

Interrelation Between Gold and Tin: A Historical Perspective

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Abstract

The historical evidence for the occurrence of some tin in gold objects and some gold in tin mini-ingots from various parts of the world has been discussed. It has been shown that their presence in each other is not intentional, but is due to their primary metal extraction process used. Both gold and cassiterite minerals were obtained as alluvial placer deposit in ancient and medieval times. Evidence for the simultaneous occurrence of gold in the same vein or alluvial placer deposits in various parts of the world has been presented. This type of deposit could be concentrated to either a cassiterite-rich or gold-rich concentrates by panning, Even the most efficient panning process used in those times, would have resulted into some amount of gold in cassiterite concentrate and cassiterite in gold concentrate. Such concentrates would give rise to some gold in the extracted primary tin, and some tin in the extracted primary gold. If tin bronze was used for making alloyed gold containing higher amounts of copper, at least a part of tin present in the alloyed gold might have originated from it.

Introduction

The occurrence of tin in certain historical gold objects has been reported in the literature. Also, there is available a historical literary reference of Indian origin which mentions the occurrence of gold in tin mini-ingots. It is most unlikely that either gold in tin or tin in gold were added intentionally in antiquity. It appears that they were present in each other naturally. In the present paper the archaeological and literary evidence related to gold objects containing tin, and tin mini-ingots containing gold have been reviewed. The reasons for the natural presence of tin and gold in each other have been discussed. The word “historical” has been used in a wider sense in this paper.

The Occurrence of Tin in Gold

There are several references discussing gold objects and pieces found from archaeological excavations, which inform us about the presence of tin in historical gold objects. The chemical analysis, based on emission spectroscopy, of about 3300 European gold artifacts dating from the Bronze Age to the beginning of the Christian era indicates that these contain tin, and in some cases platinum too¹. Gold objects belonging to the close of the Bronze Age in Central Europe, i.e. around the turn of the 2nd to the 1st millennium BC, were found from a wide geographical area extending from the Balkans to Spain and Ireland. These are characterized by their relatively high tin content (> 0.1 wt%) and copper content around 2 wt%, and are designated as “gold N”. The centroid of the distribution of this type of gold appears to lie in the present Schleswig-Holstein/Denmark, the British Isles and the areas of Brittany accessible therefrom, and the western part of the Iberian peninsula (Figure 1). Tylecote² has given the chemical analyses of some European gold objects on the basis of the results reported by Hartmann³ and Eluère⁴, as shown in Table 1. The tin content varies between 0.002-0.43 wt% together with copper content in between 0.01-3.4 wt%. It is interesting to note that there are some gold objects, which have low copper in the range 0.01-0.04 wt%. There is no definite trend in their tin content. While one object (from Kerouaren) has very low tin of the order of 0.002 wt%, two gold objects (from Rondosse and Kerivoa) have tin content as high as 0.15-0.23 wt%. Müller⁵ has reported chemical analysis of gold finger rings of La Tène period between the fifth and the first centuries BC, from Switzerland and the Upper Rhine in Baden (Table 2). Many gold rings contained tin in small amounts in the range 0.003-0.13 wt%. About 7 kg of gold based and silver based jewellery were found in fortified dwellings in the Celtiberian area of the Douro river, during the period 1980-1987. These hoards were buried at the end of the first century BC. Perea and Rovira⁶ have given the chemical analysis of several of these gold objects, which shows that these contain tin in the range 0.02-0.3 wt% (Table 3). Many historic Iberian gold

Table 1*Chemical composition of gold artifacts from Hungary, Germany and France*

Site	Description	Ag, wt %	Cu, wt %	Sn, wt %	Cu:Sn	Ref. No
<i>Hungary</i>						
Bihar (1500 B.C.)	Bowl 1	< 11	0.30	0.004	98.69:1.31	3
	Bowl 2	18	0.44	0.010	97.78: 2.22	3
	Bowl 3	< 15	0.38	0.011	97.19: 2.81	3
	Bowl 4	20	0.29	0.019	93.86:6.14	3
<i>Germany</i>						
Fritzdorf	Beaker	10	0.22	0.006	97.35: 2.65	3
Terheide	Bowl	15	2.20	0.100	95.65: 4.35	3
Ostfriesland	Bowl	17	2.10	0.057	97.36:2.64	3
Gronnebek	Bowl	12	0.35	0.065	84.34:15.66	3
Albersdorf	Vase	17	3.40	0.190	94.71:5.29	3
Albersdorf	Beaker	21	0.43	0.430	50:50	3
Unterglauheim	Beaker	16	2.10	0.150	93.34:6.66	3
Unterglauheim	Beaker	22	2.30	0.170	93.12:6.88	3
<i>France</i>						
Cazevielle	Bead	2	0.28	0	100:0	4
Kerouaren	Appliqué	5	0.02	0.002	90.9:9.10	4
Kerivoa	Lunulae	7	0.05	0.004	92.59:7.41	4
Kerouaren	Twist	10	0.12	0.14	46.15:53.85	4
Kerouaren	Headband	5	0.31	0.032	90.64:9.36	4
St. Martin / L	Ring	6	0.16	0.023	87.43:12.57	4
Lannion	Spacer	15	0.33	0.067	83.12:16.88	4
Alvignac	Ring	13	0.21	0.015	93.34:6.66	4
Rondossec	Collar	15	0.01	0.15	6.25:93.75	4
Kerivoa	Lunulae	8	0.04	0.23	14.81:85.19	4
St. Potan	Lunulae	10	0.06	0.043	58.25:41.75	4
Kerivoa	Torque	15	0.05	0.019	72.46:27.54	4

**Figure 1***Distribution of material group "Gold N" ¹*

artifacts contain tin below 1.0 wt% together with varied amounts of copper⁷. Similarly many Central European Celtic gold coins contain tin as minor elements in the range 0.1-0.5 wt%⁸.

The Cu:Sn ratio of all the gold objects reported in Table 1-3 has been calculated and is shown in the respective table. Figure 2 shows the relationship between copper and tin

contents of gold alloys reported in Table 1-3. No particular trend is observable. However, it can be seen that a large proportion of the gold objects studied have copper within 4 wt% and tin within 0.15 wt%.

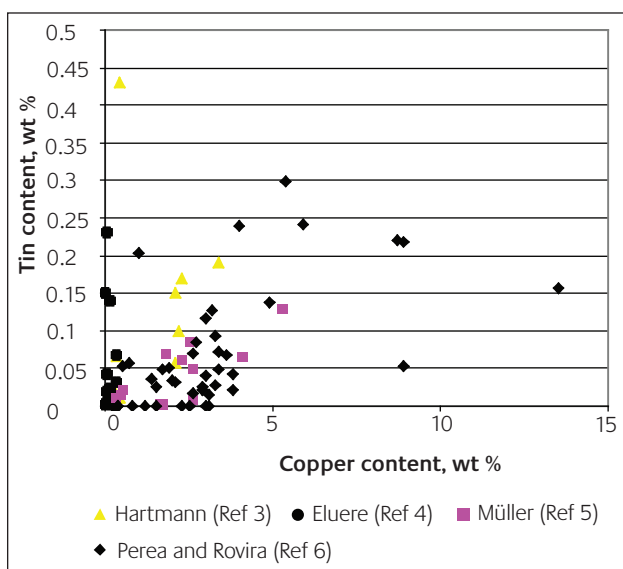
Eluère⁹ has drawn our attention towards an interesting feature of early gold objects. She states that many early gold objects are fairly rich in sand and other river-bed minerals, some times even cassiterite. Eluère and Raub¹⁰ have stated that the earliest archaeological gold objects, e.g. of Varna, were either totally or partially sintered from alluvial placer gold particles. They have studied the microstructure of such a gold leaf, and found that it contains non-metallic inclusions derived from the residual sand and river bed minerals present in the panned alluvial gold concentrate. Such historical gold objects were manufactured by sintering of panned alluvial gold dust along with the residual sand and riverbed minerals, followed by beating/hammering into the required shape. Presence of the small amounts of non-metallic impurities does not affect the malleability of gold in any significant manner. This shows that in ancient times gold and cassiterite were found together in the panned concentrate.

The Harvard-Sardis team carried out extensive archaeological excavation in Sardis, and several important finds related to gold refining were found there. Sardis was the capital of Lydia in the center of western Anatolia. Croesus

Table 2

Chemical composition of La Tène period gold finger rings from Switzerland and the upper Rhine in Baden⁵

Site	Analysis. Ref. No	Wt. of finger ring, gm	Au, wt %	Ag, wt %	Cu, wt %	Sn, wt %	Cu:Sn
Münsingen	Ha 0495	1.8	68.4	30	1.6	0.003	99.81:0.19
Münsingen	Ha 0476	2.6	57.4	37	5.3	0.13	97.61:2.39
Münsingen	Ha 0474	9.7	92.4	7	0.5	0.021	95.97:4.03
Muri	Ha 0472	8.0	87.5	11.5	0.46	0.015	96.84:3.16
Münsingen	Ha 0494	4.4	71.4	26	2.6	0.009	99.66:0.34
Münsingen	Ha 0493	5.5	90.3	8	1.7	0.003	99.82:0.18
Belp	Ha 0475	3.6	66.9	29	4.1	0.065	98.44:1.56
Kirchenthurnen	Ha 0491	4.7	82.4	15	2.5	0.085	96.71:3.29
Kirchenthurnen	Ha 0492	7.6	87.3	10	2.6	0.049	98.15:1.85
Münsingen	Ha 0479	3.5	70.6	27	2.3	0.061	97.42:2.58
Muri	Ha 0473	8.0	89.6	8.5	1.8	0.070	96.26:3.74
Stettlen	Ha 0471	7.0	97.2	2.5	0.31	0.011	96.57:3.43

**Figure 2**

Relation between copper and tin content of Au-Ag-Cu alloys containing tin

ruled Lydia from 561 to 547 B.C. The dating of the gold refinery resembles that of the reign of King Croesus (561-547 B.C.). Meeks¹¹ has described these finds in detail, together with their chemical analysis. More than 300 pieces of gold were recovered from this site. They were in either hammered foil or round/irregular globule shape. The chemical analyses of many of these gold globule samples were carried out on the as received surface on SEM with EDX facility. An interesting point to note is that about 80 gold pieces contained tin in the range 1.4-8.6 wt%. One specimen contained tin as high as 19.3 wt%. The chemical analyses of some of the gold pieces found in Sardis are given in Table 4. Some gold globules were covered with a fused non-metallic layer; the analysis of which is also given in Table 4. The outer layer contained very high amounts of tin, together with other elements. Meeks is not very sure about the origin of tin in these gold pieces. In this connection, however, he mentions the Agatharchides' description of gold parting in which he has used cement containing both lead and tin. Apparently,

he thinks that tin in gold has come from the surroundings, i.e. cement mixture. It is interesting to note that only about 8 gold globules out of about 100 globules analysed have shown the presence of tin in various amounts.

The refining of gold at Sardis was carried out in solid state, which was commonly known as cementation or parting process. In principle, it consisted of heating impure gold containing silver as the major impurity along with a cement mixture consisting of common salt and a carrier such as alum, sulphatic material, brick dust or clay. As a result, the silver is removed from the gold. Craddock¹² has discussed the type of carrier used in gold refining at Sardis, and thinks that the cement used was probably just common salt. However, he has not ruled out the presence of carrier in the cement. He has stated that the surface deposits on the treated gold foils from Sardis have the appearance of a cement with an inert carrier such as brick dust or clay.

It is proposed that actually these starting gold globules found from Sardis, also contained tin apart from silver. During parting reaction, the tin would also get removed along with silver (Appendix 1). This hypothesis also gets support from the fact that the outer mineralized layer covering the gold globules contained a large amount of tin. These tin containing gold globules are actually either partly refined globules or the starting unrefined raw globules.

The Occurrence of Gold in Tin

There is an Indian description of early fourteenth century A.D. on the occurrence of gold in tin mini-ingots, written by Ṭhakkura Pherū. This reference has not yet received the attention of the historians of metallurgy. Ṭhakkura Pherū was initially a treasury officer of Alauddin Khilji. After the death of Khilji, Ṭhakkura Pherū became Mint Governor in the kingdom of his successor Sultan Kutubaddin in 1316 A.D. He wrote several books on subjects such as gemology, metallurgy, astrology, mathematics, coins etc. In one of his books "Dhātūtpatti", Ṭhakkura Pherū has written two verses in

Table 3

Chemical analysis of some of the gold objects obtained from the Celtiberian area of Douro river⁶

Ref. No.	Description	Au, wt %	Ag, wt %	Cu, wt %	Sn, wt %	Sb, wt %	Cu:Sn
PA1001A	Belt (band)	73.6	23.2	3.1	0.015	nd	99.52:0.48
PA1001B	Belt (end)	75.5	20.7	3.6	0.067	nd	98.18:1.82
PA1001C	Belt (back fastener)	72.5	24.2	3.1	tr	0.007	100:0
PA1001D	Belt (obverse fastener)	75.9	20.7	3.0	0.041	tr	98.66:1.34
PA1002	Torc (necklace)	85.9	10.5	3.4	0.049	0.009	98.58:1.42
PA1003	Torc (necklace)	72.6	18.0	8.7	0.221	0.018	97.53:2.47
PA1004A	Torc (necklace)	76.0	19.4	3.8	0.043	tr	98.89:1.11
PA1004B	Torc (end piece)	86.9	10.8	2.1	0.031	0.012	98.55:1.45
PA1004C	Torc (end piece)	85.3	11.8	2.6	0.070	0.011	97.38:2.62
PA1006A	Symmetr. Brooch (lining)	83.7	12.2	3.8	0.022	nd	99.43:0.57
PA1017	Pendant ear-ring (down side)	85.1	12.5	2.0	0.034	0.012	98.33:1.67
PA1017A	Pendant ear-ring (top side)	86.5	11.1	1.9	0.050	0.015	97.44:2.56
PA1018A	Pendant (body)	94.6	4.7	0.4	nd	0.017	100:0
PA1018B	Pendant (ring)	94.9	4.1	0.8	nd	0.018	100:0
PA1019	Ring	93.3	4.9	1.5	nd	0.013	100:0
PA1019A	Pendant (body)	87.5	9.4	2.9	0.026	0.011	99.12:0.88
PA1019B	Pendant (ring)	89.4	7.4	3.0	nd	0.014	100:0
PA1020A	Pendant (body)	93.3	5.3	1.2	nd	0.018	100:0
PA1020B	Pendant (ring)	93.2	5.0	1.5	0.026	0.017	98.30:1.70
PA1021A	Pendant (body)	89.8	7.4	2.6	0.018	0.003	99.32:0.68
PA1021B	Pendant (ring)	89.1	7.7	2.9	0.022	0.014	99.25:0.75
PA1022	Finger ring	88.6	7.3	3.3	0.094	0.016	97.23:2.77
PA1023	Ring	94.1	5.3	0.4	nd	0.016	100:0
PA1024	Pendant	79.0	17.4	3.2	0.128	0.006	96.16:3.84
PA1025A	Ring (twist)	84.7	11.8	3.3	0.028	0.005	99.16:0.84
PA1025B	Ring	85.6	11.4	2.7	0.085	0.006	96.95:3.05
PA1026A	Finger ring (flat front)	95.6	3.6	0.5	0.053	0.014	90.42:9.58
PA1026B	Finger ring (ring)	95.3	3.5	0.7	0.058	0.031	92.35:7.65
PA1028	Ring	93.1	5.2	1.4	0.035	0.022	97.56:2.44
PA1030	Finger ring	89.5	8.5	1.7	0.048	0.096	97.26:2.74
PA1031	Finger ring	65.9	32.3	1.0	0.204	0.024	83.06:16.94
PA1038A	Brooch (knob)	92.5	4.6	2.5	nd	0.029	100:0
PA1038B	Brooch (pin)	91.9	4.6	2.3	nd	0.044	100:0
PA1039B	Ring brooch (bow filigree)	53.3	39.7	5.9	0.242	nd	96.04:3.94
PA1039C	Ring brooch (ring knob)	53.1	40.9	4.9	0.137	0.013	97.28:2.72
PA1039D	Ring brooch (coating wire)	56.9	37.0	5.4	0.299	0.016	94.76:5.24
PA1039E	Ring brooch (pin)	61.8	33.9	3.0	0.116	0.044	96.28:3.72
PA1039F	Ring brooch (braided str.)	49.6	37.0	8.9	0.218	0.105	97.61:2.39
PA1059A	Torc (necklace)	52.5	33.9	13.5	0.157	nd	98.85:1.15
PA1069A	Ring brooch (bow)	69.2	26.4	4.0	0.239	nd	94.37:5.63
PA1069B	Ring brooch (knob)	78.4	17.8	3.4	0.073	nd	97.90:2.10
PA1069C	Ring brooch (coating wire)	75.9	14.9	8.9	0.053	0.008	99.41:0.59

Prākṛta language on the topic of the occurrence of noble metals in the base metals, such as tin and lead¹³. He has reported the presence of gold in tin, and of silver in lead. Ṭhakkura Pherū states that tin contains gold to the extent of thirty-four *jau* gold in one hundred *tolā* tin (Verse 26). The relevant verse in question is as follows:

*evaṁ ca bhāṇiya saṁpai kudhāumajjhe sudhāu
bhāṇimoham |
kaṇviya raṅge kaṇayam tolāya saya java cauttisam ||*

Ṭhakkura Pherū has described elsewhere the relationship between various weight measurements. In his book *Dravyaparīkṣā* (Verse 38), he has given the following relationship between various weight measurements prevalent in his time:

16 *jau* = 1 *māṣa*
4 *māṣa* = 1 *ṭaṅka*
3 *ṭaṅka* = 1 *tolā*

According to the above, there are 192 *jau* in 1 *tolā*. Thus

Table 4

Surface SEM-EDX analysis of some gold globules containing tin found at Sardis in elemental weight percentage¹⁷

Sample Description	Au	Ag	Cu	Fe	Sn	Pb	Mg	Al	Si	K	Ca	Mn	P	Remarks
<i>Sample 45673Z</i>														
Gold globule An1	84.2	3.6	-	-	8.6	-	-	-	-	-	-	-	-	
Gold globule An2	93.7	3.1	-	-	3.2	-	-	-	-	-	-	-	-	
Gold globule An3	77.6	3.1	-	-	19.3	-	-	-	-	-	-	-	-	
Gold globule surface An5	84.8	3.1	-	0.8	3.4	3.3	-	1.7	1.8	0.7	0.4	-	-	
<i>Sample 45675V</i>														
Gold globule An12	95.8	2.3	-	-	1.9	-	-	-	-	-	-	-	-	
Gold globule An13	95.4	2.6	0.6	-	1.4	-	-	-	-	-	-	-	-	
<i>Sample 45680Q</i>														
Gold globule An3	58.9	38.9	0.6	-	1.6	-	-	-	-	-	-	-	-	
Gold globule An5	58.4	38.9	1.1	-	1.6	-	-	-	-	-	-	-	-	
<i>Sample 45673Z</i>														
Gold outer surface layer An4	2.0	-	0.7	24.1	58.1	-	2.7	1.9	3.4	0.6	0.8	0.9	-	
Gold outer surface layer An6	6.1	-	1.1	24.1	58.1	1.8	5.8	4.5	8.7	1.2	2.9	2.0	0.6	Cl: 0.4, Ti: 0.4

according to Thakkura Pherū tin contained about 0.17wt% gold. From the description given by Thakkura Pherū, it is not clear whether such a tin was produced within India or was imported from some other country. A possible source of this type of tin is discussed later.

The above reference of Thakkura Pherū indirectly suggests that in all probability Indians were recovering gold from auriferous tin mini-ingots.

Discussion

In ancient times, the most important source of gold was alluvial placer deposit. This type of deposit was also common in medieval times. Alluvial placer gold is derived from the weathered out rocks containing vein gold deposits. Gold is highly resistant to weathering. The gold particles along with weathered rocks are washed down the mountains, and are subsequently deposited in the sand and gravel of rivers. Naturally occurring gold contains several impurities; the most notable ones have been silver and copper. During weathering and transport of gold particles, silver and copper present in gold particles are more susceptible to dissolution or leaching

depending on the pH value of the surrounding environment. Thus the overall silver and copper content of the alluvial placer gold is somewhat less than the vein gold from which it has been derived.

The silver content in natural gold may vary between 1-40 wt%^{14,15}. The maximum amount of copper in natural gold is much more difficult to define. Hartmann and Sangmeister¹⁶ stated that only 0.01 wt% copper is usually present in natural gold, and is reported never to exceed 1.5 wt%. Tylecote¹⁷ mentioned that copper content in natural gold is less than 1 wt%, but also stated that more than 3 wt% copper is almost certainly alloyed gold. It is reasonable to assume, as Pingel¹⁸ has done elsewhere, that the limit of copper in natural gold is around 2 wt%.

The principal ore of tin in ancient and medieval times was cassiterite. It is also very resistant to weathering, and is obtained as alluvial placer deposit. This type of cassiterite deposit was an important source of tin in ancient and medieval times.

Thus, the mining method and the mineral beneficiation technique used in the past for gold and cassiterite, have some similarities, as both were obtained as alluvial placer deposits. Both these minerals were beneficiated by simple

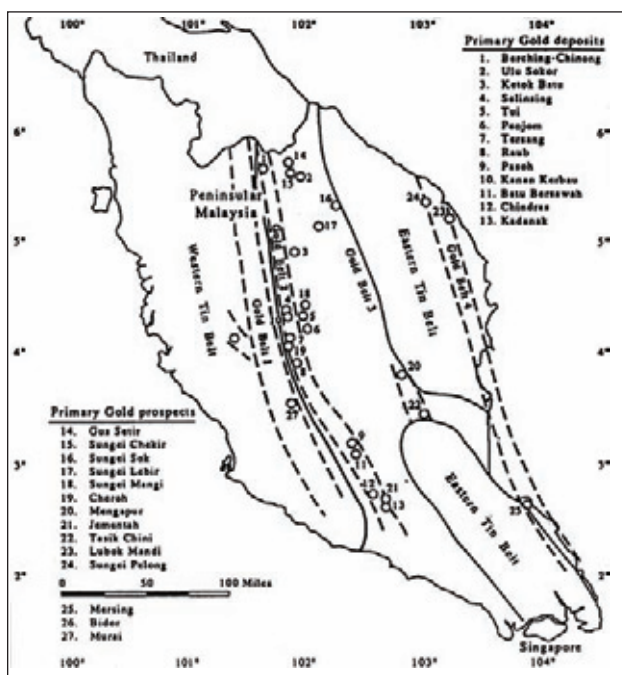


Figure 3
Peninsular Malaysia showing mineral belts and primary gold occurrences²⁶

process of panning, due to their high specific gravity, in the ancient and medieval period.

In order to understand the reason for the presence of gold and tin in each other, it is necessary to discuss the mechanism of the formation of alluvial placer deposit, which follows.

When a water stream containing mineral grains of different specific gravity and size flows, their simultaneous deposition can occur depending on fulfilling certain conditions. Rubey¹⁹ has introduced the concept of “hydraulic equivalence” for understanding the behaviour of simultaneous deposition of grains of heavier and lighter minerals. He stated that the grains of same settling velocity are deposited together. He assumed the spherical shape of grains. Rubey stated that the settling velocity of grains is a function of grain size and the specific gravity of grains and introduced the concept of “hydraulic equivalence size”. According to it, smaller size grains of heavier minerals deposit together with larger size grains of lighter minerals. Reid and Frostick²⁰ have mentioned the quartz settling equivalents of cassiterite and gold. A spherical cassiterite (specific gravity 7) grain has a diameter, which is only 0.52 of its quartz equivalent, while gold (specific gravity 19), due to its even higher specific gravity contrast has a diameter only 0.32 of quartz grain. Thus the mean grain size of the heavier mineral is finer than that of lighter mineral quartz. However, there are weaknesses inherent in the simple theory of settling equivalence as a dominant parameter in the formation of placer deposit. In the actual practice the diameter of heavier and lighter particles are different than those predicted by this theory. Factors such as particle shape, orientation of particle in the stream and particle surface texture affect the settling velocity of the particles. Many times the shape of the particle is flaky, which is far away from spherical. It has been shown

that major sorting and concentrating process take place on the river channel rather than higher up in the flow. Such entrainment equivalence and dispersive equivalence have been introduced to explain the actual settling behaviour of sediments. Another factor, which controls dispersion, is the trapping of small placer particles in bed interstices.

From the foregoing discussion, it is apparent that if the water stream contains gold, cassiterite and quartz particles, all the three types of particles would deposit together but have different mean sizes. Thus the size of gold flakes deposited with fine size cassiterite particle would be very fine, and their separation during panning of auriferous cassiterite would be rather difficult. Such an auriferous alluvial placer deposit can be concentrated into gold-rich or cassiterite-rich concentrate by panning. If it is desired to obtain a gold-rich concentrate, the complete separation of gold particles from cassiterite or quartz would lead to a greater loss of gold in the tailings. In general, the common practice was to pan gold concentrate along with some residual mineral impurities from the auriferous sand to decrease the gold loss in the tailings. When such a gold concentrate containing residual cassiterite and quartz impurities is melted in a crucible under oxidizing atmosphere, almost pure liquid gold would be obtained, and the residual oxide impurities such as cassiterite and silica would be removed as a slag. Raub²¹ prepared a gold leaf containing 1 wt% cassiterite concentrate by sintering a mixture of gold powder and cassiterite concentrate followed by mechanical deformation. It was melted four times in air in a quartz tube (each time solidifying), and was analysed. Cassiterite impurities disappeared completely from the gold as slag. Similar results were also observed for the gold foil prepared from alluvial placer gold containing non-metallic impurities from river bed.

Alternatively, if such a gold concentrate is melted under reducing atmosphere, cassiterite would get reduced depending on p_{CO}/p_{CO_2} ratio in the atmosphere, and would enter into liquid primary gold. At room temperature gold has a solid solubility of about 3-4 wt% tin. If the concentration of tin is more than this amount then inter-metallic compounds between gold and tin form. The residual silica and unreduced cassiterite are slagged off. It must be stated that if such a gold containing tin is further melted in oxidizing atmosphere, refining would take place and tin would be removed as oxide in the slag.

On the other hand, if such an auriferous alluvial placer deposit is panned for the recovery of cassiterite, some gold particles would be present in cassiterite concentrate. When such a cassiterite concentrate is charged in a tin smelting furnace, gold would get collected in liquid primary tin. It is interesting to note that the equilibrium solid solubility of gold in tin is small, about 0.2-0.3 wt% at room temperature.

A pertinent question that may be asked is as to how both cassiterite and gold particles occur together in stream. There could be two possibilities. In the first case separate streams containing cassiterite and gold derived from respective

weathered deposits, mixed together at a certain point to form a composite stream containing both cassiterite and gold. In the second case it is possible that the weathered mineral deposit contains both cassiterite and gold together and thus the stream carrying such a deposit would have both these minerals from the beginning.

There are several references, which state that gold and cassiterite can be present simultaneously in the deposit. Pliny was perhaps the first writer who pointed out the presence of cassiterite along with gold deposit. He writes²²:

“...The Greeks applied to it the name cassiteros... It (Cassiterios) is also found in the gold mines called “Alutiae” through which a stream of water is passed that washes out black pebbles of tin mottled with small white spots, and of the same weight as gold in the bowls in which it is collected, and afterwards are deposited in the furnace, and fused and melted in to white lead (tin)”.

There are some more references, which show that auriferous cassiterite was found in ancient times. The alluvial placer deposit obtained from the stream in the Taurus foothills near Nigde, Turkey contains cassiterite and gold in addition to other minerals. Old workings in the nearby mountains were also found. The radio-carbon dates suggest that tin was mined in Taurus Mountains in the Early Bronze Age²³. Earl and Özbal²⁴ have presented the analytical data for a large number of samples of ore, both unground and ground, collected from the old working sites of Göltepe and Kestel mines in the Central Taurus region of Turkey. It shows that the ore carried tin values of a very “patchy” nature, and is frequently found with gold and tin mineralization. Most of the mines consisted of limestone rock, with hematite mineralization. There has been an acid igneous quartzite intrusion in the lowest level.

Malaysia has been an important tin and gold producing country in ancient times. Both cassiterite and gold were largely mined as alluvial placer deposit. Peninsular Malaysia consists of three main geological domains, which are parallel to peninsula. Scrivenor²⁵ designated them as Western tin belt, Central gold belt, and Eastern tin belt, as shown in Figure 3. There is evidence that gold also occurs in the central parts of the tin-rich Western belt. On the basis of the style of mineralization, Yeap²⁶ proposed to divide the gold mineralization in Peninsular Malaysia into four belts or zones which are parallel, some juxtaposed and some overlapping with the Eastern and Western tin belts or zones. It is not unlikely that tin and gold could deposit together in alluvial placer deposit because of the proximity of tin and gold belts. In this context, the statement of Emmons²⁷, who was Professor and Head of Geology and Mineralogy at the University of Minnesota, U.S.A. in the early part of the twentieth century, is of importance:

“In west Malaya, however, the gravels of some of the tin placers carry considerable gold.”

Gerini²⁸ has discussed the old gold working sites in Malay

Peninsula. Many of the gold mines developed by European enterprises in the Malay Peninsula in the nineteenth century were originally worked at a very early age. The evidence of gold working were innumerable in the area, and were termed “wonderful” by the Europeans, who first noticed them. Gerini has opined that the Hindus (Indians) visiting /migrating to the Malay Peninsula planned, directed and superintended the gold mining activities at most of these sites in ancient times.

Al-Masudi (943A.D.) has mentioned the existence of tin, silver, gold and lead mines around the countries of Kalah and Panhang (Pahang) in the Malay Peninsula²⁹:

“...Mines of tin (al-rasas) and mountains of silver. There are also mine of gold and lead (al-rasas al-abyad)”.

In 1909, Lajonquiere³⁰ published a report on the archaeological work carried out in the Malay Peninsula, according to which a large number of Hindu colonies were situated in ancient times in widely remote centers, such as Chumphon, Caiya, the valley of river Bandon, Nakhon Sri Dhammarat (Ligor), Yale (near Patni) and Selensing (in Pahang) on the eastern coast, and Malacca, Province Wellesley, Takua Pa, and the common delta of river Lanya and Tenasserim on the western coast. Further, Lajonquiere states that the group of people occupying the colonies at Selensing, Panga, Puket and Takua Pa prospered by the exploitation of tin and gold mines. It is not unlikely that some of the alluvial placer cassiterite and gold concentrates from this area contained some amounts of gold and cassiterite respectively.

There are several reports, which state the association of cassiterite with gold deposit even in pre-modern and modern times. In the discussion that follows, a brief discussion of such deposits has been given below.

Gold dust and flakes and even small gold nuggets have been reported to be present in most of the alluvial tin deposits in Cornwall. In 1602 A.D., Richard Carew in his “Survey of Cornwall” has reported the occurrence of gold in alluvial placer tin deposits of Cornwall, and its subsequent separation³¹. The amount of gold thus recovered was small. Even after careful panning, some gold must have been left in the cassiterite concentrate, and some cassiterite in the gold concentrate. This would have resulted in tin and gold in each other after smelting. In this connection, it is interesting to note that a necklace made in 1802 A.D. from the gold recovered from the alluvial placer tin deposit of Ladock, is on display in the Royal Cornwall Museum. Most of the gold recovered was sand-grain size and rarely larger than ‘a split pea’. It is interesting to note that about 90% of the British prehistoric gold objects contain traces of tin measured in ppm. Wing-Easton³² has noted that for a long time several places along the eastern coast of Banca island in Indonesia have been known for the recovery of tin ore from Beach sand, occasionally with a little gold. In Lower Myanmar, various tributaries of the Tenasserim river, besides carrying cassiterite, were also auriferous³³. Cassiterite–Gold deposits have been available in the Tin Range, Port Pegasus,

Stewart Island, New Zealand. These deposits were worked during the period 1888–1914 for recovering gold and cassiterite^{34,35}. Morgan³⁶ identified cassiterite in the gold sluicing concentrate at Montgomery's and neighbouring place in the Blackball-Healy's Gully district, New Zealand. Morgan also noted stream tin in auriferous gravels near the coast line north of Ten Mile Creek. Stream tin was recovered from gold placer mining operation in the Hot Spring and Ruby districts in Alaska in the beginning of twentieth century³⁷. Hutton³⁸ recognized cassiterite in the dredge concentrate from the Barry town, Blackballo, Snowy river, Grey river, Ngahere, Atarau, Arahura, Rimu, Kaniere and Slab Creeks gold dredges in New Zealand. Patyk-Kara³⁹ has reported gold and tin-bearing placer deposits in the coastal area east of the Kolyma river mouth in the eastern arctic shelf of Siberia. Suthakorn⁴⁰ has discussed in detail the important types of tin deposits found in Thailand. The primary Pilok tin deposits in Thong Pha Phum district in Thailand contains cassiterite along with gold locally. Similarly, small amounts of gold is found locally in tin-bearing quartz veins in Ratchaburi, Suanphung district. Small amounts of gold have been reported to be present in the cassiterite bearing veins in Prachuap Khiri Khan Area. Suthakorn has reported that alluvial placer tin ore deposit in many localities in Thailand is associated with gold flakes. Such a type of auriferous tin ore deposit is found in Pilok area, Kanchanaburi province, Bang Saphan area, Prachaua Khiri Khan Province and Tomo area, Narathiwat province, etc. It is interesting to note that the word "Kāñcana" in Kanchanaburi means gold in Sanskrit language, which had a great influence in South East Asia in ancient and medieval times. Several alluvial placer deposits near Haystack Mountain, Alaska resulted in both gold and tin ore in the pan concentrate apart from other minerals⁴¹. Cabri et al⁴² have reported the presence of gold in cassiterite of Corvo ore body, in the Iberian Pyrite Belt of Portugal. Kovalenker⁴³ has stated that an upper palaeozoic gold deposit in the Chatkal-Kuraminisk ore region (middle Tien Shan) has revealed tin mineralization. The principal tin mineral includes cassiterite. Thomson et al⁴⁴ have discussed the characteristics of gold deposits in Tungsten-tin provinces in the northern Cordillera of the US, the Hereynides of Europe, the Altaid Orogen of eastern Australia and the polymetallic belt of south western Bolivia. Marcoux et al⁴⁵ have discussed Au-Ag-Sn-W(Bi) ore deposit of Cirotan, West-Java, Indonesia. Gavrilo and Onikhimovskiy⁴⁶ have stated that tin and gold occur in the same deposit due to similar chemical and siderophile properties. They have further stated that Ag and W act as geochemical "bridges" that link Au and Sn and promote their co-precipitation in ores.

For the sake of the present discussion, it is reasonable to divide the Au-Ag-Cu alloys mentioned in Table 1-3 in two groups, viz. those obtained from natural gold, and those made by alloying. From the point of view of silver content, all the gold samples in Table 1-3 can be considered as natural gold. However, from the point of view of copper content, it is not the case. As discussed earlier, gold alloys containing less than 2 wt% copper could be considered as "natural gold", while those containing above this amount as "alloyed

gold". It can be seen that the tin content in gold alloys containing less than 2 wt% copper varies from 0 wt% to 0.43 wt%. Similarly, in gold alloys containing more than 2 wt% copper, the tin content is between 0-0.3 wt%. In case of natural gold objects containing tin, the source of tin is the cassiterite left out in the gold-rich concentrate obtained after panning. On the other hand, there could be another source of tin as well, in addition to the above described one, for the alloyed gold objects. This would depend upon as to how copper was added during the preparation of gold alloy. Copper could have been added either as copper, copper-arsenic alloy or tin bronze. If the copper content was increased by adding copper or copper-arsenic alloy, all the tin in the alloyed gold must have been derived from the left out cassiterite, as discussed earlier. However, it is also possible that the copper addition was done in the form of tin bronze. In this situation, a part of the tin present in Au-Ag-Cu alloys would have been derived from the tin bronze. If it is known as to how much tin and copper come from the addition of tin bronze, one can estimate the composition and amount of tin bronze used for making a particular alloyed gold containing tin. However, it is very difficult to fix this value. We have to content ourselves by speculating about the proportion of tin and copper coming out from tin bronze addition. If it is assumed that in case of alloyed gold containing more than 2 wt% copper, all the tin and copper are derived from tin bronze, one can estimate the composition of tin bronze used for alloying. Such a relationship has been shown in Figure 4. It can be seen that such bronzes used for alloying contain tin in the range 0.5-7 wt%, which is within the limit of the composition of ancient tin bronzes.

It is difficult to identify the origin of the auriferous tin ingots as reported by Thakkura Pherū, and of the staniferous gold as found from different places. However, an attempt has been made to throw some light on this subject.

Hartmann and Sangmeister⁴⁷ have discussed the source of the staniferous gold used in the European gold objects dating from the Bronze Age to the beginning of the Christian era, found from various parts of Europe. They have cautiously expressed the tentative view that the staniferous gold originated somewhere in east Europe and was traded by a maritime route via the Baltic, the North Sea, and the Atlantic to various parts of Europe. Muhly⁴⁸ also thinks that the chemical analysis of the ancient European gold artifacts indicates a possible Bronze Age trade in gold, and a connection between sources of gold and sources of tin.

The contact between India and various countries of Southeast Asia dates back to pre-historic times. Southeast Asia was described in Sanskrit literature by generic terms- *Suvarṇabhūmi* (The Land of Gold) and *Suvarṇadvīpa* (The Island of Gold). The contact between India and Southeast Asia became vigorous at the time of the beginning of the Christian era, and strengthened continuously in the subsequent centuries. There was intense commercial and trade activities between India and Southeast Asia in the

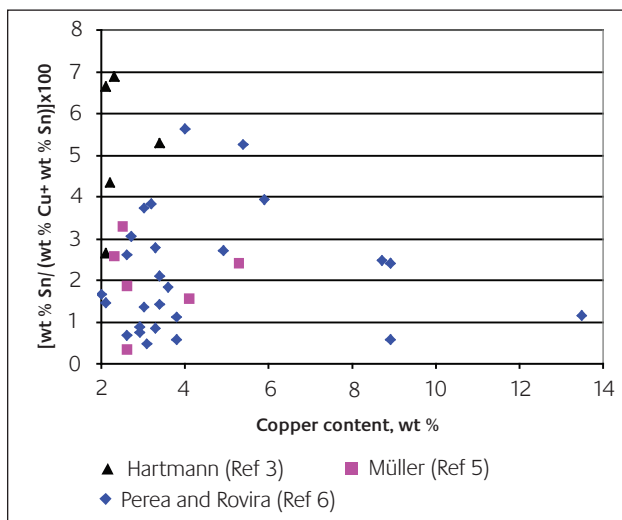


Figure 4
Relation between copper content and $[\text{wt} \% \text{ Sn} / (\text{wt} \% \text{ Cu} + \text{wt} \% \text{ Sn})] \times 100$ of Au-Ag-Cu alloys containing tin

medieval period. Gold and tin were the two most important metal commodities besides other products such as spices, aromatic woods, fragrant resins, silk and precious stones, which were imported to India from Southeast Asian countries. In view of the occurrence of the gold and tin mineralization together in some parts of Southeast Asia, as discussed earlier, and the commercial contact between India and Southeast Asia, it seems probable that some of the tin ingots imported from Southeast Asia to India were auriferous in nature. However, some sources other than Southeast Asia for auriferous tin mini-ingots cannot be ruled out.

Concluding Remarks

There has been some archaeological evidence which reveal that historical gold objects, such as jewellery, bowl, beaker etc., obtained from different parts of Europe, contained small amounts of tin. The tin content varies from as small as 0.002 wt% to about 0.4 wt%. It is interesting to note that these objects were essentially made from ternary Au-Ag-Cu alloys. The tin content in some of the gold pieces obtained from the archaeological excavation carried out at Sardis, is as high as 1-8 wt%. All the references of gold objects containing tin discussed in the present paper are from Europe. It would be interesting to study the gold objects of other parts of the world from this point of view.

There is an Indian description of early fourteenth century A.D. written by Ṭhakkura Pherū, which states that tin mini-ingots contained about 0.17 wt% gold. This reference is unique, and has not yet attracted the attention of the historians of metallurgy. It seems that in medieval times India was getting the supply of tin mini-ingots from the area where auriferous cassiterite was available. Some selective parts of Southeast Asia could have been the probable location for it. However, some sources other than Southeast Asia for auriferous tin mini-ingots cannot be ruled out.

Cassiterite and gold can sometimes occur simultaneously in the same vein or alluvial placer deposit. Ground vein or particulate alluvial placer auriferous cassiterite deposit could be concentrated to either a cassiterite-rich concentrate or gold-rich concentrate by panning process. Despite all the possible precautions taken during panning, the cassiterite-rich concentrate would have contained some gold particles, and the gold-rich concentrate some cassiterite particles in historical times. When such a cassiterite-rich concentrate is smelted at high temperatures, the residual gold would get collected in the liquid primary tin. On the other hand, when such a naturally occurring gold-rich concentrate is melted in somewhat reducing atmosphere, the cassiterite is reduced to metallic tin, which enters into liquid primary gold phase. It is generally believed that historical Au-Ag-Cu alloys containing more than 2 wt% copper were not just derived from naturally occurring placer gold concentrate, but were made by alloying it. It is quite possible that tin bronze, in lieu of copper or copper-arsenic alloy, was also used for the addition of copper in such gold alloys. In such cases, at least a part of tin might have been derived from tin bronze addition.

Thus the occurrence of tin in some historical gold objects, and gold in some tin mini-ingots, as discussed in the present paper, does not seem to be the result of intentional alloying practice, but is due to the extraction of gold from natural stanniferous alluvial placer gold concentrate, and tin from natural auriferous cassiterite concentrate respectively.

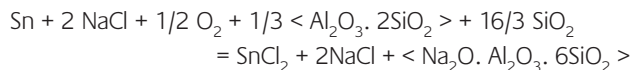
When gold containing tin is repeatedly melted as a result of recycling, the residual tin would be oxidized and transfers in to slag. Thus the amount of tin in gold would decrease after repeated melting. On the other hand when tin containing gold is used as an alloying element in making alloys such as bronzes, the gold being a noble metal would still remain in the final product. It would be interesting to carry out a more systematic chemical analysis of historical tin bronzes from this point of view.

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Appendix 1

The removal of tin from impure gold during salt cementation process, i.e. heating of impure gold along with a mixture of salt and brick dust powder, is believed to occur according to the following reaction:



The standard free energy change and equilibrium constant value of the above reaction at 925 K are approximately – 47 kcal and 1.25×10^{11} respectively.

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