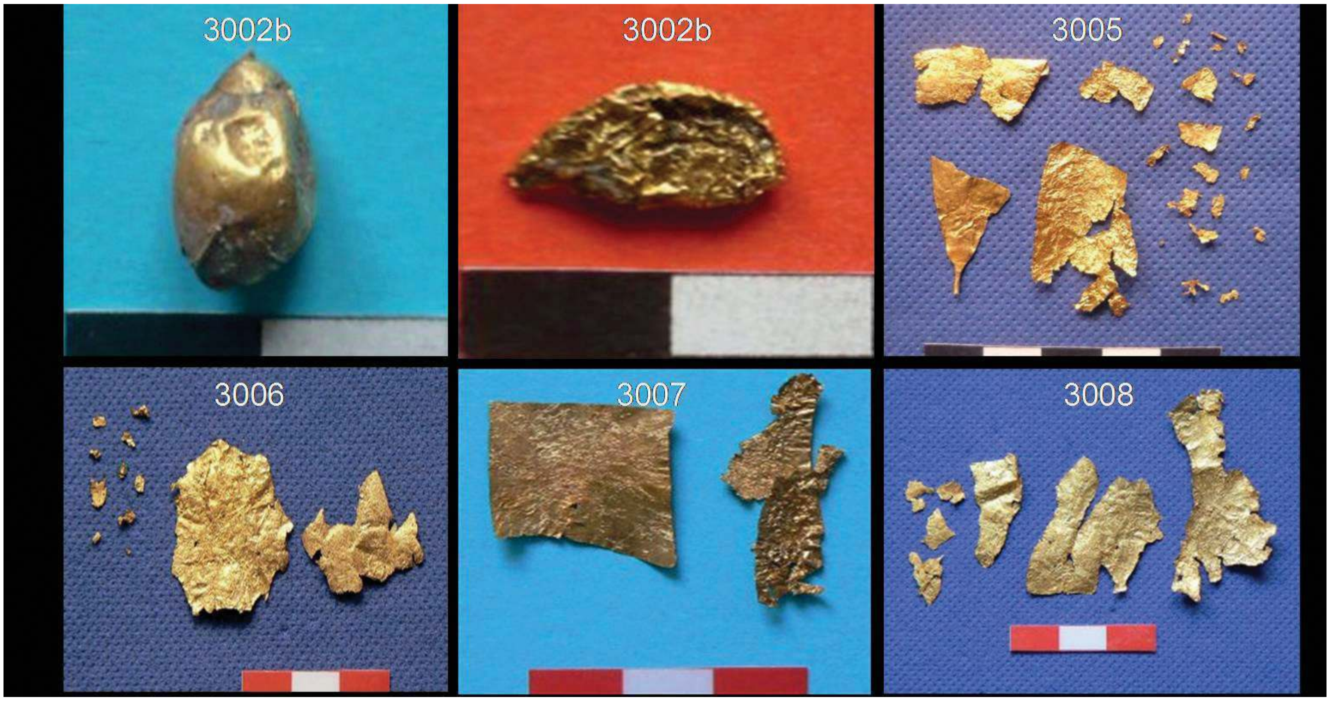


Figure 4 : Deyan Lesigyarski *et al.*, Thracian Golden Wreath from Kabyle, Bulgaria: Chemical Composition (p. 151)

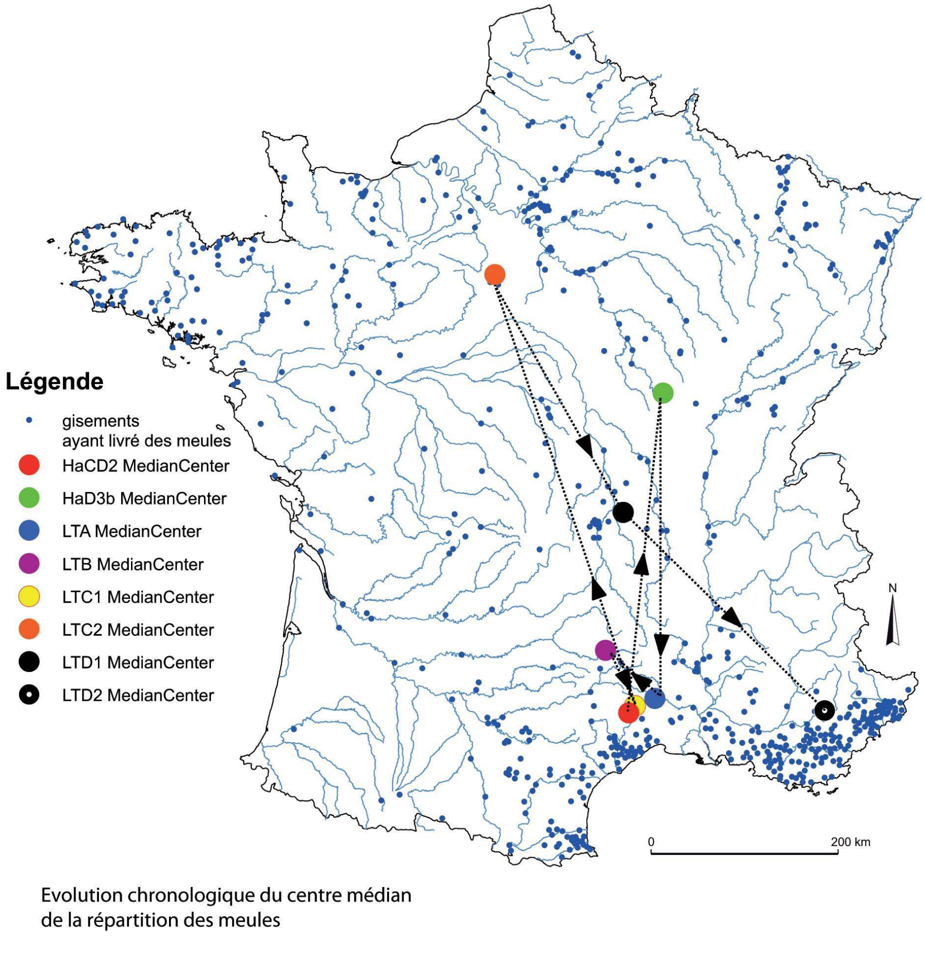


Figure 3 : Olivier Buchsenschutz *et al.*, Une base pour l'élaboration de modèles de peuplement de l'Âge du Fer en France (p. 163)