Solid state diffusion bonding of gold: a Sanskrit reference from Vedic literature

R K Dube

ABSTRACT: A Sanskrit reference from the Chāndogya Upaniṣad in the form of a simile, dealing with joining of gold pieces by solid state diffusion bonding without the use of any external filler metal, is discussed. Both unrefined native gold and alloyed gold were used in antiquity. The presence of a base metal oxide film is detrimental to good quality diffusion bonding. The ancient Indian process used consisted of cleaning the surfaces to be joined, followed by heating them together in close contact. The generic term lavaṇa was used for substances used for cleaning the gold surfaces. Alternatively, lavaṇa substances were used to create a thin layer of pure gold on the surface by either depletion gilding or a cementation process. Such lavaṇa-treated gold pieces kept in close contact were heated leading to diffusion bonding. Availability of lavaṇa substances in the Sindhu (Indus) valley and adjoining areas is also discussed.

Introduction

Gold is a metal par excellence. One characteristic is that pure gold pieces can be joined by solid-state diffusion bonding, without requiring any external filler metal. This is made possible due to the absence of an oxide film on pure gold when heated in air, its low elastic modulus, together with its rapid self-diffusion characteristic. A good quality joint between pure gold pieces can be achieved at temperatures above about 430°C, even without applying pressure. Humpston and Baker (1998) reported that a bundle of gold wires converted into a single rod after heating at 500°C for one hour. Pure gold rods placed in contact with each other, joined readily at 500°C (Humpston and Jacobson 2004, 43).

Solid state diffusion bonding of gold is an age old process. However, the published information on this subject is far-fetched and sketchy (Tylecote 1967). The present paper discusses a Sanskrit reference belonging to Vedic literature, which refers to the joining of gold by solid state diffusion bonding without any external filler metal.

Documentary evidence

In the Chāndogya Upaniṣad (4.17.7-8) there is the following reference regarding how one makes good the defect in the *yajña* ceremony performed by the priest:

> *tadyathā lavaṇen suvarṇaṁ saṅdadhyātsuvarṇen rajataṁ rajatena trapu trapuṇā sīsaṁ sīsena lohaṁ lohen dāru carmaṇā* ||7||
> *evameṣāṁ lokānāmāsāṁ devatānāmasyāstrayyā vidyāyā vīryeṇa yajñysya viriṣṭaṁ saṅdadhāti bheṣajakṛto ha vā eṣa yajñyo yatraivaṅvidbrahmā bhavati* ||8|| (Śaṅkara 1998, 438)

Jha (1942, 218) provided an English translation of the above passages:

> Just as one would join gold by salt, silver by gold, tin by silver, lead by tin, iron by lead, wood by iron, and wood by leather [7]. So does one make up the defect in the Sacrifice (*ie yajña* religious ceremony) by the virility of these regions, of these divisions and of the science of the three Vedas. That Sacrifice verily becomes well-healed up where there is a Brahman-Priest knowing this [8].

Upaniṣads are an important part of the vast Vedic Sanskrit literature. Winternitz (1972, 236 and 310) opined that the beginning of the Vedic period lies between 2500–2000 BC, while the end of the period is between 750–500BC. Further, he stated that the Chāndogya Upaniṣad is one of the earliest Upaniṣads. The date of its composition can thus be assigned to approximately1000BC. On the basis of an astronomical statement cited in one of the Upaniṣads, Vaidya (1930, 173) concluded that the Upaniṣadic age can be placed between 2500–2000BC.

The composer of the above passages explained the process of making up the defects in the religious ceremony (*yajña*), through a simile which deals with the joining of materials, including different metals and wood. The joining of metals, such as silver, tin, lead, and iron by brazing/soldering is referred to. Filler metals are stated for each case. However, in the case of joining of gold, there is no mention of any metallic filler material, but instead *lavaṇa* is mentioned. In modern times, the most common meaning of *lavaṇa* is salt, *ie* common salt, and accordingly *lavaṇa* was translated as salt above. In fact, the appropriate translation of *lavaṇa* above was not considered, leading to confusion in understanding the cited reference.

Solid state diffusion bonding of gold with the aid of *lavaṇa*

Since there is no mention of any metallic filler material for the joining of gold, it is logical to believe that the reference in question deals with the solid state joining of gold without any external filler metal. In other words, it is an example of the solid state diffusion bonding of gold pieces, with the aid of *lavaṇa*. Questions which arise are: What is the actual meaning of the term *lavaṇa* in the present context? and, What is its function in the joining process?

In general, pure gold was not used in ancient times or even in modern times, for most common applications, due to its poor hardness. In order to increase its hardness, gold was used in alloy form. The most common alloying elements were silver and copper. There could also have been some other base metals present in the gold. In ancient times, it is reasonable to assume that unrefined native gold was also used in large amounts for making different types of products. In general, native gold contained some silver, copper and iron, together with traces to minor amounts of several other elements (Boyle 1979, 17). On the basis of a survey of the gold deposit literature, Boyle (1979, 335) stated that the purity of most placer deposits is in the fineness range of 500-999.

When gold containing base metals is heated in air, a coating of their oxides is formed on its surface. If this is allowed to remain, it prevents proper metal-to-metal contact during solid state joining. Hence, it is essential to remove the oxide film from the impure gold pieces to get a good quality joint. The substance referred to as *lavaṇa* was used as a flux, which helped in providing proper metal-to-metal contact, leading to a sound and firm joint between alloyed gold pieces. The question is what material was used as *lavaṇa* in the present context.

The meaning of *lavaṇa*

Today, the word *lavaṇa* is widely used for salt, and in particular for common salt (NaCl). Monier-Williams (1990, 898) has given the meaning of the word *lavaṇa* as salt, especially sea salt, rock or fossil salt, factitious salt or salt obtained from saline earth. Before we discuss the meaning of *lavaṇa* in the present context, it is desirable to trace the history of this term in India. *Lavaṇa* is quite an old term, as it is referred to in the Atharvaveda (7.76.1) (Satavalekar 1985, 92). One important Sanskrit book on Indian medicine, Suśruta Saṁhitā, dealt with the terminology *lavaṇa* in detail. In it (Sūtrasthānam, 42.26), a number of substances were included under the category of *lavaṇa*:

> *saindhavasauvarcalaviḍapākyaromaka-sāmudrakapaktrimayavakṣāroṣaraprasūta-suvarcikāprabhṛtīni samāsena lavaṇo vargaḥ* ||26||
>
> 'Substances such as *saindhava*, *sauvaracala*, *viḍa*, *pākya*, *romaka*, *sāmudraka*, *paktrim*, *yavakṣāra, ūṣaraprasūta*, and *suvarcikā* belong to the category of *lavaṇa*' (Ghanekar 1977, 232).

Here *Saindhava* is rock-salt from Sindh province of the erstwhile united India. *Sauvarcala* is sochal salt which is prepared by boiling down soda with emblic myrobolan [Indian gooseberry, *Phyllanthus emblica*]; it is also a type of natron. *Viḍa* means black salt. *Pākya* is saltpetre. *Romaka* is a kind of salt extracted from saline earth called *rumā*. According to some, it is the salt extracted from the lake Sambhar in Ajmer, India. *Sāmudraka* is the salt produced from sea water. *Paktrima*, which is also known as *pākima*, is the substance obtained by cooking or boiling; it refers to saltpetre. In Sanskrit, alkali was termed as *kṣāra*. *Yavakṣāra* is an alkali produced from the ashes of burnt barley-corns. *Ūṣaraprasūta* refers to the substance obtained from saline soil. *Suvarcikā*, also known as *svarjikā kṣāra*, is either natron or saltpetre (Monier-Williams 1990; Apte 2014).

In a different reference (46.322) (Ghanekar 1977, 293):

> *yavakṣārasvarjikākṣāroṣakṣārapākimaṭaṅkaṇa-kṣāraprabhṛtayaḥ* ||322||

the Suśruta Saṁhitā also included *ṭaṅkaṇa* (borax) in

the list of *lavaṇa*. The composer of the Suśruta Saṁhitā mixed up substances belonging to both salts and alkalis, as they are known in modern times, in the category of *lavaṇa*. Clearly, he used the word *lavaṇa* in a generic sense, which included both salt and alkali groups.

Similarly to Suśruta Saṁhitā, Kauṭilya also used the term *lavaṇa* generically. In his list under the *lavaṇa* category, Kauṭilya enumerated six substances: *saindhava*, *sāmudra*, *viḍa*, *yavakṣāra*, *sauvarcala*, and *udbhedaja* (Gairola 1962, 194). This list also included both salts and alkalis. In the Rasaratnasamuccaya (10.68), *ṭaṅkaṇa* is also included in the list of alkaline substances, together with *yavakṣāra* and *svarjikākṣāra* (Sharma 1977, 154).

Caraka Saṁhitā, a Sanskrit work composed in the 2nd century BC, prescribed alum as an ingredient for some medicinal preparations (Dube 1993; Sastri and Chaturvedi 1984, 852 and 858). The words used for it were *saurāṣṭrikā* (*ie* produce of the Saurāṣṭra area, part of Gujarat state, India), and *kāṅkṣī* (produced in the Kaccha area in Gujarat state, India). It is reasonable to assume that the alum was known in the period of Upaniṣadic literature. It is interesting to note that the red dyed cotton fragments found at Mohenjo-daro suggests the use of alum as a mordant in ancient India (McIntosh 2008, 334). Further, rock salt and alum are often found in the same crystalline form in nature and have nearly the same hardness and specific gravity (Melloni 1835, 232). There is a fair chance that alum was also considered as a substance under the category of *lavaṇa*. It is interesting to note that the term‘alum salt’ was in use along with nitre salt, borax salt, vitriol salt, etc, even in pre-modern times (Lieber 1835, 509), and that the compound Sanskrit word *arkalavaṇa* has been used for saltpetre (Monier-Williams 1990, 89).

In ancient times most of the substances belonging to the category of *lavaṇa* were not pure in the strict sense but contained some other related materials. Notes of the impurities present in some of the substances referred to in the context of *lavaṇa* are available in pre-modern literature. For example, naturally occurring borax from the Ladakh area in India contained sodium chloride as an impurity (Cunningham 1854, 239), and saltpetre also usually contained sodium chloride as an impurity. Baden-Powell (1868, 81-2) stated the chemical analysis of a number of saltpetre samples (produced in the Shahpur district of Multan in the former united India) contained sodium chloride from a very low amount to as high as 60 wt%, depending on the grade. Clearly, many naturally occurring substances of the *lavaṇa* category contained common salt (NaCl) as an impurity.

It is interesting to note that there were two Sanskrit terms in use, *lavaṇakṣāra* and *kṣāralavaṇa* , meaning respectively ‘alkali containing salt’ and ‘salt containing alkali’ (Mishra 2008, 686).

From the above discussion, it is clear that the term *lavaṇa* does not mean simply salt or common salt. Further, it might have consisted of more than one saline material**.** In the context of the cited reference in the Chāndogya Upaniṣad, it has been used in a generic sense. A variety of chemical substances, such as common salt, borax, saltpetre, alum, *yavakṣāra*, *sarjikākṣāra*, etc were included in the category of *lavaṇa*.

Discussion

Vedic references were composed in Sūtra (concise) style, wherein details were not provided. However, an attempt has been made to reconstruct the procedure adopted for joining gold according to the reference under consideration. The process of solid state diffusion bonding of gold consisted of two steps. The first and most important step was the cleaning of the gold surfaces, followed by the heating of gold pieces, kept in close contact with each other, without using any external filler metal.

As stated earlier, both unrefined native gold and alloyed gold contained base metals. As a result, a film of base metal oxides is formed on such gold pieces during heating in air. This film is detrimental in making a good join between gold pieces by diffusion bonding in the solid state. Borax is a unique substance, in the sense that it is capable of dissolving most metallic oxides. It also forms fusible products with silica. If a small quantity of borax is sprinkled on the surface of the heated gold containing base metals, the oxides present on the surface are dissolved in it, resulting in an oxide-free gold surface. In this way, a strong joint is formed by diffusion bonding between the gold pieces which are otherwise impure, in the sense that they contained base metals as impurities. It is interesting to note that the greatest commentator Śankarācārya, born in the later part of 7th century AD, has taken borax as the meaning of the word *lavaṇa* in his commentary on the passages of the Chāndogya Upaniṣad under discussion here (Upadhyaya 1963, 49; Śaṅkara 1998, 438).

Some other substances in the category of *lavaṇa* would also ensure a firm joint between impure gold pieces by diffusion bonding. However, the process route for achieving this goal differs from when borax is used for this purpose.

In antiquity, two processes related to gold metallurgy were developed: enhancement of gold content on the surface of the impure gold (*ie* depletion gilding), and through-and-through solid state refining of thin pieces of impure gold (*ie* a cementation process). These two processes were closely related to each other in the sense that they were based on the *lavaṇa* type of substances described earlier. In the present context it is logical to discuss these two processes in order to show how they were used for joining gold. Before doing that, it is worthwhile to introduce an important Sanskrit word –*saindhavikā* – used by Kauṭilya in his discussion on gold refining in his famous treatise Arthaśāstra (Gairola 1962). This word is derived from the word Sindhu (Indus), and means 'Indus earth'. The Indus valley and its adjoining area have been credited with the origin of Vedic culture and civilization. Sindh province was an important source of natural saline materials. In the context of the Sindh province, the remarks of Stocks, as quoted by Hughes (1976, 6), are worth mentioning:

> 'The alluvial tracts (in Sindh) nevertheless contain much saltpetre, and in South Sindh, where the soil is largely mixed up with sand, it is so impregnated with common salt as to produce by evaporation after simply pouring water over it, an abundant supply of that article.'

Clearly, the 'Indus earth' could have been either common salt-rich or saltpetre-rich earth from Sindh province, depending on the location of its source.

Diffusion bonding of depletion gilded gold

The scientific examination of archaeological gold specimens recovered from different places, such as gold rings from Nahal Qanah Cave and gold chisels from Royal cemetery at Ur, suggest that surface enhancement of impure gold specimens was practiced in antiquity (Craddock 2000, 27-8).

The Leyden Papyrus X, written in Greek capital letters similar to those used during the 3rd century AD, has given a recipe for 'gold polish'. Its English translation is as follows:

> 'For treating gold, otherwise called, purifying gold and rendering it brilliant: Misy, 4 parts; alum, 4 parts; salt, 4 parts. Pulverise with water. And having coated the gold (with it), place it in an earthenware vessel deposited in a furnace and luted with clay, (and heat) until the above-named substances have become molten, then withdraw it and scour carefully.' (Caley 1926, 1155).

A literary reference for the use of 'Indian salt' for the surface enhancement of impure gold dinars is available in a medieval text by Mansur ibn-Bara (13th century AD):

> 'Sixth chapter on the polishing of gold before stamping. After the gold flans are made round, they are placed in a pot and heated to redness. Pulverised Indian salt in a little fresh water is added to it. Then a fire is kindled under it with such intensity that the salt dissolves as lead does. It [the molten salt] is decanted into a mould; then the dinars are removed to be washed with cold water, then fine sand. They are dried in a pot above a gentle fire and then stamped. When the strength is contaminated with this salt, it clarifies it, with the help of the exalted Allah.' (Levey 1971, 35 and 66, cited by Craddock 2000, 29)

Craddock (2000, 29) identified the Indian salt with the Indus earth of the Arthaśāstra, and stated that it was likely to have been saltpetre. It is interesting to note Barbosa also stated that the salt derived from the blocks of the rock-salt hills in Hurmuz was called Indian salt (Dames 1918, 91). It is equally possible that the 'pulverised Indian salt' of Mansur ibn-Bara was pulverised rock-salt.

Depletion gilding of gold alloys, in particular of tumbaga alloys, was much practiced in Central and South America. Spanish priest Fray Bernardino de Sahagún (*c*1499AD) described the depletion gilding of the cast gold alloy necklace, which consisted of treating it with pulverised alum followed by washing and rubbing. The process was repeated. Final rubbing was carried out with a mixture of 'yellow earth' and a little salt, termed 'gold medicine'. The gilded gold exhibited a 'very yellow' colour. The details of the process and some of the ingredients used were kept secret (Dibble and Anderson 1959, 75).

During the pre-modern period, several references dealing with the surface enhancement of gold are available. The process, known as 'colouring of gold', consisted of heating the impure gold pieces in a mixture of chemical substances, such as salt, saltpetre and alum. The impurities, such as copper, silver etc, present near the surface of the gold pieces dissolved in the surrounding mixture, leaving behind a thin layer of pure gold near the surface. It is interesting to note that some substances in the category of *lavaṇa* were used for colouring gold even in the pre-modern times. A typical composition of the mixture employed for the colouring of gold was: 1 part of salt (*ie* common salt), 1 part of alum, and 2 parts of saltpetre (Gee 1913, 149). The process consisted of grinding the mixture to powder form, and heating it in an earthen pot for their dissolution. The mixture was

well stirred during heating. The work was suspended in it and kept under continual motion. Subsequently, it was taken out and cleaned to remove the adhering mixture. The process of dipping the work piece in the heated mixture and taking it out was repeated two or three times. In this way, a shining layer of pure gold was formed on the surface of the work piece.

A late 19th-century description shows that a mixture of iron sulphate, copper sulphate, potassium nitrate and sodium chloride was used for the surface enhancement of impure gold coin blanks in Japan (Gowland 1896, 409).

Sanskrit and other references from India

In India, there was a tradition of subjecting gold alloys to the *varṇapuṭa* treatment for improving its *varṇa* (colour). Clearly, it refers to the enhancement of gold content on the surface of impure gold. *Puṭa* is a common Sanskrit term in the preparation of a variety of Indian medicines. Monier-Williams (1990, 31) has given one of the meanings of *puṭa* as 'two vessels joined together for the formation of medicinal substances'. It is interesting to note that the method of preparing Indian drugs, in which the various ingredients were heated in a fire in this way, was termed as *puṭapāka*.

Rasahṛdayatantra, a Sanskrit text dealing with Indian medicinal preparations and composed by Govinda Bhagvatpāda, has discussed a process for the enhancement of gold content on the surface of impure gold based on the *lavaṇa* type of substances, as discussed earlier (Rasaśāstri 1989, 199).This reference to the 'colouring' of gold has not yet received the attention of scholars, and is being discussed here for the first time. Govinda Bhagvatpāda is considered to be the teacher of the great Indian philosopher Śankarācārya around the later part of the 7th century AD (Upadhyaya 1963, 140 and 281). However, no work of Govinda Bhagvatpāda on the subject of Indian Philosophy has yet been found. A few stanzas from Rasahṛdayatantra have been cited by Mādhavācārya (*c*1296 AD) in his popular treatise Sarvadarśana Saṁgraha. He also addressed Govinda Bhagvatpāda as '*prācīna* ācārya', *ie* ancient scholar (9.17) (Sharma 1964, 378 and 382). Thus, the text was certainly composed at least a few centuries before the 13th century AD. Sharma (1975, 458) stated the date of composition of Rasahṛdayatantra was the 10th century AD so it is not unreasonable to consider 9-10th century AD as the upper limit for the date of its composition.

Caturbhuja Mishra (2008) provided a Sanskrit commentary on this text. Rasahṛdayatantra described the various ingredients used in the *varṇapuṭa* (improvement in the colour of gold) treatment of the impure gold (18.11):

kṣitikhagapaṭuraktamṛdā varṇapuṭoyaṁ tato deyaḥ

In his commentary, Caturbhuja Mishra (2008) translated the words *kṣiti* as alum, *khaga* as yellow iron sulphate (ferric sulphate), *paṭu* as rock salt and *raktamṛdā* as *gairika* (iron oxide). According to the above verse, the process for the creation of a thin layer of pure gold on impure gold pieces consisted of applying a paste prepared from the fine size powders of alum, ferric sulphate, rock salt and iron oxide on the surface of the impure gold piece, followed by heating it in a closed earthenware vessel. The ratio of these ingredients in the mixture was not given.

Viccajee (1908, 24-5) described the surface enhancement of gold coin blanks as prevalent in Hyderabad, India in early 20th century. The mixture used for it consisted of common salt (2 parts), alum (1 part) and nitre (1 part). Some native jewellers of Hyderabad were using a nitre-rich mixture, consisting of nitre (2 parts), alum (1 part) and common salt (1 part).

The process route for the solid state diffusion bonding of impure gold via creating a thin layer of pure gold on the surface, would have consisted of heating the impure gold pieces along with different types of *lavaṇa* materials, as discussed above, followed by heating the depletion gilded pieces kept in direct contact with each other.

Diffusion bonding of gold that is surface cleaned by cementation

Lavaṇa type of substances, such as common salt, alum and saltpetre, can also be used in a different way for treating impure gold prior to joining. The process consisted of heating thin sheets of impure gold surrounded by a cement mixture in an earthenware vessel in the solid state. The Cement is a mixture of an active agent and a carrier. The most common active agent used in antiquity was common salt. However, alum, saltpetre and ferric sulphate were also used, either alone or in combination. Finely ground brick dust or clay was the most common carrier. Pure gold was obtained after heating the vessel for a prolonged period. The impurities, such as silver and copper, present in impure gold reacted with common salt and formed their respective chlorides, which were absorbed by the cement and wall of the pot. After cooling of the pot, pure gold was taken out. This solid state gold refining process is known as cementation. If the objective was the surface cleaning of impure gold pieces, the pieces would have been heated only for a shorter period of time.

Micro-porosities formed on the surface layer during refining could have been removed by burnishing. Thus the process for solid state bonding of gold via this route consisted of surface cleaning of impure gold pieces by the cementation process, burnishing of the refined gold surfaces, followed by heating the pieces kept together in direct contact in the solid state.

Although gold refining in solid state (cementation) has a very old history, most of the early literary references do not provide detailed descriptions. An exception is the Sanskrit reference given in Arthaśāstra, which is discussed below. The method described by Theophilus (*c*12th century AD) was based on the cement mixture containing common salt (2 parts) and tile or burnt reddened furnace-clay (1 part), lightly sprinkled with urine (Hawthorne and Smith 1963, 109). A 12th century AD manuscript of the *Mappae Clavicula*, which was believed to be composed in the early 9th century AD, described the gold refining process in which impure gold sheets were heated in a bed of mixture containing cow dung ashes and common salt (Smith and Hawthorne 1974, 65). The *Book of Minerals* compiled by Albertus Magnus in the 13th century AD stated that a mixture of 'powder made of soot, salt, and finely ground brick'was the cement material for gold refining (Wyckoff 1967, 230).

Agricola (who died in 1555 AD) presented several cement compositions in his well-known treatise *De Re Metallica*. The cement mixture formulations given by him can be divided into two broad groups. The first group consisted of mixtures prepared from brick dust, refined/rock salt, saltpetre, together with ingredients such as sal-ammoniac and/or vitriol, while the mixtures belonging to second group contained brick dust, refined/rock salt and vitriol. It is interesting to note that most of formulations presented by Agricola belonged to the first group, *ie* formulations containing saltpetre (Hoover and Hoover 1950, 453-4). Ercker presented at least three formulations for the cement mixture used for gold refining in his book *Treatise on Ores and Assaying*, first published in 1574 AD. His typical cement formulation consisted of brick dust, salt and white vitriol in the ratio 4:2:1 by weight, and moistened with urine or sharp wine vinegar. It is important to note that the cement capable of producing beautiful colour in gold, contained saltpetre. Its composition was brick dust 14 lot, haematite 4 lot, *crocus martis* 1 lot, verdigris 1 lot, white vitriol 6 lot and saltpetre 3 lot (Sisco and Smith 1951, 184). *Pirotechnia*, another work of the 16th century AD, described the cementation process for gold refining based on the cement mixture consisting of fine powders of old tiles or brick dust, common salt and vitriol in the ratio of 24:8:3 by weight (Smith and Gnudi 1943, 203-4). Another work of the same period, the *Probierbüchlein*, also prescribed a similar cement mixture, containing brick dust powder (2 parts) and salt (1 part), moistened with vinegar or children's urine (Sisco and Smith 1949, 144 and 151).

The Chinese text *Huang Ti Chiu Ting Shen Tan Ching Chüeh* (Explanation of the yellow Emperor's canon of the nine-vessel spiritual elixir) described a three step process for the refining of crude gold. The last step consisted of heating gold leaves along with a mixture of yellow alum, poplar balsam and mud. This work is considered to be of the 2nd century AD, but it could have been of the 4th or 5th century AD (Needham and Gwei-Djen 1997, 57). Yellow alum was possibly the hydrous sulphate of iron and aluminium called halotrichite, perhaps mixed with alunogen (Schafer 1963, 217). In Nepal, the cement mixture also contained borax (1/8 part) in addition to brick dust (2 parts) and common salt (1 part) (Campbell 1834).

Sanskrit and other references from India

Several Sanskrit texts, belonging to ancient to pre-modern periods, mention the cementation process for the refining of gold. Although there have been indirect references to gold refining in Vedic literature, Kauṭilya mentioned very clearly, for the first time, the cementation process for gold refining in his well-known treatise, Arthaśāstra. A number of scholars have put forward detailed arguments in favour of the date of Kauṭilya's Arthaśāstra as fourth century BC (Law 1914, x; Jacobi 1918, 102; Smith 1924, 45 and 160; Kane 1926; Bhandarkar 1926; Jayaswal 1943, 378-96). In his description for the preparation of standard gold for the making of gold products of different colours, Kauṭilya (Adhikaraṇa 2, Prakaraṇa 31) described a two-stage process for the purification of impure gold, as follows:

> *samasīsātikrāntaṁ pākapatrapakvaṁ saindhavikayojjvālitaṁ nīlapītaśvetaharita-śukapotavarṇānāṁ prakṛtirbhavati* |
>
> '(The impure gold) heated with equal amounts of lead (in liquid state), followed by transforming (the solidified gold obtained from the previous treatment) into leaves like thin pieces by fire (*ie* by hot/warm working), and lighted up with Indus earth, becomes the standard base (for the making of gold products) of blue, yellow, white, green and parrot-feather colours' (Shastri 1990, 216).

Clearly, the heating of impure gold in the liquid state along with lead refers to the gold refining by cupellation, wherein the base metal impurities present in the gold

were removed. The heating of thin strips of gold, which had previously been refined by cupellation, along with Indus earth refers to the cementation process of gold refining, wherein the silver present in the gold is removed in the solid state. Kauṭilya used the Sanskrit term *saindhavikā*, meaning Indus (Sindhu) earth containing common salt, which acted as the cement material used for gold refining. However, varying amounts of saltpetre could have also been present in it, depending on the location of its source, as discussed below.

Rasārṇava, an important treatise on the Rasaśāstra branch of Āyurveda discipline, is one of the earliest works of its kind. Ray (1903, lxxxiii) stated that Rasārṇava was composed in the 12th century AD. Rasārṇava (7.102) described the cementation process:

> *mṛttikā mātulungāmlaiḥ pancavāsarabhāvitā* ।
> *sabhasmalavaṇā hema śodhayet puṭapākataḥ* ॥
> 'Impure gold is soaked in a mixture of earth and citron juice for five days, followed by heating the mixture together with cow dung or wood ash, and salt in a *puṭapāka* vessel. As a result, gold gets refined.' (Ray and Kaviratna 1985, 116).

As discussed above, *Puṭapāka* refers to the heating of the mixture in a sealed vessel assembly prepared from an earthenware dish covered with a similar type of dish upside down.

There are some other references, which describe the use of saltpetre-based cement for solid state gold refining in India. Āīn-i Akbarī prescribed saltpetre based cement for the solid state refining of impure gold (Blochmann and Phillott 1989, 21). The cement consisted of equal amounts of saltpetre and brick dust of 'raw bricks'. This method was called the *Salonī* process, meaning the process of imparting a beautiful appearance in terms of colour to the impure gold. Clearly, this method was using raw soil used in brick making. Maclagan (1890, 21-2) described the details of the cementation process used for the solid state refining of gold in India. Mixtures of common salt and brick dust (also known as *surkhī*) or *oplā* (also spelled as *uplā*) ashes, *ie* ashes of cow dung cakes, were used as cement material in places such as Delhi, Hissar, Ambala, Gurudaspur, Gurgao (Gurugram) and Jallandhar in northern India. At Amritsar, the cement consisted of alum, common salt and *oplā* ashes. It is also interesting to note that a mixture of powdered nitre (saltpetre) and *kāhī miṭṭī* (an earth containing iron sulphate) was also used for gold refining in Jallandhar. Viccajee reported that the cement mixture consisting of red (*ie* baked) brick dust (2 parts) and sodium chloride (1 part), together with tamarind water, was used for gold assaying in Hyderabad (Viccajee 1908, 17).

Availability of *lavaṇa* substances in the Indus Valley area

The Indus valley and its adjoining area have been credited for the origin of Vedic culture. In the present context, it is pertinent to explore the historical aspects of the availability of *lavaṇa* type of substances in this area. It is well-known that Indus valley and its adjoining area have been an important source of different types of saline materials since time immemorial. The Sanskrit word *Saindhavikā* was cited for the saline Indus earth in the famous treatise Arthaśāstra, as discussed above. Salt was produced along the coast line in Kathiawar and Kutch and on the edges of the Rann of Kutch from time immemorial (Aggarwal 1937, 233). Āīn-i Akbarī mentioned salt-pits in Tatta (also spelled as Tattah and Thatta), situated in Sindh Province (Jarrett and Sarkar 1989, 339). A detailed description of the availability of *lavaṇa* type of substances in the Indus valley region is available in pre-modern references. The valley of Indus in most part was flat in appearance, and was strongly impregnated with salt. Such tracts were known as *pāṭa* (Aitken 1907, 2). In many parts of Sindh province, in particular Tatta in the Lower Sindh, efflorescence of muriate of soda (sodium chloride) and saltpetre formed after the fall of rains, due to the decay of animal and vegetable matter on the upper surface of the soil (Winchester 1855, 274). The waters of the Indus river in the lower part of its course exhibited 'a foul and muddy appearance', and also 'these waters contain, in a state of solution, a proportion of saline ingredients, principally common salt, carbonate of soda, and nitrate of potash (saltpetre)' (Carless 1855, 490). The soil of many parts of Sindh contained a large proportion of common salt, together with carbonate of soda and saltpetre (Heddle 1855, 418). It is interesting to note that mixtures of common salt and alkalis were called as *lavaṇakṣāra* in Sanskrit (Tarkavacaspati 1962, 4824). M'Murdo (1834, 229) stated that the country around Tatta and various other places of the Sindh province produced great quantities of refined saltpetre from crude saltpetre. He also stated that the shores of the delta in Sindh were a huge source for the supply of common salt. In the 17th century AD the East India Company was purchasing saltpetre from Tatta of the Sindh province for export to Britain (Foster 1921, 311).

Hughes (1876, 472) mentioned the names of the places in the Sindh province, which were producing salt, saltpetre and alum. Delhoste (1855, 262) mentioned that the hills between Sonmeeanee to Sehwan, and thence to Kurachee (Karachi) in the province of Sindh contain alum. Nitrous earth, from which saltpetre was produced,

was known in India from very early times. According to an estimate reported by Leather and Mukerji (1911, 4 and 7), most nitrous earths in India contained 3-5% nitrate and, in addition, several percent of chlorides and sulphates. They also reported that the crude saltpetre obtained from Indian nitrous earth contained 27-68% potassium nitrate together with 17-35% sodium chloride. Punjab province was an important producer of high quality saltpetre from its local nitrous earth (Cotton 1919, 303-4).

Burnes stated that alum was found in the hills of the Salt Range (Burnes 1834, 52). Alum was also available in the Saurāṣṭra region of Gujarat state of India, and for this reason one of its names in Sanskrit was *Surāṣṭrajā*. Further, another name of alum in Sanskrit was *hemaśodhanī*, as it was used in the refining of gold (Sharma and Sharma 1979, 286).

Indian gold beads of archaeological origin

An important characteristics of diffusion bonding of similar materials carried out under optimal conditions of surface cleanliness, pressure, temperature and time, is the absence of physical interface whatsoever between the pieces being joined (Kazakov 1985, 43). Some of the gold beads discovered from the excavations carried out in the Sindhu (Indus) river valley exhibited a characteristic of no sign of solder or very fine joint. For example, Marshall (1931, 519) stated that some of the disc-shaped gold beads (0.4"(10mm) in diameter and 0.2"(5mm) long) which were a part of a necklace discovered from the DK area of Mohenjo-daro, had 'a very fine joint'. Mackay (1938, 501 and 515) reported that a barrel-shaped bead of thin gold foil over a resinous base found from the DK area of Mohenjo-daro showed 'no evidence of solder'(it was 0.29"(7.4mm) in diameter and 0.3"(7.6mm) long). He also stated that a tapered cylindrical gold bead (SD 3132; 0.7"(18mm) long by 0.1"(2.5mm) in diameter at one end and 0.05"(1.3mm) at the other) showed 'no sign of lapping or soldering'. It is difficult to opine on the actual manufacturing method used for these gold beads in the absence of scientific studies. However, there is a strong possibility that the gold beads reported by Mackay might have been made by diffusion bonding.

Joining of gold with the aid of *lavaṇa* type of materials, in the reference under consideration, has been used as a simile. The details of the process are not to be expected from this reference. The possible procedures adopted for the diffusion bonding of gold have been discussed above. Flat-flat geometry as used in lap joints of barrel-shaped beads would have been the most amenable for achieving good quality diffusion bonding of gold. Lap joint area of such gold bead preforms would have been easily accessible for light hammering to ensure intimate contact between the surfaces to be joined.

Summary and concluding remarks

In Sanskrit literature, there are several indirect references dealing with principles of metallurgy. One such reference in the Chāndogya Upaniṣad, in the form of a simile dealing with the joining of gold, has been discussed here. According to this reference, gold pieces were joined in the solid state with the aid of *lavaṇa.* This is an important ancient reference for the solid state diffusion bonding of gold, without using any external filler metal. The process consisted of two steps: cleaning of the gold surfaces to be joined, followed by heating together the gold pieces in direct contact. In ancient times, the Sanskrit term *lavaṇa* was used in India in a generic form for a variety of saline and alkaline substances. Chemical substances, such as common salt, saltpetre, borax, alum and different types of alkalis were classified under the general category of *lavaṇa.* Further, both unrefined native gold and intentionally prepared alloyed gold were used for making different products in ancient times. Such gold pieces also contained metals which formed oxide films on their surface during high temperature working. This oxide film prevented the formation of a good joint between gold pieces. *Lavaṇa* was used for removing such oxide films. If borax was used, all the metal oxides present on the joint surface would have dissolved in it, resulting in clean gold surface. As a result, a good quality solid state diffusion bonding between such gold pieces was achieved. Alternatively, different types of *lavaṇa* materials were used to create a thin, pure gold surface layer on the impure gold by either depletion gilding or a cementation process. Subsequently, such *lavaṇa*-treated gold pieces kept together in intimate contact, were heated in the solid state, leading to diffusion bonding. A variety of *lavaṇa* type materials, such as common salt, alum, saltpetre etc, were readily available in the Indus (Sindh) valley and the adjoining area. The description of joints of some of the gold beads found from the excavations carried out at Mohenjo-daro suggests the strong possibility of being made by diffusion bonding.

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The author

Professor R K Dube was with the Department of Materials Science and Engineering, Indian Institute of Technology, Kanpur, India from 1978-2012. He specializes in the area of unconventional metals processing, in particular wrought powder metallurgy. He has published widely in his core area of interest and has also extensively researched and published on metallurgy in ancient and medieval India, in particular metallurgy as described in Sanskrit texts. He has contributed papers in the multi-volume project 'History of Science, Philosophy and Culture in Indian Civilization'. Professor Dube is the recipient of the Best Metallurgist Award from the Government of India, IAEC Golden Jubilee Award and Gold Medal, BHU Gold Medal, Indian National Science Academy Teachers Award among others. The book *Problems in Metallurgical Thermodynamics and Kinetics*, co-authored by Professor Dube and published by Pergamon Press, Oxford has been translated into Spanish, Chinese and Persian. He served as a member of the Editorial Board of the journal *Powder Metallurgy* published by IoM3, London. He is a Fellow of the IoM3 and a Life Fellow of the Indian Institute of Metals, Kolkata. He was a member of the Central Advisory Board of Archaeology, and also of the Second Sanskrit Commission, Government of India, representing the area of Science and Technology.
Address: Plot No 15, Ravindrapuri Extension, Lane No 2, Durgakund, VARANASI 221005, (U. P.), INDIA
Email: rkd@iitk.ac.in