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Gold *Bhasma* and Silver *Parpam* Used in Indian Traditional Medicines. Scientific Validation of their Interaction with Human Cells

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Review Article

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Abstract

Bhasmas in Ayurveda and *parpams* in Siddha medicine are unique herbo-metallic/mineral preparations, effective remedies, fabricated from highly purified metals, treated with a variety of herbal decoctions and incinerated at high temperatures to, finally, obtain a metal ash, having a significantly reduced size and free of toxic effects. The processing techniques of bhasmas and their use as medicines have been described in ancient texts of Ayurveda such as *Rasa Shastra*, *Charaka Samitha* and *Sushruta Sambita*. It has been emphasized that, while *Siddha* medicine is close to Ayurveda, *Siddha* has been closely linked to the Tantric religious movement, traced back to the 6th century AD and it is believed that Alchemy played a more central role in *Siddha* medicine than in Ayurveda. Some of the most important *bhasmas* and *parpams* are briefly described; their fabrication and properties are mentioned.

The study of the interaction of gold *bhasma* and silver *parpam* with human cells investigated by our group is described in the second part of this work. In this section, the cellular uptake and localization of the gold and silver particles in cancerous and normal cells have been elucidated by using, principally, the hyperspectral imaging method that combines the image with the spectral information.

Keywords: Herbo-metallic Drugs; Ash; Cellular Entry; Cellular Localization; Nano-Bio Interactions

Introduction

In the Hindu spirituality, *Bhasma* is a more general term, used for any ash product, for example, *Vibhuthi*, the sacred ash, used in Hindu rituals as symbol of purity. In the modern system of medicine, metals and minerals as such, cannot be used for therapeutic purposes as they may be harmful. In Ayurveda and Siddha, the most important Indian traditional medical systems, the use of metals is recommended in the form of *bhasmas* (herbo-metallic drugs, literally meaning ashes), claimed to be free of toxic effects, when prepared properly and used in proper doses [1,2]. Similar herbo-metallic drugs are in use in other Asian countries as well [3]. The ancient Ayurvedic and Siddha practitioners had learned how to use the metals in a non-toxic, absorbable form, through transformations achieved, using various herbal products**.** The therapeutic potential of herbo-metallic drugs has been advocated in ancient texts of Ayurveda such as *Rasa Shastra*, *Charaka Samitha and Sushruta Sambita* that contain descriptions of the processing techniques of metals and their use as medicines. Interestingly, formulations containing mercury (*Parada*) are rarely mentioned in

Charaka Samitha and the first recommendations are only for external use. References on other metals such as gold *(Swarna*) can be traced back *to Charaka*. *Tamr*a (Copper): known during pre-Vedic times (in *Charaka*, the term *Arka* is used for *Tamra*). Charaka advocates the use o*f Tamra Patra* in some pharmaceutical procedures and strongly recommends copper vessels to keep water [4].

Usable forms of metals were produced with the development of *Rasa Shastra,* the science of alchemy. Both Rasa Shastra and Siddha have devised methods for detoxifying metals, before they are reduced to ash. They are called *suddhi murai* in Tamil and *sodhana* in Sanskrit, (*chunnam/bhasman*) but their methods are different [5].

In Siddha, the metals are purified, either by the repeated heating of sheets or coarse powders of metal and immersion in various plants' decoctions, or by *marana*, by "destroying" of the metal by use of herbs**,** so that the metals lose their identity and become fine powders. After the metals underwent extensive purification (called *shodhan* in Sanskrit), they are incinerated several times in earthen pots, at specific temperatures, to make them ready for human consumption**.** In both Ayurveda and Siddha, it is believed that metals and minerals, combined with herbs, help the assimilation by the human body. It is thought that the toxic effects of heavy metals are neutralized by the medium (carriers) as well [2].

The Siddha pharmacology has been closely linked to the Tantric religious movement that can be traced back to the 6th century AD. The alchemical part of Siddha appears at least from the time of Tirumular's Tirumandiram (6th or 7th cent. AD), where various alchemical preparations are mentioned.

Alchemy is also found in Sanskrit texts from North India, from about the same period, and only later became an integral part of Ayurvedic medicine called Rasa Shastra. In the classical treatises of Ayurveda, only certain metals and minerals are mentioned in the late classical treatises of the 7th century AD, by the author Vagbhata. It has been emphasized that Alchemy plays a more central role in Siddha medicine than in Ayurveda and in Rasa Shastra, mercury and sulphur are the corner-stones of Siddha pharmacology. The most important ingredient in almost every Siddha alchemical preparation is mercury. Common to both Siddha and Ayurveda are six pharmaceutical preparations: calcinated metals and minerals (chunnam), powders (churanam), decoctions (kudinir), pastes (karkam), medicated clarified butter (nei), and medicated oils (ennai).

Generally, to prepare bhasmas, metals are made into coarse powders by hammering and subjected to Shodhana (purification). For this, the metals are heated to red hot or melted and quenched in a particular liquid media for specified duration. Then, the purified materials are subjected to Marana, then mixed with specific drugs for incineration (Maraka Dravyas) and levigated (Bhavana) by particular liquid media for a specified time [6-9]. Chakrikas (pellets) are prepared from the levigated mass and taken into earthen crucibles faced together, with the junction sealed by a mud smeared cloth. This apparatus, called Sarava Samputam is subjected for heating in a traditional Puta (heating grade) or an electric muffle furnace. Burning is continued for a specific time limit and when cooled down, the apparatus is opened to get the incinerated powder. The pharmacology of the different bhasmas is described and therapeutic uses, diet and adverse effects are indicated [1].

The pharmaceutical processes like Shodhana (purification/detoxification), Marana (incineration/ calcinations) transform bhasmas in highly effective forms, without any inconvenient effects in the therapeutic dose [10].

Comparing the fabrication of traditional bhasmas and the green synthesis, it can be seen that both are using plant extracts, but their role is different. While, during the fabrication of bhasmas, it is believed that the plant extracts help to detoxify it and make it easiy absorbable by the body, in the green synthesis, the plant extracts work as reducing agents. The incineration process of bhasmas removes all the organic compounds from the plant extracts, leaving the pure metal, or/and metal oxide, while green synthesis may need additional separations and purifications [11].

Important Indian Ayurvedic *Bhasmas*. History

Tamra Bhasma: (incinerated copper) was called Loha in the Vedic literature. It is important among the bhasmas for the treatment of many diseases but improperly prepared Tamra Bhasma has been quoted as a poison. In the Vedic period (in Rigveda), there are references to Ayas, a redish metal, mentioned in context with various ornaments. In mythology, it was believed that lord Prajapati has created the ores in the womb of the earth. Tamra is said to have originated from the semen of Kartikeya, which fell on the earth. By another myth, Tamra is the blood and fat of Gudakesha Asur, a demon killed by lord Vishnu. The use of metals in therapeutics was initiated in the Samhitas. It is indicated for internal use as Rasayana (rejuvenation) and for treatment of asthma and cough. It was believed that water stored in Tamra pots becomes free of all toxic effects and may cure diseases of the eye as collyrium [12]. In Samgrahas, pots made of Tamra are also used to store medicines. The use of Tamra was mentioned in the Tantric literature but was kept under secrecy until the 7th-8th century A.D. Even in the ancient period (Samhita period, 2nd century B.C. to 2nd century A.D.) many methods of the 8th century preparations have been mentioned and, after the

8th century, new methods have been elaborated. Generally, the raw materials are Tamra wires 0.5 mm diameter, without coating on the wire. The wire is converted to thin sheet by passing through a roller press. Due to heating and quenching in liquids of variable pH, Tamra larger particles disintegrate into fine particles that facilitate marana. The liquid treatment helps in dissolving impurities of the raw material, while the heat treatment removes the volatile and thermo labile impurities. Various supporting materials are used for marana of Tamra: Parad (Mercury), Gandhaka (Sulphur), Hinguls (HgS-cinnebar), Hartal (As2S3-orpiment), ManahshilaAs2S2 – realgar with the bhawana of lemon juice.

Lauha Bhasma: Iron is a noncontroversial metal for therapeutic use (as hematinics) since centuries, in east as well as in west. These drugs are known to induce some adverse reactions, thus alternative iron-based drugs from Ayurveda and Siddha were necessary. *Lauha Kalpas* are formulations with *Lauba Bhasma* the major ingredient along with herbal ingredients. Some *Lauha Kalpas* contain mercury as well [13].

Loha, Aayasa (iron) and some of its alloys were known to ancient civilizations. *Rasa Shastra* describes *Loha* as a rejuvenator that acts as a restorative. As in the case of copper, iron vessels are recommended in certain procedures. In these texts, emphasis is given to purification of metals and their conversion to fine powders [4].

Lauha Bhasma has been used for centuries, mainly for the treatment of ailments due to iron deficiency.

The preparation of *Lauha Bhasma* involves *samanya sodhana* (a general purification step), *vishesha sodhana* (a special purification step), *Bhanupaka* (reaction under sunlight), *sthalipaka* (roasting of contents in iron vessel), and *puta* (calcinations). *Bhanupaka* is a green process that utilizes *Triphala* decoction, an aqueous extract of the mixture of three dried fruits, *Terminalia chebula*, *Terminalia belerica,* and *Phyllanthus emblica* (Indian gooseberry. The raw material used for the preparation of *Lauha bhasma* is *Kanta Lauha* (iron powder). It is heated to red hot and immersed in liquids like sesame oil, butter milk, cow's urine, rice gruel. The thermal treatment with each liquid is repeated thrice and, after this, the powder is heated again and immersed in *Triphala* decoction seven times. The plant extracts were analyzed by LC-MS/MS to authenticate the plants [14,15].

Mercurial and Gold Preparation

There is a plethora of mercury containing preparations. In Ayurveda there is *Kajjali, Parpati, Rasasindura,* and *Makaradhwaja* and medicines like *Garbhapala Rasa* which have lower mercury content. Similar mercury medicines like *Linga Chendooram* exist in Siddha tradition. All forms of mercury are considered toxic. Mercury is known to affect the nervous system and has been shown to accumulate in kidney. It is important to ascertain the chemical form in which mercury is present as well as the presence of free mercury and sulfur in the preparation. The ancients purified the raw mercury and sulfur by using plants and prepared HgS, having a very low solubility (1×10^{-54}). However, the solubility of HgS may be higher in the gastrointestinal tract due to the changing pH conditions and the action of digestive enzymes. Standard texts mention eight stages of purification of mercury when natural metallic impurities are removed. For the fabrication of HgS, classical texts have recommended to use a large excess of sulfur. *Poorna chandrodayam* (PC) is an elixir of mercury and gold widely used in Siddha medicine. It is a scarlet red colored powder, soluble only in *aqua regia* [16].

It was found by XRD and XPS that the main component was HgS . The content of gold, mercury and sulphur was found to be: 9.78, 78.11 and 11.95 g, respectively. An Ayurvedic medicine of similar composition is called *Makaradhwaja Sindoora* prepared with aloe juice, instead of banana stem juice. It was prepared from purified gold, purified mercury and purified sulphur in the ratio 1:8:16 and treated with red cotton flowers for 2 days, dried and further triturated with banana stem juice for two more days, dried and processed by the *kupi pudam* technique. Another mercurial drug of Siddha medicine is *Gowri Chinthamani Chendooram*, used in the treatment of various diseases such as tuberculosis, skin diseases, osteoarthritis, etc. [17].

Interactions of Nanomaterials with Biosystems. Cellular Entry and Localization of Gold *Bhasma* and Silver *Parpam* in Human Cells

Studies on the cellular localization of gold and silver utilized in traditional Indian medicine, in the form of *bhasmas* in Ayurveda and *parpams* in Siddha are scarce and their mechanism of cellular entry is not known. In this regard, our group's comparative study on the chemically synthesized gold nanoparticles and the incinerated ancient Indian gold, *Swarna Bhasma,* proved to be compelling [18,19]. The particle sizes as well as the chemical composition of Swarna Bhasma and Veli parpam have been determined by using modern physical methods, especially, SEM with EDX, XRD, and ICP-MS (Inductively-Coupled Plasma Mass Spectroscopy) as well as DLS (Dynamic Light Scattering). Most importantly, the SEM images show that *bhasmas* (and *parpams*) are micro-sized aggregates of nano-sized crystallites, probably, due to the repeated high-temperature incineration processes. Both the gold *bhasma* and the silver *parpam* were incubated with

a variety of cells (HeLa and HFF-1 for the gold *bhasma* and A549, HCT 116, and HeLa for the silver *parpam*) for different durations and the SEM and hyperspectral images of particles inside the cells were studied.

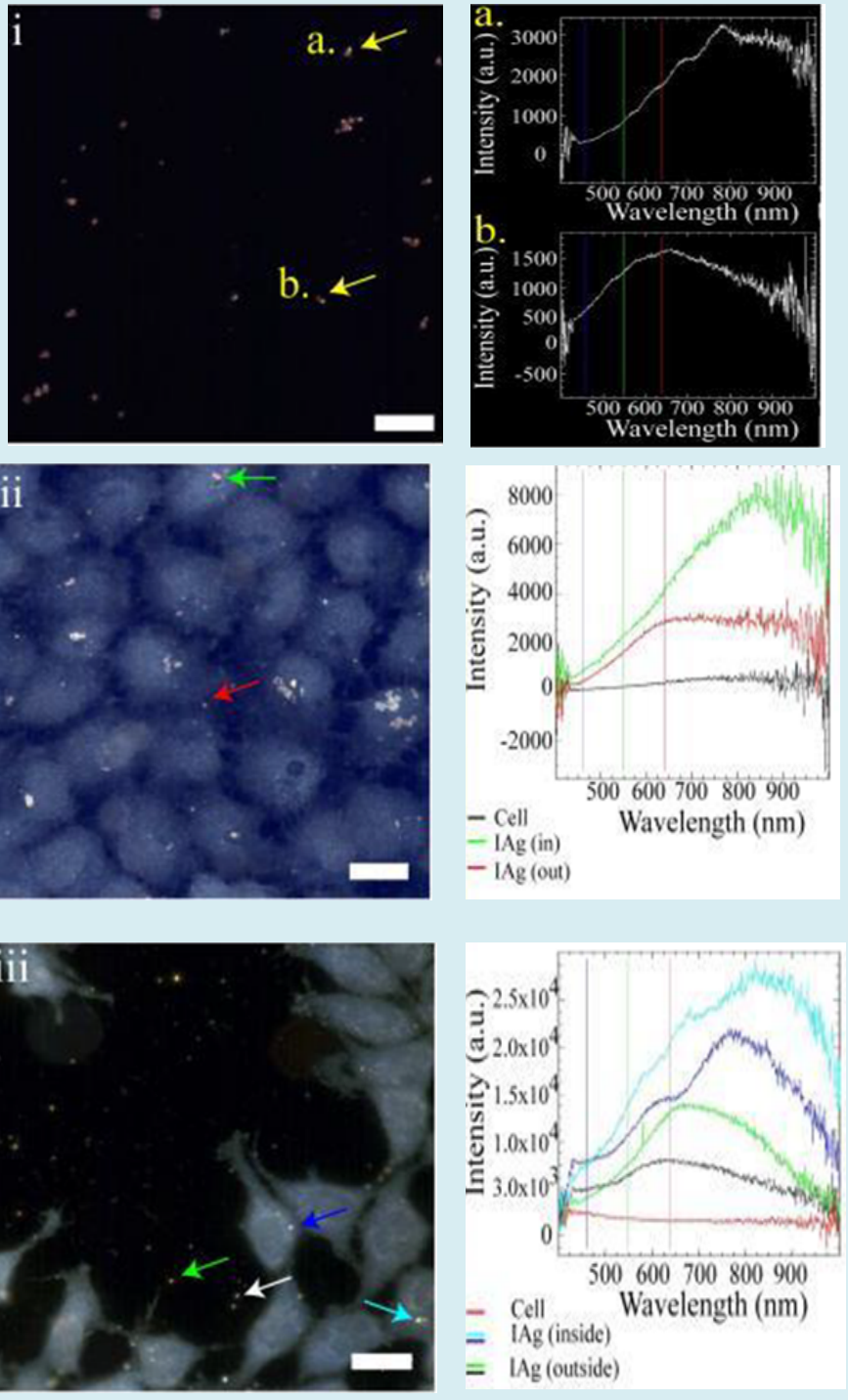

Figure 1: Hyperspectral images of Veli parpam (incinerated silver particles), alone (i), inside A549 cells (ii), and inside HCT116 cells (iii) (scale bare 20um) [20].

In addition to the localization of silver particles in cells, the image shows the spectral characteristics as well. The broad bands inj the spectrum belong to silver aggregates.

It was found that the uptake and localization of particles vary, (depending on the cell types and that, generally, there is a preferential uptake by the cancer cells, compared with normal, non-cancerous human cells. It was also found that the Swarna bhasma aggregates are able to enter the nucleus during the cellular division. The nuclear localization may suggest the utilization of Swarna bhasma particles as good carriers for drug delivery. Under the conditions of the experiments, it was found that, neither the gold bhasma, nor the silver parpam imparted toxicity to the human cells.

Conclusion

In this short review paper, we described, briefly, the herbo-metallic Indian traditional medicines, the Ayurvedic *bhasmas* and the Siddha *parpams* used for thousands of years to heal various diseases. The history, fabrication and properties of some of the most important metal bhasmas are shortly described. The paper is further focused on the recent contributions of our group to elucidate the pathways of entering of bhasmas and parpams in human cells and their localization in sub-cellular components. The traditional medicines studied in this work imparted no toxicity to human cells.

Bhasmas and *parpams* are, historically, prominent parts of the past and present of the Indian health care system. Their scientific validation, by using modern investigation methods, is a continuous process, bringing novel information in support of their therapeutic properties and lack of toxicity.

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