Golden Artifacts from the Royal Tombs of Ur, Mesopotamia

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Introduction

At the confluence of Tigris and Euphrates, the Early Dynastic period was a time of great wealth and prosperity for the city of Ur and a time of secure long distance trade relations. Countless finds, which were recovered from the tombs of the Royal Cemetery of Ur are not only witnesses for an extraordinary degree of craftsmanship, they are number of precious artifacts crafted from (noble) metals and semi-precious stones. Only very few analytical investigations on these invaluable objects of enormous cultural importance were performed in the past, and those that were mostly were motivated by pressing preservation issues. Based upon the available analyses (Plenderleith, 1934, pp.294), it is apparent that there are different types of gold alloys present in the Royal Tombs and the availability of different types of gold is also supported by the textual evidence in cuneiform tablets (Reiter, 1997). Moorey (1994), however, stated that the interpretation of the textual evidence for the kinds of gold and silver and their origins is anything but simple.

Visual inspection of the gold artifacts shows that there are variations of color and this should be to be expected with gold alloys: The gold objects of Ur appear yellowish, whitish and reddish with changing tints. Details of color variations must have been certainly well-known already in early gold smithing, so that it is reasonable to assume that the different colors could have been produced deliberately.

As a first stage of the larger joint venture between the Penn Museum, the Deutsches Bergbau-Museum Bochum, the Goethe-Universität Frankfurt and the British Museum, a series of non-destructive elemental analyses on selected gold and silver objects were performed in 2009 at the Penn Museum.

Our question were the following:
1. Do the artifacts, which are conventionally termed as “gold” consist of pure gold or are they gold alloys?
2. If the artifacts were made of alloys rather than of pure gold, was the alloy produced deliberately by adding other components e.g. silver and / or copper to the gold, or do such alloys have a natural origin?

Materials and Methods

From the Penn Museum’s collection, we have analyzed 30 objects: Three bowls, one tumbler, 19 jewelery objects and 7 tools and weapons. These artifacts were predominantly selected from two tombs, the tomb of Puabi (PG 800) and the Great Death Pit (PG 1237), from which most of the gold objects were excavated. Most of the gold artifacts from the Royal Tombs are made of sheet gold, which was presumably hammered out, e.g. from a bullion. These sheets were used to manufacture the many meters of ribbons for hair decoration or the large number of leaves used in pendants. Sheet gold was also used for making the famous bulls heads, which decorate the wooden lyres from the Great Death Pit, or for gilding parts of the Rams in a Thicket. Also, the tumblers and vessels were hammered out from bullions. Among all gold objects from the Royal Tombs, massive cast objects are rare. Only a few chisels, a dagger and a socketed adze could be identified as cast.

A handheld portable X-ray fluorescence spectrometer (pXRF) was used for the elemental analysis. The pXRF is an energy dispersive system and applicable for the determination of main and minor element composition of inorganic materials. The method lacks high percision and cannot be applied to obtain bulk elemental compositions; it only analyzes the small area of the surface. Corrosion, as a natural process of weathering, may affect gold artifacts buried in the soil. It is well-known that archaeological gold artifacts as well as natural gold nuggets occasionally may be “coated” with a layer of gold of higher fineness than in the bulk. In this case, the information gained with pXRF would not be represent-

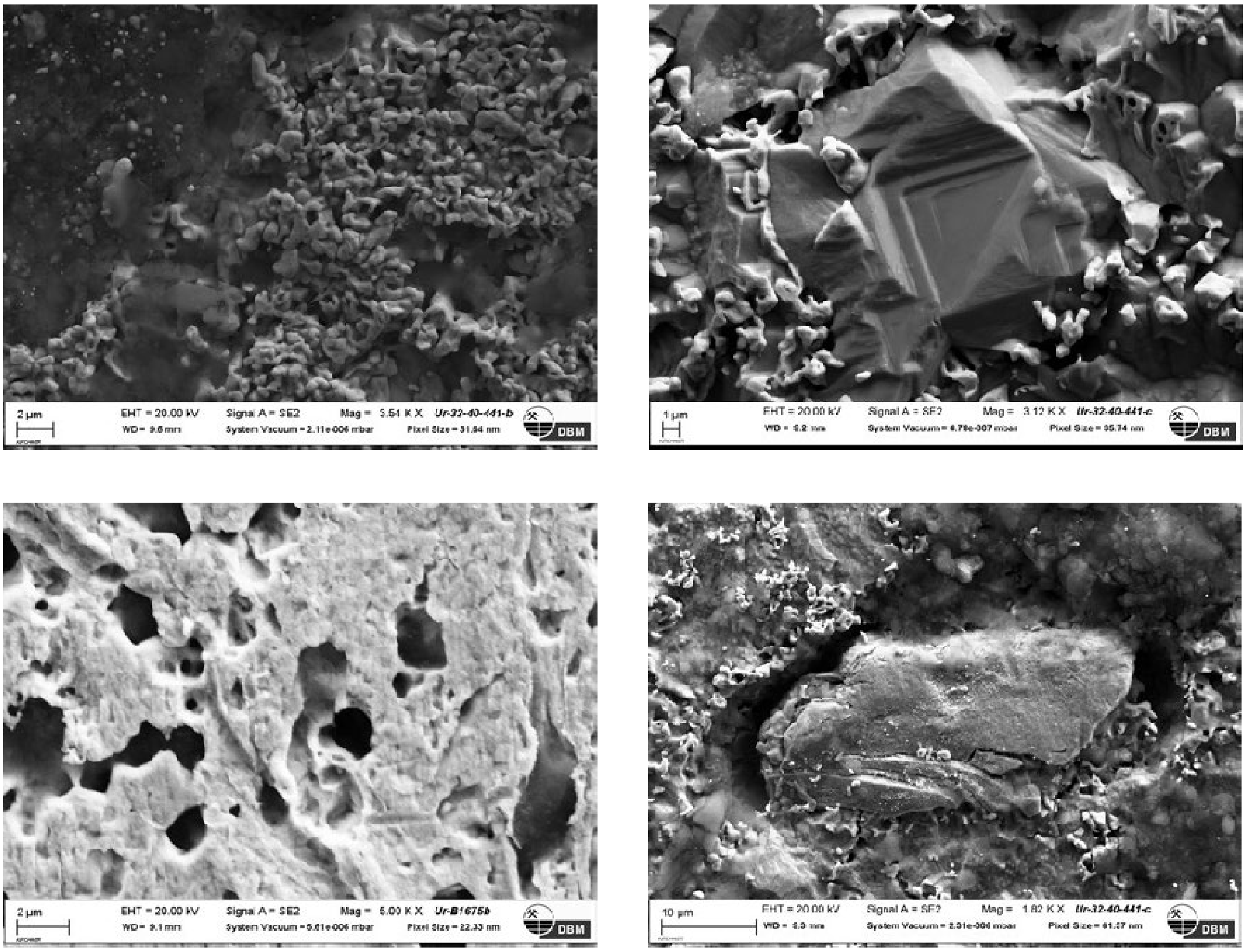

Figure 1.Scanning electron microscope secondary electron images illustrating the textural features of surfaces of some hair ribbons from Ur. a: Formation of a typical spongy texture caused by leaching of silver from gold by corrosion. b: Remains of spongy gold and formation of authigenic gold at the surface. Note network of anhedral gold buds sitting on etched crystals of the AuAg alloy. c: Nanoporous spongy texture caused by dissolution of silver from gold. d: One of the frequent inclusions of platinum-group elements (PGEs) in the gold alloy. Note the well crystallized habitus of the inclusion. Sample numbers under every image.

ative for the composition of the core metal. To explore this potential problem, selected pXRF analyses were cross-checked by electron microprobe measurements of core metal and only very slight differences were found (Jansen, in prep.). Therefore, pXRF was used as a first step, and the method was applied on object where destructive sampling was not permitted. However, only through the sampling of objects can the most high precision analytical methods be applied to gain the largest amount of information.

Besides elemental analyses, we investigated the surficial microtexture of a few samples by scanning electron microscopy (SEM). Microtexture analysis contributes to the understanding of technological information, e.g. crystallization state (cast or worked), dissolution or oxidation (corrosion / leaching processes) and tool marks (manufacturing techniques).

Results

The results are divided into four parts exploring different aspects of the analysis of the gold objects.

Tarnish and Corrosion

The golden color of the objects is frequently stained by red and black patches covering the surface. These splotches consist of thin layers of clay enriched with iron- and manganese hydroxides. They have to be treated as post-burial formations.

The surfaces of gold-silver-alloy objects are dull and show corrosion phenomena visible under the scanning electron microscope. This corrosion led to a spongy texture with irregular particles with gently sinuous outlines forming vermicular structures, which is called cellular honeycomb (Figure 1a). Below this surficial texture

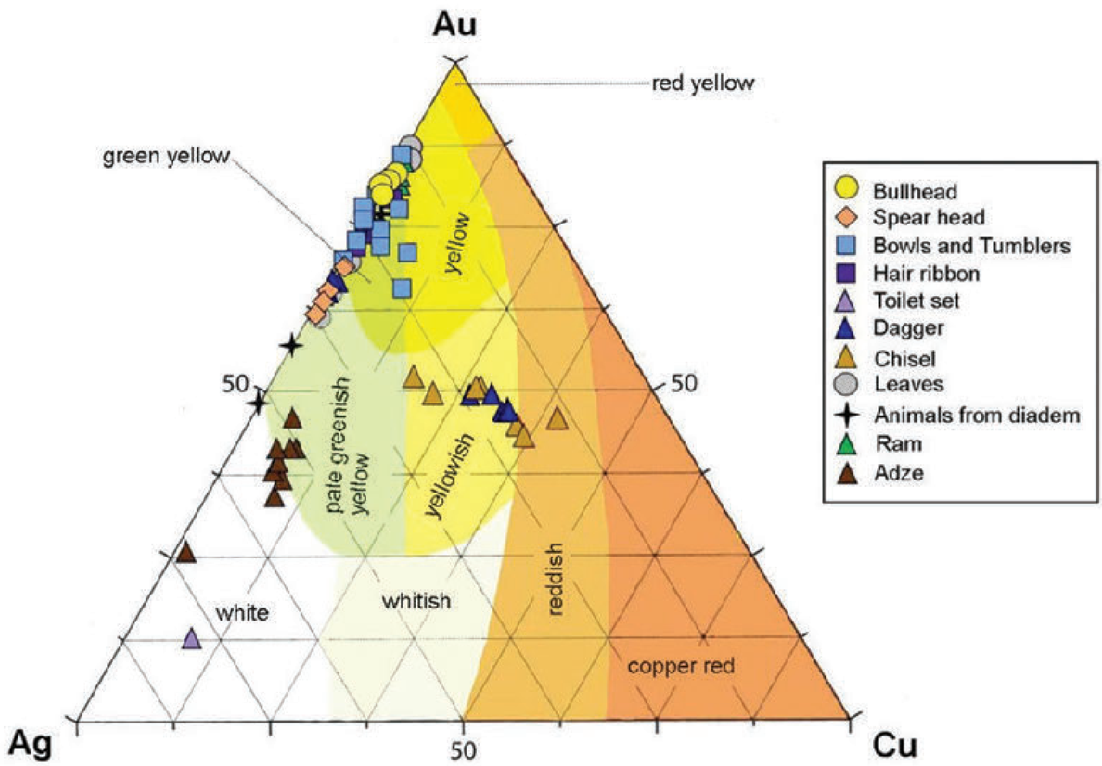

Figure 2. Multiple analyses of gold objects from the Royal Tomb of Ur plotted in the ternary system Au-Ag-Cu indicating the variation of colors achieved by specific mixtures of gold, silver and copper. Note that the variation of colors depends on the portion of single alloy components. This results in stability fields for the different colors of gold alloys. The analyses indicates that the majority of the objects plot within the field of yellow and greenish-yellow gold, while objects high in copper plot within the reddish-yellowish field.

etched crystals of AuAg are visible (Figure 1b). The material is interspersed by sets of minute pores (Figure 1c).

These spongy surfaces and pores result from silver dissolution from the gold-silver alloy as it is the case with widespread natural silver depletion of nuggets in sedimentary context (Falconer and Craw, 2009, p.89). As for the gold objects, silver was dissolved out of the alloy body and led to a precipitation of silver chloride. This effect was probably caused by the very typical soil conditions of the region during burial: Oxidation in salt-rich marshes and *sabkhas* with a surplus of chlorides (Möller, 1995). In the archaeological context, this phenomenon also occurs in artificially depleted gold observed at mid-1st millennium BC Sardis, where gold was refined. Comparable textures were described by Meeks (2000) and Geckinli, et al. (2000).

Chemical composition

Silver, and subordinately copper, are the predominant alloy components of the gold objects from the Royal Tombs. Additional traces of elements such as Sn, Zn, Pb, As, Fe and Ir are rare and in most cases below 1 wt. %. The variety of the gold alloy compositions as found in the objects from the Royal Tombs are well represented by the ternary phase diagram Au-Ag-Cu (Figure 2). The silver contents in the gold objects range between c. 10 and 35 wt. %. Many hair ribbons, leaves and other jewelery made of hammered foils belong to these alloys. There is no evidence for pure gold. This is in good correspondence with the co-existence of geological gold and silver occurences in natural alluvial placers. The silver-containing gold objects were thus most probably manufactured using natural alluvial gold-silver alloys (placers) without any further processing. Parting, i.e., a metallurgical separation process to remove silver from gold, as suggested by Levey (1959), is not known to have been practiced at Ur in the middle of the 3rd millennium BC.

Objects higher in silver (c. 50 wt.%) rarely occur and are connected with increased copper concentrations (5-8 wt.%) in the Royal Tombs. One of these silver-rich objects is a socketed adze, which is one of the few examples represented in the Royal Tombs that was cast instead of hammered. The adze has a distinctly whitish tint overall and differs from the color of other gold artifacts. The alloy composition would solidify in the range of c. 900 - 950 °C, much lower than the melting point of pure gold (1063 °C). We suggest objects of this alloy to be a deliberately produced alloys. Even if native gold may contain such high silver concentrations (Antweiler and Sutton, 1970), the copper is much too concentrated. Natural

Figure 3.Dagger # 30-12-550, PG 1054. The dagger is composed of several types of gold. For the decoration of the hilt, greenish-yellow gold was used to manufacture some 400 nails, the collar and the sheets (left). The blade consists of a gold-silver alloy high in copper, gilded by a depletion gilding process. The golden surface is only in parts preserved (right). Length of the dagger: 33 cm. A similar feature is to be observe on the blade of a dagger from the Baghdad-Museum (Internet: www.baghdad-museum.org).

gold bears only small quantities of copper (< 1 wt. %); slightly higher concentrations maybe caused by incomplete separation of gold from copper minerals during the washing of alluvial gold (Hauptmann, et al., 2010).

One of the most exciting results of our analyses is that, beside the variety of gold-silver alloys, also alloys with extraordinary high copper contents (23 – 40 wt.%) occur (Figure 2). This concerns several objects, which were all made of massive (cast) metal. The dagger blade of tomb 1054 (# 30-12-550) and two chisels (# B16724 and # B16725) from Pu-abi's grave (PG 800) belong to this group. The dagger is made of various components that consist of differnt alloy compositions. The excellently burnished blade is fabricated in one piece of a copper-rich gold-silver alloy. However, it shows slightly varying pinkish to yellowish and shining tints of gold (Figure 3). Above the guard on the hilt and on the pommel c. 400 nails with spherical heads of pale greenish-yellow color were inserted on gold sheets. These are made of a gold-silver alloy low in copper. No sheeted metal objects were produced from this high-copper alloy. We presume that these gold-silver-copper alloys were mixed deliberately with (tin-containing) copper.

Depletion gilding

The surfaces of the dagger's blade and the chisel have much higher gold and silver contents than the cores, which consist of the above described gold-silver-copper alloys. The metallurgical technique to enrich gold on the surface of gold alloys containing copper is called depletion gilding. The technique is based upon the following metallurgical procedure: An object is cast from a homogeneous ternary gold-silver-copper alloy. The entire range of gold-silver-copper ratios is possible. The variable compositions of such alloys result in characteristic colors that vary from red-orange to red gold. The lowest solidification temperature of this alloy is around 900 °C, which is about 150 °C lower than the melting points of its individual metals. The lowered melting point of the gold-silver-copper alloys is of major advantage for the goldsmith's work, especially during further casting or other processing techniques.

After an object is brought to its final shape and design, it is heated in an open fire to selectively oxidize the copper. Since silver and gold have less affinity to oxygen than copper, they remain in metallic condition. In contrast, copper reacts with oxygen to copper oxides, which form a reddish or black crust. Finally, this crust is mechanically removed by hammering. Possible remains of copper oxide will be leached out in a boiling of plants high in tannic, oxalic-, apple-, citric acid or in urine. Finally, a spongy gold-rich surface results from this process, which can be compacted by hammering and polishing.

At the current state of research, it appears that these two examples from Ur are only depleted in copper and the gold to silver ratios remain constant. We found no evidence that chemical treatments, such as heating with iron compounds and chlorides, were applied to selectively leach out the silver from the surface of the alloy, and this technique may not have been known. Had the technology of separating gold and silver (cementation) existed at this time, then one would expect to find objects from Ur made of pure gold, but this is not the case. Depletion gilding from gold-silver-copper alloys, which are named *tumbaga*, with depletion of both copper and silver was described as a characteristic technique for the Pre-Columbian South Americas (Lechtman. 1973; 1979;

Ingo, De Caro and Bultrini, 2007; Scott, 2012). It was suggested that this technique was first invented there. In compliance with previously published data from Ur (La Niece, 1995; Hauptmann and Pernicka, 2004), and our own observations, it is confirmed that a variation of depletion gilding was already known at the time when the Ur objects were manufactured.

Inclusions of platinum group minerals

One of the most remarkable characteristics of the gold artifacts from the Royal Tombs are abundance of silvery shining inclusions, which are composed of platinum group elements (PGE), particularly consisting of alloys of osmium, iridium and ruthenium. They occur in unusual and unparalleled quantities in most of the gold artifacts from the Royal Tombs (Jansen, et al., 2016) (Figure 1d). Generally, PGEs are contaminants in placer gold neighboring (ultra-) basic rock, where such PGE-bearing minerals occur in primary mineralizations. Primary gold deposits are geologically very different from this type of deposit. Gold and PGEs are thus associated with each other only in geological environments with secondary mineral deposition such as placer deposits, in which the PGE minerals can combine mechanically with the placer gold. The presence of PGEs in gold artifacts confirm that the gold, which was used for the production of the objects originated from secondary placers rather than from a primary gold source in solid rock. The high frequency of the PGE inclusions might be an indication that the gold was not, or often, re-melted (Meeks and Tite, 1980), because then the PGE being solid inclusions would probably have been reduced step by step from the liquid gold. It may be possible to determine the provenance of the PGE-containing placer gold used for the Ur artifacts through isotope analysis as reported by Jansen, Hauptmann and Klein in this volume.

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