



Indigenous knowledge on ancient Indian alchemical alloying

N. Anantha Krishna¹ · Debi Prasad Mishra² · Y. V. Mohan Reddy³ · B. Chandra Mohan Reddy⁴

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Abstract

In recent times, the metallic drugs synthesized through traditional Indian alchemy processes are gathering much attention owing to their therapeutic nature and physico-chemical properties. The extractive metallurgy and alloy formulation of this tradition have hardly been explored as it involves sturdy pyro-technological treatments to extract the essential medicinal properties of hard substances like minerals, metals and stones. However, through modern characterization tools, it is possible to investigate the submerged sensible patterns, and tap their new dimensions of applicability. In the present work, a critical analysis is attempted to study the materials and pyro-technological treatments followed in alloying process of Indian Alchemy. The inner crucible coatings incorporated in the ancient Indian alchemical systems are studied by Thermo Gravimetric Analysis (TGA). It is found that the tradition of using materials like Borax, ghee etc., traces back to Indian philosophies of 600 BCE. Also, it is believed that they used to extract active principles from ores, minimizing their metallic dosages and without alteration in their potency for treatment. The alloys synthesized and characterized involve Au, Ag, Cu, Zn and other minerals like *śilājī* and *svarṇamākṣika* (Chalcopyrites).

Keywords Ayurveda · Indian alchemy · *Rasaśāstra*

1 Introduction

In ancient times, the methods adopted by the blacksmiths, alchemists and astronomers were in congruence with the nature. Accordingly, the perspicacious inferences on metals, flames and phase changes were ingenious and in concord with modern characterization techniques. Discerning these procedures could be a remarkable breakthrough in pyro-metallurgy. In *Atharvaveda*, the hymns for cure of diseases and of demonic possessions of diseases are known as *bhaiṣajyani*, while those which have the objective of life rejuvenation and prolongation are known to be as *ayusyani*. The later led to *rasāyana*, which is Sanskrit equivalent to

alchemy (Ray, 2014, p.37). One cannot say that the origins of alchemy is only post-*Atharvaveda* as there are earlier mentions of *soma* herb (*Ephedra*) (body energizer) preparations. The use of metals in medicines dates back to the period of Suśruta (400 BCE) however, its omnipotence was hugely propounded by eighth century specifically during Tantric period of ninth century. Indian alchemy came to full agility during tenth–fourteenth century. With increasing time, many inorganic materials piled up into their *Materia-medica* and many pyro-metallurgical procedures heaped into their texts (Prakash, 1997, p. 3).

In these incinerations and calcinations, pyro-technology forms a strong background and core component. Some of these techniques were even handed over to blacksmiths, where they quenched their weapons even in liquids like blood and urine. Their quenching techniques seem to take inspiration from alchemical procedures like *nirvāpa* (hardening) and *dravīkaraṇa* (liquification). Devarathan (2011) examined the ancient Indian alchemical concept of incineration, its related furnaces, heating systems and temperatures. Durand Charee (2004) traced the journey of the steel from Iron and explored many ancient metal making technologies and mining practices. Ranganathan (2006) unfolded the ancient Indian metal, wootz's legacy: its synthesis,

✉ Debi Prasad Mishra
dpmishraikt@gmail.com

¹ Department of Mechanical Engineering, Sree Vidyanikethan Engineering College, Tirupati 517102, India

² Department of Aerospace Engineering, IIT Kanpur and Director, NITTTR, Kolkata 700 106, India

³ Department of Mechanical Engineering, GP Reddy Engineering College, Kurnool 518007, India

⁴ Department of Mechanical Engineering, JNT University Anantapur, Ananthapuramu 515002, India

deformation and crystallography. Sherby and Jeffery (2001) have done extensive research on ancient metals, especially ancient blacksmiths, Damascus steels, and Iron. The critical evidences of studies on ancient metallurgy are an important contribution in contemplating the evolution of man, civilization and knowledge they ameliorated by their experience of innate relationship between nature: metals, plants and minerals. They quoted their entire work in a single proverb as “the best of the new is often the long forgotten past”. Their work highlighted the metallographic legacy of ancient blacksmiths, their artistic accomplishments revealing deep roots in alchemy and astronomy.

Some researchers have traced the origins of an ancient crucible metal from Sanskrit etymology. They analysed its hardness. The morphological studies indicated the presence of several particles adhered to its surface indicating the formation of liquid phase. Their research emphasizes the significance of deciphering ancient Sanskrit texts through etymology (Dube, 2014, Williams, 1990, Kant, 2002). Guisbiers et al. (2014) have studied ancient Au–Cu nano-alloys and found that Au–Cu phases were being employed, even by pre-Columbian civilizations in the decorative arts. The nano-alloy was named “*Thumbaga*” and was explored in depth, along with its molecular dynamics and nano-thermodynamics. Marianne Reibold, (2006) investigated a sample of Damascus sabres of the seventeenth century. Both cementite nanowires and carbon nanotubes were found. They proposed an interlink between impurity segregation, nanotube formation, nanotube filling, cementite wire growth and development of huge cementite particles. Schaming and Remita (2015) have reported that nano-sciences and technologies appear as new concepts of twentieth century, but metallic nano-particles were already been in usage since ancient times. The growing body of literature survey confirms that nano-powders were in use since earlier times in ceramics, medicine and other art works.

Neogi (1979) has studied the legacy of copper in ancient India. His work encompasses copper in vedic (circa 2000 BCE–1000 BCE), Brāhmanic (circa 1000 BCE–500 BCE) and the epic age (circa 500 BCE–200 BCE). He mentioned that there was no bronze age in India, but only copper age; his work is a treatise on ancient Indian copper for alchemy, metallurgy and other archaeological sources. Bhanu Prakash (1997) has done extensive research on pyro-metallurgical extractions, and incinerations, mentioned in Indian alchemy with respect to iron, lead and copper. It lays the foundations of ancient heating system (*puṭa*) and analyses the diversifying heating ranges of the system. Also, several researchers considered the usage of metals in ancient medicine where they have studied the ancient concept of metallic drugs and their application (Dixit, 1993, pp. 1–3; Jha et al. 1989, p. 17; Jha, 1990; Joshi, 1993; Mitra and Sayakhare, 1993; Nataraja, 1993; Parish, 1992).

Jagtap et al. (2014) work transpires on purification and detoxification procedures (*śodhana*), with special reference to copper in medieval Indian medicine. It elucidates several metallurgical phenomena like hydrogen embrittlement, nano-crack formation and bio-beneficiation, as innate aspects of the traditional process. Shuchi Mitra (2009) studied the effect of media in detoxification procedures employed in traditional alchemy. He emphasized the concept of bio-sorption and bio-accumulation as reasons behind toxic metal separation during the *śodhana* process. Kapoor (2010) worked on critical analysis of ancient Indian metal based medicines. The work analyzed heavy metals and incinerated metals (*bhaṣmas*). It draws parallels between *bhaṣmas* and Oxide of metals. It also emphasizes on safety aspects and standardization in metallic medicines.

Arun Kumar Biswas (1987) revealed the antiquity of science in the thirteenth century through study on *Rasaratna Samuccaya* dealing with the art of mineral processing. The work accentuated all the alchemical concepts authored by Vagbhata. Mira Roy (1984) conceptualized the alchemical seed principle mentioned in *Samarāṅgaṇasūtradhāra*. It was applied to reverse engineering machinery and mechanical contrivances as mentioned by the author of the text King Bhoja. It emphasized that the aforementioned principle was formulated on two innate ideas— theory of micro and macrocosm and generative principle of a seed. This triggered insight for alchemical metallurgists to reveal that any matter contained seed in it led them towards transmigration.

Barbara Obrist (2003) worked on medieval alchemy. The work focused on the portrayal of the image in alchemy and was mentioned as a Latish phenomenon with a high substance on metaphors and geometrical patterns; the part of much pictorial representations can be attributed and analyzed in the status of prevalent scientific conceptions and methods of transmitting knowledge. They investigated on traces of nano-particles in materials and nano-particle layers were found in medieval ceramics with metallic lusters. They also worked on characterization of the aforementioned specimens through SEM and TEM studies and optical properties of these layers were studied in detail during the investigation. Similar attempts were also made by other researchers (Philippe Sciau, 2012, Angelini et al., 2004, Bobin et al. 2003).

Muralidhar and Mohan Kumar (2016) conceptualized *śodhana* (pre-treatment/detoxification) procedures of different metals, minerals, etc. by surveying eighth century texts and found that detoxification is classified as *sāmānya* and *viśeṣa* (general and specific) in terms of degrees of impurity removal and also classified pyro and non-pyro technological process. Hanchezhian et al. (2010), Dhama et al. (2005), Mehta et al. (2007), Sarkar & Prasanta Kumar (2008) have worked on the effect of media in detoxification of poisonous herbal drugs, *Datura metel* Linn, *Aconitum ferox* Wall and *Semecarpus*



anacardium Linn. Their studies showed that toxic constituents transferred into media enabled the drug becoming nontoxic and emphasized the importance of specific media for internal administration. Similar findings were also attempted by other researchers on the role of different media involved in detoxification. Rabinarayan Acharya (2015), Bhargav et al. (2013), Bhargav (2012), Hanchezhian et al. (2012), Sarkar (2008), Sarkar and Prajapathi (2008) Sujatha et al. (2013), Sathaye (2012), Tripathi et al. (2008) built up a colossal collection of writing on *śodhana*. It was found to be a detoxification strategy to upgrade the potency and productivity of a mineral/metal/home grown medication and eventually diminishing its dangerous properties. Parallel work on investigative, phytochemical, phyto-pharmacological assessment of *śodhana* (detoxification) and its impacts on various materials were endeavoured by not many researchers.

Sudheendra and Reena (2012) contemplated the impacts of detoxification strategies on copper and found that components like Fe, S, Ni, Al, Si and Pb have lessened fundamentally after these systems were included. Jagtap et al. (2014) worked widely on restorative employment of old Cu drugs mentioned in *Bhaiṣajya Ratnāvalī*. More than 20 sedate equations were contemplated by them on 30 distinct kinds of ailments from fever to heart issues. Pattanaik et al. (2016) studied and characterized physio-chemical parameters of detoxified copper metal granules through *śodhana* method using SEM and XRD and found drastic variations in particle sizes before and after purification.

Kashinath et al. (2017) presented an in depth analysis on alchemical quenching (*nirvāpa*) with respect to copper detoxification and studied the effect of quenching on PH of the liquid media. Chavan (2016) suggested the role and importance of *nirmalīkarana* in detoxification of *tuttha* (CuSO_4 and H_2O). It removed physical impurities like silicon, potassium and increased sulphur, iron, zinc and reduced calcium contents. Mahapatra and Mahapatra (2013a, b) worked specifically on the role of *śodhana*, detoxification on anti-microbial activity of *tuttha* on more than six varieties of organisms. The work further provided detailed physico-chemical analysis of *tuttha* and *tuttha bhaṣma* (ancient copper drugs) prepared through three different eighth century procedures. Raman and Archana (2012) made an attempt to study alchemical detoxification treatments of Fe, their synthesis and classification. In this work, attempts are made to study the exacting pyro-technological recipes in alloy making. Accordingly, the extracts of the crucible are analyzed through morphological and thermo-gravimetric analysis.

2 Materials and methods

All the materials involved in the process are commercially pure grade and acquired from local market. The pre-treatment of metals viz. gold, copper, zinc and copper extracted

through copper sulphate are taken in equal quantities in a crucible coated with a mixture as mentioned in alchemical texts (Sastri and Krishna, 1952; Sharma, 1954; Agnivesh, 1976; Sharma, 1954; Mukherjee, 1984; Shastri, 1995; Padacharya, 1989). The crucible involved in the present work is a clay crucible with a lid on the top. Furthermore, the type of furnace involved is of *aṅgāra koṣṭi* type as mentioned in Ayurveda texts. It is a charcoal fired, single blower, crucible furnace capable of attaining 1500°C. A mixture of *guḍa* (jaggery), *guggulu* (gum of *Balsamodendron mukul* Hook), *lākṣā* (lac), *sarjikā* (an alkali material containing sodium bicarbonate), *saindhava* (rock salt), sulphur, *taṅkaṇa* (Borax), guṅja seeds (*Abrus precatorius*), Magnetite (Iron oxide), in equal quantities are mixed well with, ghee, and butter, in a mortar and pestle. Further, this is applied as a coating to the inner portions of the crucible. Also, the same mixture is grinded well with minerals or metals involved in alloying, forming a bolus. This mixture is referred as herbo-mineral coating or specific coating. The thermo-gravimetric analysis (TGA) of such coating is presented in detail by Anantha Krishna et al. (2018). The molten alloy was further characterized using Scanning Electron Microscopy (SEM), a method for high resolution surface imaging to exploit the surface topographies of the synthesized alloy. The SEM make is JEOL Model JSM-6390LV with resolution of 3–15 nm and magnification of 5× to 300,000× (both in low and high volume modes). The Thermogravimetric analysis (TGA) was performed with Diamond Thermogravimetric/Differential Thermal Analyser having temperature range, RT 1500°C. It is a differential type furnace with air, inert gas as atmosphere having 10–2 torr vacuum. Similarly such alloys involving other minerals like bitumen, chalcopyrite, iron, etc. in the above said manner were synthesized and characterized using SEM. The initial sample weights of both specimens with and without the drug are 25 mg and 24 mg respectively.

3 Results and discussions

Śodhana (a quasi-equivalent term is purification/detoxification) of raw materials in extractive metallurgy is considered to be the most essential part in Indian alchemical metallurgy. It was known as *samskāra* and was believed by the ancient Indians that this process transmutes the latent attributes of a substance which leads in summation of new ones. This belief can also be seen in western alchemical systems especially of Geber who was known to be the pioneer alchemist. Various modes of such treatments such as *svedana* (boiling), *manthana* (churning), *mardana* (grinding), *bhāvana* (impregnation) etc. are mentioned in Indian alchemical texts. Each treatment has a unique procedure for the specified raw material. Anantha Krishna et al. (2018) studied the ancient



Indian alchemical alloying process of Cu–Au–Zn alloy through thermo-gravimetric and morphological studies.

The thermo-gravimetric analysis of the aforementioned coating involving honey, clarified butter, sulphur etc. is discussed in detail by Anantha Krishna et al. (2018). The corresponding thermo-gravimetric analysis (TGA) and differential-thermal analysis (DTA) of Cu–Au–Zn alloy along with coating is shown in the Fig. 1. The degradation occurs in more than five stages with initial temperature between 90

and 100°C with evaporation of moisture content before this cut-off. After this material degrades slowly in successive stages, it finally remains with a residual mass of 44.29% at 799.6°C. Similar observations can be found when such analysis is made on Cu–Fe alloy along with coating with a residual mass of 44.94% at 799.7°C shown in the Fig. 2. The inflection points in both the cases can be attributed to the presence of inflammable substances present in the coating such as sulphur and some resins like lac, *Commiphora*

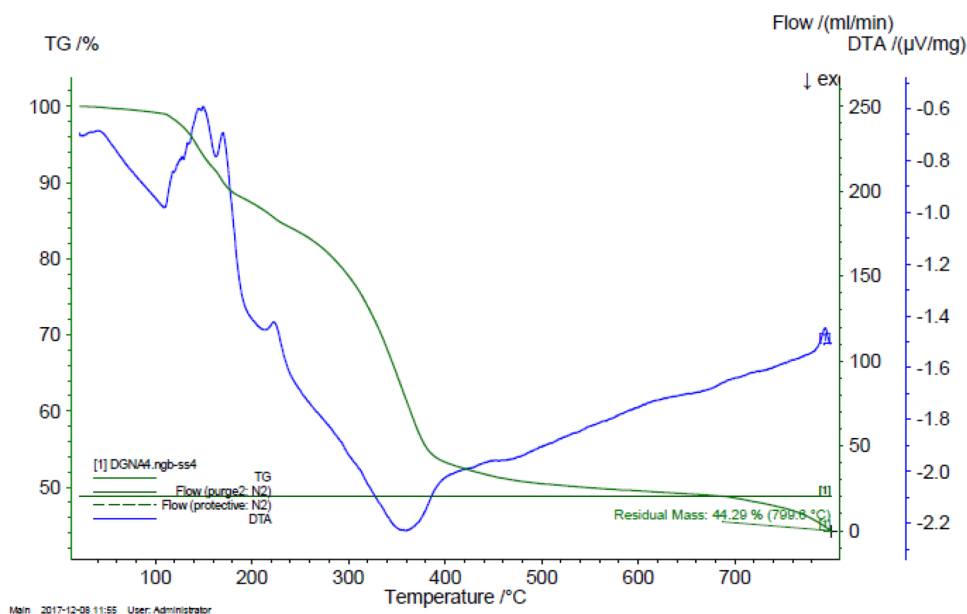


Fig. 1 TGA and DTA of Cu–Au–Zn Alloy including crucible coating mixture

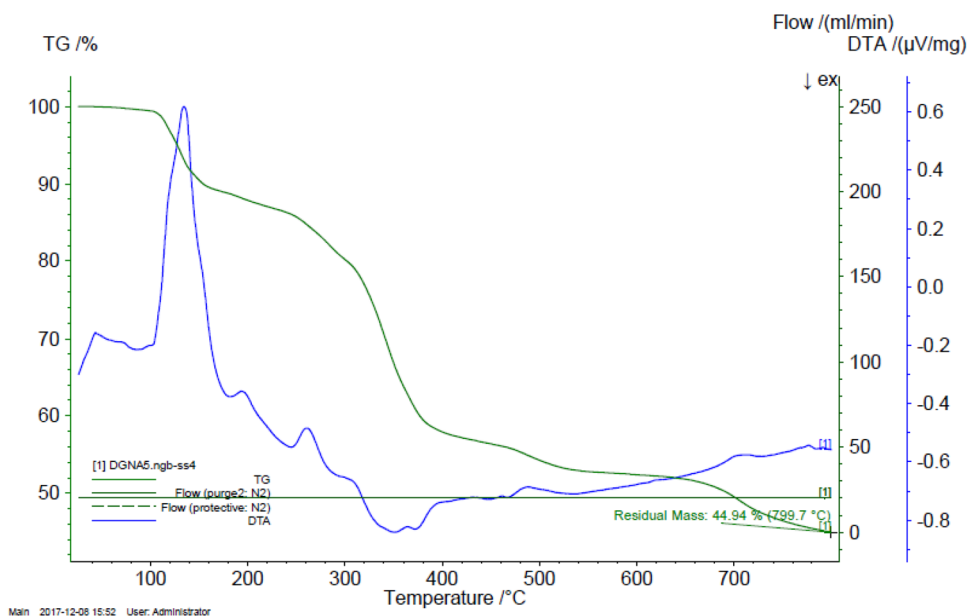


Fig. 2 TGA and DTA of Cu–Fe alloy including crucible coating mixture



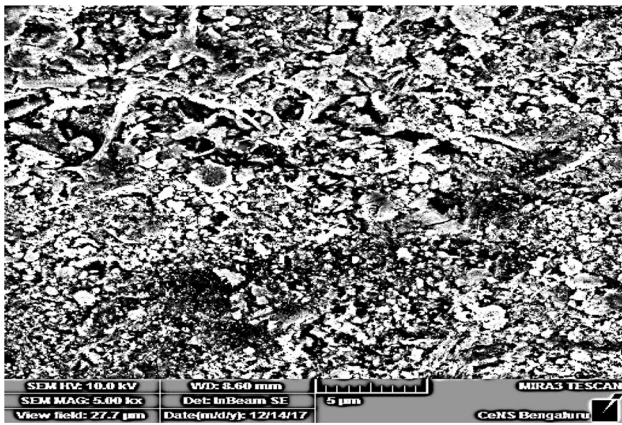


Fig. 3 SEM image of 5000× magnification depicting bitumen and iron oxide

wightii and damer resin etc. This is evident from DTA curves from both images.

4 Morphological studies

The contents of the crucible for alloying—copper, iron, bitumen, chalcopryrite are powdered in a heavy mortar and pestle and is further mixed with specific coating as mentioned in classic texts (Krishna et al., 2018; Sastri and Krishna, 1952; Sharma, 1954; Agnivesh, 1976; Hemraj, 1994; Mukherjee, 1984; Shastri, 1995; Padacharya, 1989). The prepared mixture is kept in a crucible, tightly closed and set on fire. The typical TGA and DTA of the same are depicted from Fig. 6. Here it can be observed that, in the temperature zone of 320–390°C, very slow and constant rates of melting were observed, which can be attributed to the coating involved. Also, silver, copper, iron, bitumen and chalcopryrites were considered in the next combination, and following the same lines of thought, the synthesized alloys were characterized using SEM and Edax.

5 Characterization of the crucible content

It can be observed through the high magnification of SEM photo-micrograph that very fine copper, which has been crystallized along with some large grains of the same shows that copper is administrated in two forms, one as large flakes and the other as essence extracted from copper sulphate as fine powder.

The presence of solidified grains with bitumen as dark constituents in the interface can be pinpointed in Fig. 3. Also, the iron oxide of chalcopryrite or iron oxide involved in the coating may also be suspected because copper has capabilities to dissolve in iron. The appearance of oxides

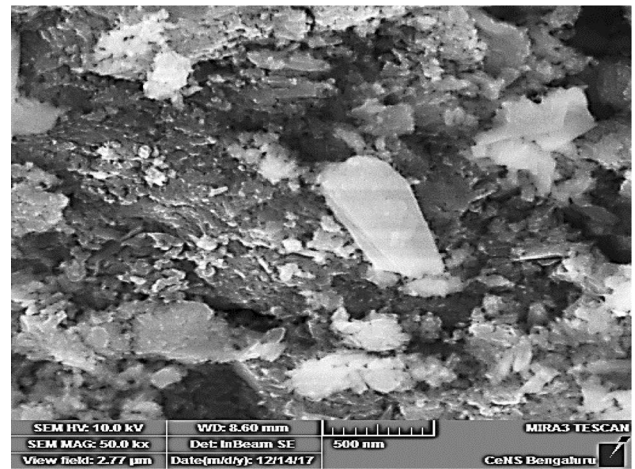


Fig. 4 Microstructure of crucible content exploring the un-vitrified lumps

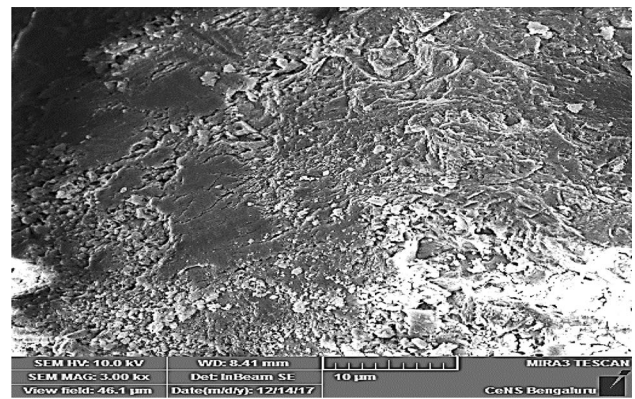


Fig. 5 SEM image at 3000× of the dark diffused particles of composite Bitumen additives

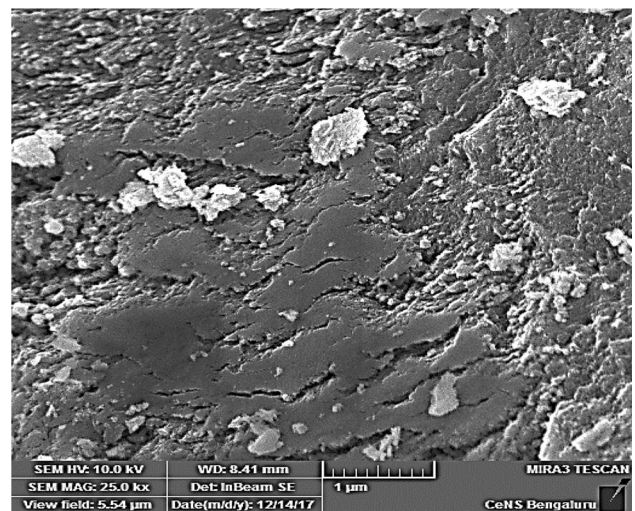


Fig. 6 Solid copper with coated additives and calcium oxide



may be due to the involvement of powdered magnetite in the coating and in the agglomerated mixture or from the oxidation of the iron metal and chalcopyrite in the sealed crucible with lid.

Further, the higher magnifications reveal copper as distinct particle and it can also be seen that some of the unvitrified lumps that were formed probably have not gone deep into the solution and might have been left as solids from base materials, which can be seen in Fig. 4. Also, the dark constituents of additives added to solidified copper are observed; it also portrays iron oxide with the solidified grains. This fine grained copper mass along with dark diffused particles of composite bitumen additives is shown in Fig. 5. The herbo-mineral coating given to the crucible form dark constituents with a minimal presence of white oxides of calcium; this can be observed in the Fig. 6. Further magnification reveals same evidences along with oxide particles, fine lumps and some finer debris adhered to the base of the solid metal. This metal could be iron with copper. Alternatively, the smooth surface could be copper and the fine porous surface between the solid could be chalco-pyrites. The Edax evaluation shown in Fig. 11 supports this argument with high iron and oxygen content; hence, the rough matrix could be with higher iron content.

The surface morphology shown in Fig. 7 is the powdery and sintered surface of different particles with some fine copper particles in between the grains. There is a possibility that the additive powders formed a lump with some solid particles of copper. Being at lower magnification, the powdered non-metallic surface shows the grains bonded together as lumps. The composite matrix with metal solidified containing un-dissolved non-metallic ore particles is shown in Fig. 8. The free solid copper is observed as isolated particles in the powdered and solidified lumps. Probably, the copper atoms are bond and hold the powder lumps.

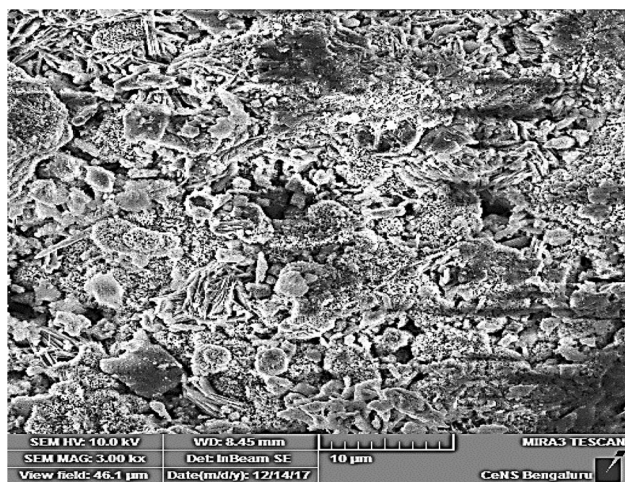


Fig. 7 Lump formed from additive powders and solid Cu particles

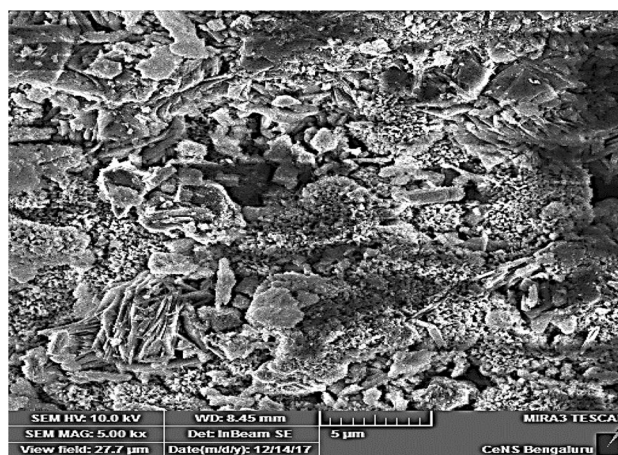


Fig. 8 The composite matrix with metal solidified containing un-dissolved non-metallic ore particles

This is supported by the evidence from alloying chapters of *rasāyana* texts such as *Rasaratna Samuccaya* and *Rasahrdayatantram*. Fig. 9 shows the SEM photo-micrograph at 10,000× which has resolved more of copper solid which is solidified. However the non-metallic composite particles formed honey comb structure.

Fig. 10 shows the SEM photo-micrograph at 50,000× which has resolved the surface which shows uneven surface with both metallic and powdered and sintered matrix. There are possibilities that all the ore particles along with diffused bitumen molecules could be present. The Fig. 11 and corresponding Table 1 show the presence of low carbon (from bitumen) and high iron (from chalco-pyrites). The extracted alloy images from top portion of the crucible are shown in Fig. 12. Its corresponding Edax image is shown in Fig. 13 with rich copper content along with carbon due to

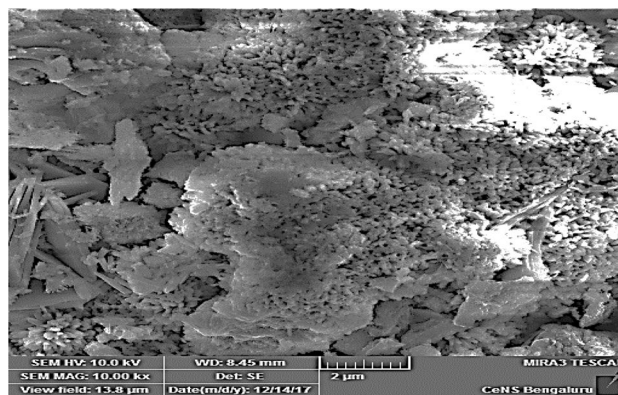


Fig. 9 Solidified copper with non-metallic composite particles forming as honey comb structure



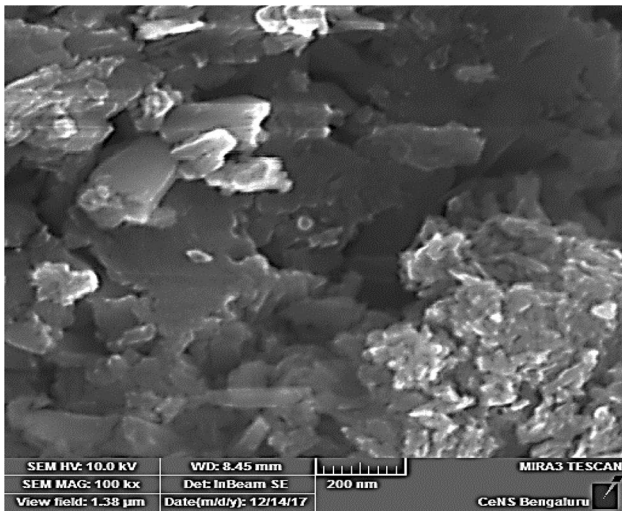


Fig. 10 Uneven surface with both metallic and powdered and sintered matrix

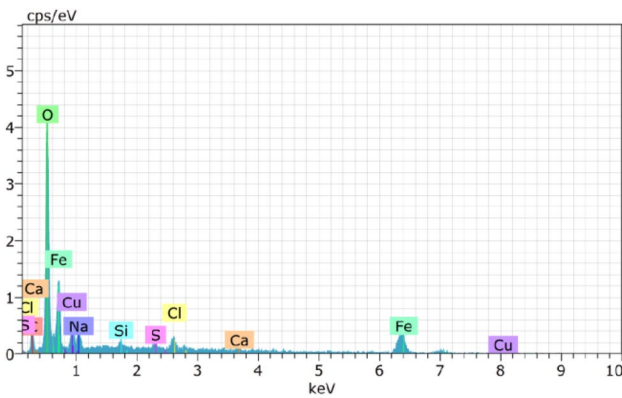


Fig. 11 Edax image of bottom crucible contents with Cu, Fe and its ores

Table 1 Elements in bottom crucible contents with respect to Cu, Fe and its ores

Element	Series	wt%
Carbon	K	4.03
Oxygen	K	29.19
Sodium	K	1.88
Silicon	K	0.63
Sulphur	K	0.46
Chlorine	K	1.35
calcium	K	0.60
Iron	K	63.72
Copper	L	6.82

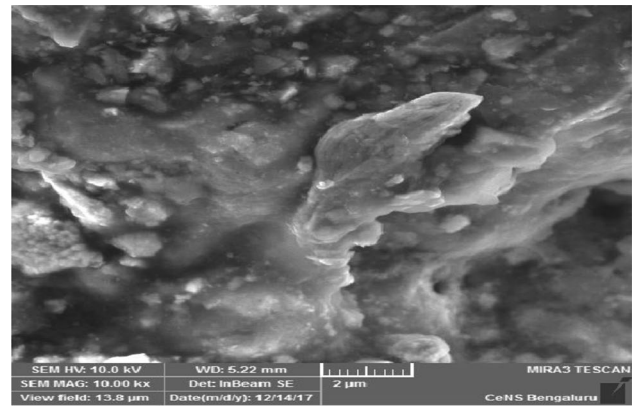


Fig. 12 Top portion of the alloy (Cu, Fe and minerals) with presence of carbon and other debris

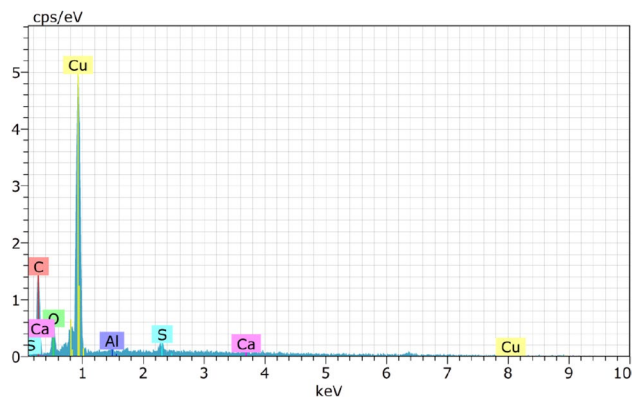


Fig. 13 Edax image of top portion of the crucible (Cu, Fe and minerals) with presence of carbon and with no traces of Fe

Table 2 Elements present from the top portion of the crucible (Cu, Fe and minerals) with presence of carbon and with no traces of Fe

Element	Series	wt%
Carbon	K	23.82
Oxygen	K	5.54
Aluminium	K	0.49
Sulphur	K	1.14
Calcium	K	0.87
Copper	L	68.14

the herbo-mineral coating as depicted in Table 2. Also, the minimal presence of sulphur can be seen due to its usage to accelerate the melting process as a coating material. Similar attempts were made with silver and the aforementioned materials instead of iron as shown in the Fig. 14 which can be confirmed with the Edax of the same in Fig. 15 and Table 3.



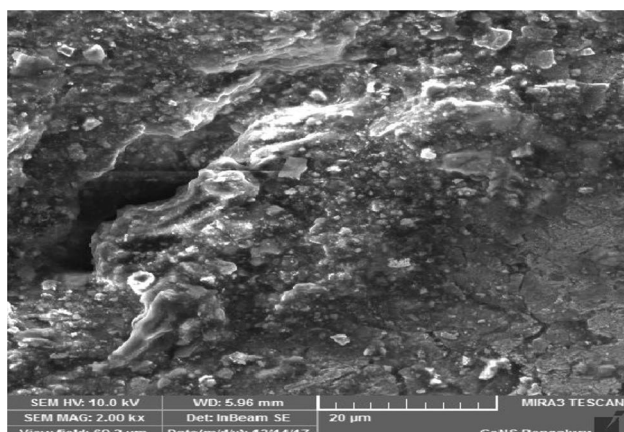


Fig. 14 Top portion of the alloy (Cu, Ag, Fe and minerals) with presence of carbon and other debris

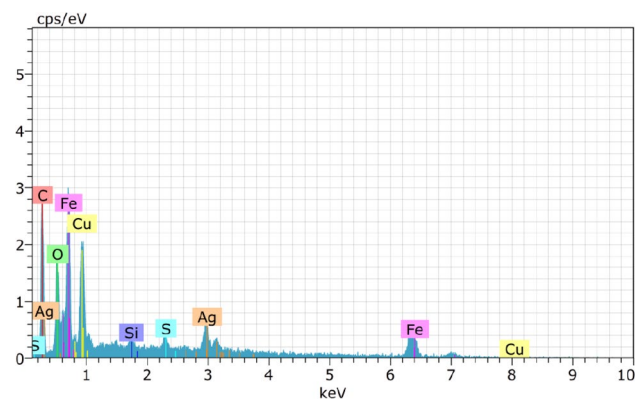


Fig. 15 Edax image of top portion of the crucible (Cu, Ag, Fe and minerals)

Table 3 Elements present in the top portion of the crucible (Cu, Ag, Fe and minerals)

Element	Series	wt%
Carbon	K	17.47
Oxygen	K	8.91
Silicon	K	0.33
Sulphur	K	0.68
Iron	K	49.93
Copper	L	20.60
Silver	L	6.95

6 Conclusion

In conclusion, alloys involving different metals and minerals are characterized by SEM-Edax and TGA. TGA studies are performed on herbo-mineral crucible coating during the process of alloy making and metal extraction.

It revealed the significant thermal stability during 500 to 690°C with 75% weight loss at 1010°C and with final residue of 11% at 1200°C. Further, the morphology of the crucible content reveals rich iron at the bottom part of the crucible due to agglomeration of bitumen and chalcopryrite. The detoxification procedures like heating and dipping, trituration, shade drying, etc. might have enhanced the metal extraction and herbo-mineral coatings which might have aided in arresting the molten state of metals and further strengthen the process of alloying.

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