PREPARATION AND PHYSICO CHEMICAL CHARACTERIZATION OF THE BHASMAS OF ZINC AND CALCIUM AND THE INTERMEDIATES OBTAINED DURING THEIR SYNTHESIS

A THESIS SUBMITTED TO THE UNIVERSITY OF PUNE FOR THE DEGREE OF PHILOSOPHY (IN CHEMISTRY)

BY

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Certificate

Certified that the work incorporated in the thesis, "Preparation and physico chemical characterization of the bhasmas of zinc and calcium and the intermediates obtained during their synthesis" submitted by Mr. Mahesh Bhagwat, for the degree of Doctor of Philosophy, was carried out by the candidate under the supervision in the catalysis and inorganic chemistry division of National Chemical Laboratory, Pune, India. Materials obtained from other sources have been duly acknowledged in the thesis.

Dr. (Mrs. Veda Ramaswamy)

Research Guide

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Mahesh Bhagwat

Pune

“There is not even a single material on earth that does not have medicinal value”
- Charak

This forms the basis of Ayurved, the traditional medicinal system that is being practiced in India for centuries.

Contents

// INTRODUCTION //

1.1 INTRODUCTION TO AYURVED

1.1.1 Ayurved grows global

It has taken nearly 5000 years for the world to wake up to the possibilities of Ayurved. People across the globe are becoming increasingly disgruntled with the detrimental effects of the drug therapies of the modern medicine[1]. This has caused a necessity of an alternative medicinal system which is being felt throughout. On the medicinal front, homeopathy, Chinese herbal medicine, aromatherapy, acupuncture had the rapidly increasing demand as alternative routes of cure. Ayurved is one of them.

However, the globalization of Ayurved is not merely because of the perceived failures of the modern medicine, but also as a shift in the social patterns in the west where the younger generations became interested in alternative lifestyles and cultures. Ayurved preaches the art of living for humans so as to have a ‘healthy life’. It believes in following appropriate measures through mind control and yoga so as to make the body tough and capable of defending any external aggressions of illness and about diet control to prevent formation of disorders in the human system. People started exploring these various aspects related to the Ayurvedic literature ranging from its spiritual origin, yoga, diet control, mind control

amongst others. The Ayurvedic literature describes in detail ‘The way of life’ and does not limit itself merely to an encyclopedia of traditional medicines[2-3].

With the changing life styles in India, this concept of a healthy life took a backseat and the face of Ayurved that came forward slowly was limited to that of a medicinal system. From the role of the preacher of how to live a healthy life, Ayurved shifted to a position of a medicinal system, a system that describes in detail diagnosis of disorders in human body and the remedies for various illnesses.

The origin of Ayurved dates back to a few hundred centuries[4]. Since then, Ayurved is the traditional medicinal system that is practiced in India on the basis of the literature available in the form of the ancient texts compiled mainly by *Vaidya* Sushrut, *Vaidya* Charak and *Vaidya* VagBhat.

There is not even a single material on earth that does not have medicinal value. -Charak.

Charak describes the medicinal potential of the vast range of naturally occurring materials on earth as perceived by Ayurved[5]. India had a wide range of flaura and fauna spread across the country. The use of these herbs as valued medicines was explored in the early stages of practise of Ayurved. The herbal drugs theory developed and flourished to the extent of becoming household remedies for common health problems. This branch of Ayurved flourished even outside India due to the fact that these drugs could cure certain chronic diseases for which the modern medicine had no full proof remedy. Further these medicines had

recognition that they showed no side effects with the recommended doses.

Although the significance of various metals for human body was well understood by the founders of Ayurved, the actual preparation and use of the related drugs can be found in the Ayurved literature written only in the later years[6]. These materials were not used due to the fact that they have a mineral origin and it was probably not known so as to how these could be administered to humans. The use of minerals as drugs developed with the branch of Ayurved known as Rasashastra. Origin of Rasashastra has a mythological reference wherein the origin of Rasa (Rasa=mercury and the minerals derived from that) is said to be from God Shiva. The 'Rasashastra' describes in detail the significance of these minerals to human body, their origin, and the methods to convert them into the form of Bhasmas[6-9]. The use of the minerals for the preparation of Bhasmas, the details of their preparations and their use as drugs are well documented. It is only in the last 500 years that this branch of Ayurved is developed. However, these drugs could not be promoted outside India. In fact, they have been banned in most of the countries in Europe and elsewhere. Not only the patients but even the Ayurvedic practitioners showed apathy towards this branch of medicines. This was in spite of the fact that these medicines are being used routinely in India effectively, and in fact without any noticeable side effects with the recommended doses.

The Bhasmas of various metals including heavy metals like iron, copper, silver, lead, gold, zinc are routinely used in medication for oral administration to patients as well as external applications. The preparations of these Bhasmas involve

use of mercury and arsenic and their formulations. Most of these metals are regarded as toxic in various forms and hence their use as drugs raises many questions. Although Ayurved is the recognized traditional medicine in India, their use outside India is certainly questionable, particularly when they have stringent rules and regulations related to food and drugs. Thus, this branch of Ayurved remains dormant and finds applications only in India. Further, there is an increasing awareness amongst the pharmaceutical companies in India, practitioners and the end users in India about the requirement of a detailed scientific study of these Bhasmas.

1.2 INTRODUCTION TO BHASMAS

‘ Aushadi’ (medicine) forms a vital part of Ayurved, which lists the wide range of materials of medicinal values. A variety of herbs are commonly used in various formulations as drugs. While for some herbs, the whole plant body is used, the use is limited to a particular part of the plant (roots, leaves, flowers, stems,

fruits) for some others. The herbs are commonly used in the forms of *Kadhas* (decoction) and *Tailas* (oils) or *kalp* (paste). Herbs being living materials are known as 'Sendriya Padartha' (organic mater) i.e., materials that can be easily assimilated into human blood. On the other hand, the materials of mineral origin, which are also of siginificant importance to human body, however, cannot be used as such. They are broadly labeled as 'Nirendriya Padartha' (inorganic mater). They have to be converted into a form that can be readily assimilated into the body. Hence, these metals are converted into products known as 'Bhasmas'[7-9].

Bhasmas are inorganic in nature and are prepared by converting a metal into its compounds like carbonates, oxides, etc. Bhasmas of iron, calcium, copper, tin, silver, gold, lead and zinc are commonly used. Bhasmas are used for both, oral administration as well as external applications. Bhasmas are prepared according to the methods mentioned in the Ayurvedic texts and the overall preparation procedure involves converting the metal into a 'non-metallic' form by treating the metal with a range of animal and herbal products and some Ayurvedic mixtures. Various synthetic procedures for the preparation of the bhasmas are quoted in ancient Ayurvedic texts. The methods are broadly classified on the basis of the use of Kajjali and Hartal (Ayurvedic preparations containing mercury and arsenic, respectively) and herbal products in the preparation of the bhasma. According to Ayurved, the bhasma involving use of Kajjali in its preparation is supposed to have a greater medicinal value as compared to the bhasma prepared by other methods.

Preparation of the bhasma involves two major steps

1.2.1. Shodhan

A long procedure during which, the metal (used as the source for preparation of the bhasma) is subject to heat treatments followed by treatments with a number of herbal and animal products.

The ancient books describe this procedure a requisite before the process of Maaran. This process is said to be done to ‘purify the metal and bring it to a form which can be readily converted into the Bhasma’ during the Maaran process.

The Shodhan Process is of two types

1.2.1.1 The Samanya Shodhan:

It involves heating a metal piece over fire and dipping the hot metal piece into *Til* Oil (Sesame Oil). On cooling to room temperature, the metal piece is again subject to heat treatment and again poured into the oil. Each time fresh oil is used. This act is repeated seven times. After treatment with oil, this entire procedure is repeated by using Butter-milk, Cows milk, cows urine, soup of hulga and Kanji (an Ayurvedic preparation involving use of a variety of herbal products) in the given chronological order. After following this procedure, the product obtained is generally in the form of granules or a powder. The Samanya Shodhan is performed

on a variety of hard metals like, iron, copper, etc.

1.2.1.2. The Vishesh Shodhan

Vishesh Shodhan follows the Samanya Shodhan. The procedure is similar to that of the Samanya Shodhan. However, the raw materials (herbal or animal products) used during the Vishesh Shodhan differ from metal to metal. More than one type of materials have also been suggested that can be used in the process for a particular metal. The products obtained after the Samanya Shodhan are subject to heat and are again dipped into a herbal or animal product. The products used for the treatment are different for different metals. For soft metals like tin, zinc etc., which are low melting, the procedure of Samanya Shodhan can be skipped.

1.2.2. Maaran

This involves converting the ‘purified metal’ into the bhasma. It is performed so as to ‘kill’ all the metallic properties of the metal. Thus, a metal, which is essential for the human body, is converted from a ‘toxic’ or ‘harmful’ form into a form (Bhasma) that can be used as a drug.

Maaran is a procedure that follows the Shodhan. The product obtained after the Shodhan process is triturated with a number of herbal or animal products for hours. The herbal products to be used during triturating are different for different

metals.

Then the triturated products are mixed with other Ayurvedic preparations like kajjali, manasheel etc. that are inorganic in nature. The resultant mixture is then calcined in the furnace described in the Ayurvedic literature.

The product is checked for the various tests described in the literature to find whether the bhasma is finally obtained. If the tests are negative, the act of triturating and calcination is repeated till the bhasma is obtained.

1.3. MYSTERIES TO BE SOLVED

Although Bhasmas have been used as effective drugs for centuries without any noticeable side effects, certain factors related to their preparations have remained in disguise. Though the materials have been well established drugs, their current research is limited to the study based on Ayurvedic pointy of view. Very few reports[10-16] are available where attempts have been made to understand chemical properties of the material. These reports fail to throw light on the physico-chemical characteristics of these materials which could help in determining the mechanism involved in their preparation and the unsolved mysteries related to the preparation and use of these Bhasmas.

1.3.1. Synthesis of Bhasma involves very long procedures

The preparation of Bhasmas involves performing ‘Bhavanas’ on the metal with help of various herbal or animal products. These treatments involve trituration of the metal with these substances for hours together. These lengthy procedures last from a few days to a few months. This makes the preparation highly time consuming.

1.3.2. Not cost-effective

The preparation procedures are not only highly time consuming, but also involve use of raw materials or reactants like Kajjali that are rather expensive. The calcination treatment requires a lot of fuel. This does not make the over all preparation procedure cost effective.

1.3.3. They involve use of toxic metals like Hg, Pb, As.

Heavy metals including iron, lead, zinc, silver, gold and copper are commonly used for medication in the form of their Bhasmas. The preparation of their Bhasmas involves use of Ayurved formulations containing toxic metals like mercury and arsenic.

1.3.4. Availability of Raw materials.

The processes involve use of specific raw materials including certain fresh animal products. The herbal products are extensively required during the preparations. Availability of a particular desired species of these herbs is a concern. The possible use of alternatives to these materials has to be evaluated.

1.3.5. Lack of standardization.

The tests to check the formation of the Bhasma described in the literature are raw and designed with the limitations of developed science in that age. Further, some of these tests are highly individualistic. Some quality control measures need to be suggested that can be followed by the Ayurved pharmacies. The Bhasma samples of a particular metal available in the market through various pharmaceutical companies exhibit tremendous variety in apperance. There is a lack of standardization of the overall preparation procedure and the products.

1.3.6 No scientific explanations for many processes.

The preparation of the Bhasmas include specifications for products including use of particular raw-materials and reagents. The specifications are also with respect to some processes describing the time durations for some trituration treatments and calcinations procedures. Although the specifications help in

following a systematic methodology of preparation, there is no scientific explanation for these long procedures.

1.3.7. Reproducibility.

Although the procedures of preparations of the Bhasmas are well documented, the products obtained by following these procedures often differ in properties like appearance (color). Due to the lack of standardization of the procedures and quality control tools, it becomes difficult to understand the reasons for the non-reproducibility of the samples.

1.4. OBJECTIVE OF THIS WORK

The global herbal market is worth $ 120 billion a year and Ayurved alone represents 50 % of it. This is in spite of the fact that many Ayurvedic formulations involving the Bhasmas cannot be promoted in the global market. These Bhasmas and their formulation are being effectively used for various therapies and form popular drugs here. Thus, it becomes extremely important to study these materials in depth. It is important to understand the mechanism involved in their preparation and a thorough characterization of the Bhasmas is required as the first step towards establishing these materials as potential medicines to the world.

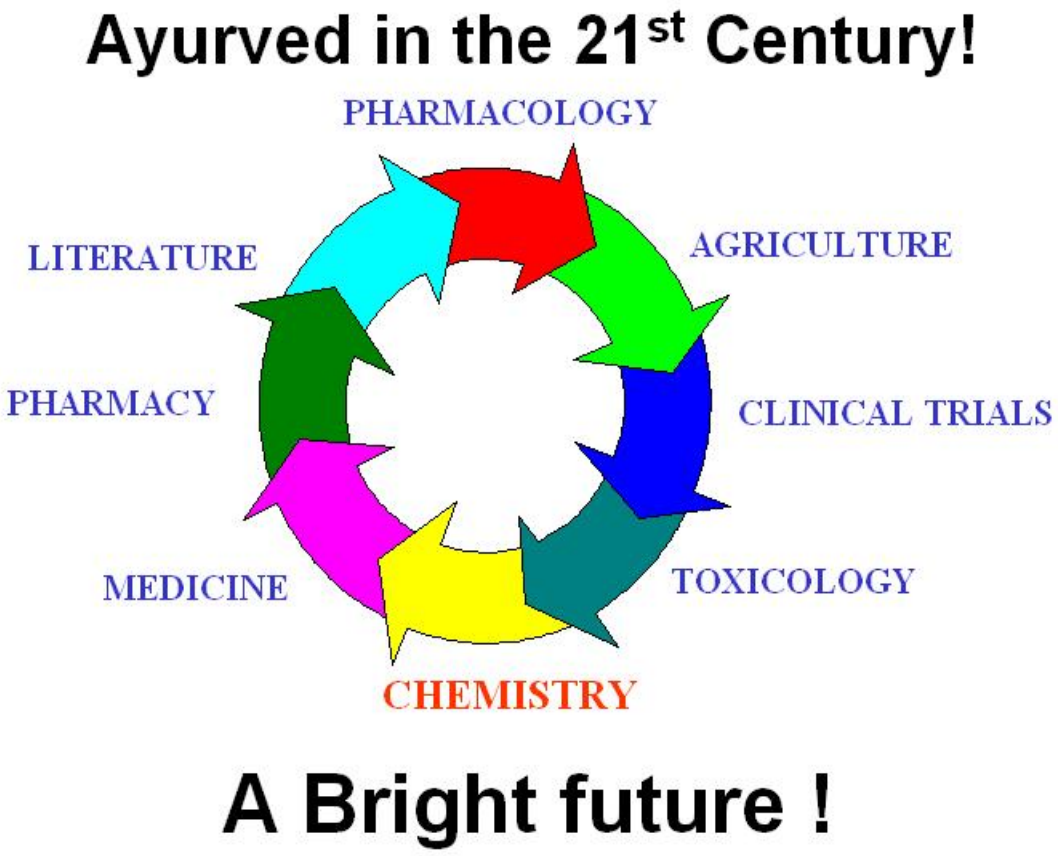
1.4.1 Taking the Bhasmas to the world

1.4.1.1. Ayurved-an ancient science is a gift of India to the world.

Till the 20th century, people were depending mainly on Ayurved & even home herbal remedies. In the 20th century, Ayurved surrendered the dominance to allopathy and was shifted to the status of alternative medicine. Allopathy flourished and prospered due to contribution from pathology, physiology, chemistry and chemical engineering. Ayurved on the other hand remained stagnant[17].

One of the reasons for the apathy towards Ayurved was that Ayurved

remained an island in ocean of knowledge. Researchers and practitioners of Ayurved remained indifferent towards advances in other sciences. Now time has come for researchers in Ayurved to open hands for researchers from Botany, Chemistry and other sciences for collaborative research. A multidisciplinary approach is required to know these valuable materials in depth[18]. Chemistry plays a vital role to understand the intricacies involved in the synthesis of these Bhasmas and their utility. It has become very important to carry out a systematic and scientific study of these medicines that would help in promoting the rich heritage of Ayurvedic medicines all over India and elsewhere in the world.



We have taken up the research work involving the preparation and characterization of these Bhasmas. The Bhasmas of zinc, calcium and tin have been prepared strictly as per the methods mentioned in the Ayurvedic text 'Rasaratnasanucchaya'.

The Bhasmas have been analyzed by various analytical techniques to determine their chemical compositions. The raw materials, intermediates obtained during the synthesis and the final products have been characterized by various instrumental techniques including powder X-ray diffraction, Infrared spectroscopy, Thermo gravimetric analysis and differential thermal analysis to understand the

probable mechanism of formation of the Bhasmas.

The Bhasma samples marketed by various pharmaceutical companies have been analyzed to understand the differences in their physical and chemical properties. Simple quality control tests have been suggested for the pharmaceuticals, which could be the first step towards the Standardization of the Bhasmas.

1.5. MATERIALS AND METHODS

The Bhasmas of Zn, Sn and Ca were prepared strictly as per the methods mentioned in the Ayurvedic text 'Rasaratnnasamucchaya' without any alterations in the procedures[7-9]. The raw materials, including the metal sources, various herbal products and other reactants required during the preparation of the Bhasmas were procured from the Ayurvedic Aushadhi shops and used as per the guidelines in the text. Although pure metals and chemicals available in the market could have been used, the materials have been procured from the places, which supply the same to pharmaceutical companies preparing these Ayurvedic medicines. Various raw materials used in the preparations have been listed in Table 1 along with their Ayurvedic names (wherever necessary).

Jasad Bhasma	Jasad (zinc metal sheets), cow's milk, lemon juice (fresh), Aloe vera gel (fresh), mercury, sulfur, fresh leaves of the plants of Neem and Aghara, lime water.
Vanga Bhasma	Vanga (tin metal ribbons), leaves of Nirgundi (fresh), lime water, ova, haldi, jeera, pimpli churna, chinch churna, lemon juice (fresh), Aloe vera gel (fresh).
Samudrapanchak	Kapardika, Shankha, Shimpla, Paval, rose water, lemon juice (fresh), Aloe vera gel (fresh).

The apparatus required during the preparation of the Bhasmas was also as per the directives in the Ayurvedic text. The apparatus (vessels, mortar and pestles etc.) were procured with the required specifications. The Maaran procedure forms an important part during the synthesis of the Bhasmas. The procedure mainly involves various calcinations steps. Various fuel including Cow dung cakes and coal, to be used during the Maarn procedures are described in the text along with the quantity to be used. Also, crucibles to be used (earthen pots) and the nature and the size of the furnace have also been described in detail. The same has been followed during the preparation of the Bhasmas.

The samples obtained during the various steps in the Maaran procedures were subject to the various tests described in the literature to check weather the sample obtained is the Bhasma or still an intermediate. Though these physical and chemical tests are sufficient to tell whether the bhasma has been formed or not, they provide very little information about the composition and the structural properties of the bhasma. With the advent of modern technology it thus becomes necessary to put the synthetic procedure under scrutiny using modern instruments. Also, few reports about the metal analysis and the structural characterization of the bhasmas are available. Thus, it becomes necessary to characterize the bhasma and the various intermediates obtained during the preparation so as to get an insight into the mechanism involved in the process of synthesis of the bhasma.

The Bhasmas of Zn, Sn and Ca and the various intermediates obtained during their preparations were analyzed to determine their chemical composition and characterized using the various techniques described below.

1.6. CHARACTERIZATION TECHNIQUES

1.6.1. Chemical analysis

The chemical analysis of the Bhasma sample is extremely important to determine the chemical composition of the material to be used as the drug. The raw materials were analyzed to determine the probable sources of the various elements in the Bhasma samples. The intermediates obtained during the preparation were analyzed to determine the changes taking place in the sample with the various treatment procedures.

The samples were analyzed qualitatively by performing spot tests. Zn in Jasad Bhasma was estimated by volumetric analysis by titrating with EDTA using Eriochrome black T indicator and ammonia-ammonium chloride buffer[19]. Elements with lower concentrations were analyzed by EDAX to determine the relative atomic percentages of the major and minor elements on a JOEL-JSM-5200 Scanning Electron Microscope with Energy Dispersive X-ray analysis facility (Kevex). Metal atoms with concentrations upto 1 ppm were estimated by using Atomic Absorption spectroscopy by analyzing their acidic solutions. The metals in concentrations less than 1 ppm were estimated by inductively coupled plasma on Perkin-Elmer ICP-400 elemental analyzer.

1.6.2. Particle size distribution

The Ayurvedic texts give a lot of importance to the particle size of the

Bhasmas and believe that their efficacy as a drug is strongly related to its small particle size. It is also mentioned in the Ayurvedic texts to repeat the steps in the Maaran procedures till the sample with small particle size is obtained. The Bhasma samples and the intermediates were dispersed in distilled water under constant stirring. Ultrasonication was used for effective dispersion of the powders. These dispersions were analyzed using a particle size analyzer (Fritsch Particle Sizer Analysette 22 model) to determine the particle size distribution of the bhasma samples.

1.6.3. Powder XRD studies

After determining the chemical compositions of the samples, it was important to determine the crystalline form of these materials. Hence, the Bhasma samples were characterized by the powder X-ray diffraction technique to obtain the structural information. The intermediates were also characterized by powder XRD to obtain the structural changes taking place in the samples during the Maaran procedures.

All the samples were scanned on Rigaku D-Max III VC powder X-ray diffractometer using Cu Kα radiation (λ=1.5406). The diffractometer was equipped with a Ni filter, a curved graphite monochromator and a NaI scintillation detector. Silicon was used as the standard to calibrate the instrument for the correction of 'd' spacings'. The samples were scanned in the 2θ range 20-100 ° for a period of 10

sec per step in the step scan mode. The phase composition was determined using the JCPDS Library and the search-match program available with the Rigaku machine. The lattice parameters were obtained by refining the peak position values by the least square fitting procedure. The Crystallite size (L) of the samples was determined using the Scherrer equation, L = k λ

constant (k = λ Å (Cu K

α

20.

1.6.4. Infra red studies

The Fourier transform infra-red spectra of the samples were recorded on a Shimadzu 8300 spectrophotometer. The samples were dried in an oven at 110 °C to remove moisture. The finely powdered samples were mixed with potassium bromide (I.R. grade) to form a homogeneous mixture. The sample concentration in the mixture was roughly 1% with respect to KBr. The mixture was then converted into pellets, which were analyzed on the spectrophotometer from 500 to 3600 cm^{-1} region.

1.6.5. Thermal Analysis

The raw materials and the intermediate samples were analyzed by the thermogravimetric and differential thermogravimetric analysis to study their decomposition behavior. The samples were also analyzed by the differential thermal analysis technique to study the thermal changes in the sample as a function of temperature. The analysis was done on a Setaram thermal analyser (SETARAM). The thermograms were recorded from room temperature to 1273 K at the heating rate of 10 K min^{-1} in air atmosphere.

1.6.6. pH measurements

While preparations of some Bhasmas involved reactions in acidic medium, it was necessary to monitor the change in the pH of the medium as the reaction progressed. The pH of the samples was determined using a pHmeter (Labindia) using a gel filled combined electrode.

Kapardikas were immersed in lemon juice and the pH of the mixture was determined after a regular interval of one hour for four hours to study the reaction between the Kapardikas and the lemon juice. Also, the samples obtained after each calcination process were suspended in distilled water and the pH of the water was measured in a similar way to determine the nature of the sample obtained.

Similarly, the pH changes were monitored during the preparations involving Shankha Shimpla and Paval.

1.6.7. Carbon analysis

The Maaran procedure involves trituration of the metal with a variety of herbal products followed by calcination of the mixture. The calcination of the organic mater would result in the combustion. However, in case there is a partial combustion, the organic mater would decompose leaving behind some carbonaceous mater. Presence of the carbineous mater in the sameple was determined using the microanalysis technique on a carbon detector CARLO ERBA EA-1108 analyzer.

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// JASAD BHASMA //

2.1 INTRODUCTION TO JASAD BHASMA

Jasad Bhasma is the Bhasma of zinc. The Jasad Bhasma is an important drug used in curing various disorders caused due to diabetes. It is also used for treating various eye ailments and in muscle building[1]. The drug is used for both, oral administration as well as for external applications. Jasad Bhasma is also used in combination of Vanga Bhasma (Bhasma of tin) and Naag Bhasma (Bhasma of lead), popularly known as Trivanga Bhasma[2]. The combination of the three Bhasmas is used for disorders related to reproductive system. Jasad Bhasma is administered to patients along with a vehicle like Ghee prepared using Cows milk.

Jasad Bhasma is prepared by using various methods quoted in the Ayurvedic texts describing the methods for the preparation of the Bhasmas[3]. The methods differ in the use of raw materials during the Shodhan and the Maaran procedures. The methods of preparation are broadly classified on the basis of the use of various herbal products, Haartal and Kajjali (Ayurvedic formulations involving use of arsenic and mercury, respectively) during the Maaran step[4]. It is believed that the Bhasma prepared using Kajjali has better medicinal properties as compared to the Bhasma obtained from other methods[4]. The texts describe use of Samanya Shodhan for purification of Jasad when zinc ore was used in older times as the source of zinc. When zinc metal itself is used as the source of Jasad, the

process of Samanya shodhan is not performed on the sample. Zinc being a soft metal, is subject only to Vishesh Shodhan[4].

The texts describe in detail various tests that should be performed on the samples obtained during the Maaran procedures to check whether the Bhasma has been formed or not[2-4]. While some tests to check the formation of a Bhasma are common for all the Bhasmas, some tests are specific for a particular Bhasma. The tests to check the formation of Jasad Bhasma are given in Table 1.

Table 1

Tests for the formation of Jasad bhasma.

Test	Observation
Sample +Water	Sample floats on water.
Sample +Ag metal piece, ignite	No gain in weight of Ag metal piece.
Sample +lemon juice	No effervescence.

The first test confirms the small particle size of the sample. The second test confirms the absence of metallic form of Jasad in the sample and that the process of formation of the Bhasma is irreversible. The third test confirms that the sample is not an intermediate but that the Bhasma has been obtained.

In the present work I have prepared Jasad Bhasma samples according to methods mentioned in the Ayurvedic text Rasaratnasanucchaya[2]. The Bhasmas have been prepared using different methods in which Kajjali and leaves of Aghara

and Neem plants are used during the Maaran procedure. Kajjali forms an important Ayurvedic formulation. It is used as a medicine as such and also in various formulations used for external applications. Kajjali is an important reactant used in the preparation of Bhasmas.

2.2. STUDY ON JASAD BHASMA PREPARED USING KAJJALI

2.2.1. Introduction to Kajjali

Kajjali is an Ayurvedic formulation employing the use of mercury in combination with sulfur. Mercury and Sulfur are treated separately with various animal and plant products and then mixed together. They are mixed together in various ratios based on the application of the Kajjali. Trituration of these treated products results in a fine black powder known as Kajjali. Kajjali is used in medication as such and also as an important ingredient in the formation of the Bhasmas.

2.2.2. Preparation of Kajjali

2.2.2.1 Shodhan of Mercury

Mercury was treated with an equal quantity of garlic paste and triturated for

56 hours (8h x 7days). After tritutaion for 56 hours, a blackish paste was obtained and mercury droplets were no more visible. This resultant mixture was than treated with hot water and mercury (M-1) was separated out from the mixture.

2.2.2.2 Shodhan of Sulfur

In an iron vessel, ghee obtained from cow's milk was heated till it just melted. Sulfur powder was added to this hot ghee with continuous stirring. It appeared that sulfur melted to form a dilute paste with the molten ghee. The mixture was heated to ensure that the mixture remains in a molten state. This hot mixture was poured into cold milk. On cooling, the milk was filtered. The residual sulfur cake was crushed to get a fine powder (S-1).

The mercury and sulfur obtained from the respective Shodhan processes (M-1 and S-1 respectively) were mixed together and triturated for hours. It was observed that the two products mixed well to obtain a black powder with luster. As the trituration was continued, luster of the black powder decreased. After about 16 hours of trituration, a fine black powder without any luster was obtained. This product is Kajjali (K-1).

2.2.3. Preparation of Jasad Bhasma using Kajjali

2.2.3.1 Shodhan of Jasad

In this process, the zinc metal (used as the source of Jasad) is treated with various herbal or animal products so as to convert it into a form, which can be readily converted into the bhasma. Zinc metal was melted in an iron vessel and the molten metal was poured into cow's milk. On cooling, the mixture was filtered and the residue was again heated and poured into cow's milk. This treatment was repeated 21 times, each time fresh milk was used. It was observed that the zinc metal sheet was converted into granules. More and more amount of zinc was getting converted into fine granules with the continued treatments. After the 21 milk treatments, zinc was converted into a gray powder (Z-1).

2.2.3.2 Maaran of Jasad

During Maaran, the product obtained after the Shodhan process is treated with various herbal and inorganic products and calcined in the furnace. The process of Maaran is repeated till the final product, bhasma is obtained.

Zinc obtained after the Shodhan process (Z-1) was mixed with an equal quantity of Kajjali. This mixture was triturated to obtain a fine powder. To this

powder was added equal quantity of lemon juice and triturated for 8 hours followed by addition of aloe vera gel. The mixture was again triturated for 8 hours and to get a fine black powder. The powder was then moulded manually to obtain small tablets that were transferred to an earthen crucible covered with an earthen lid. The earthen crucible was calcined in an traditional furnace shown in Fig.1. The furnace was filled with cowdung cakes that gave the desired temperature when fired. The temperature at the furnace reached around 1100 °C. The furnace was allowed to cool naturally (about a days time).

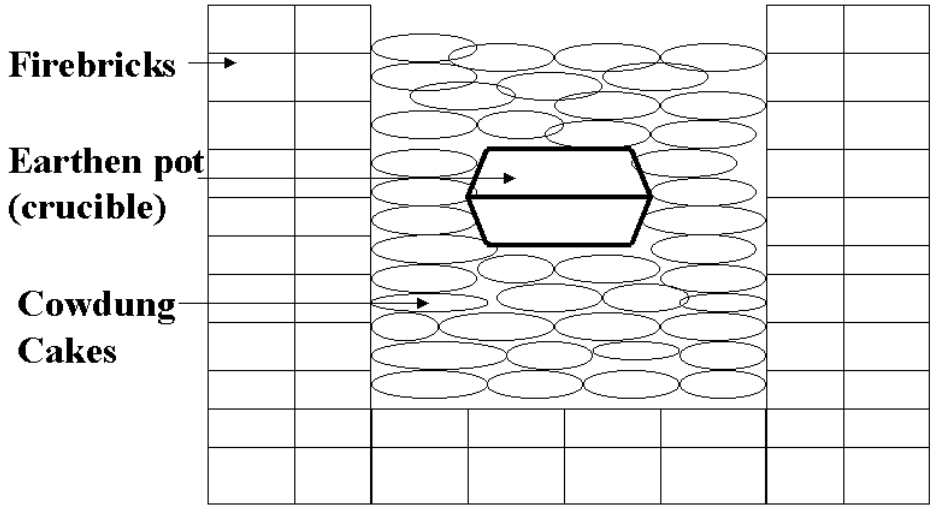


Fig. 1 Traditional furnace for a 'Gajaputa' as described in Ayurvedic literature

The sample obtained after the calcination procedure (JB-1) was subjected to the tests mentioned in Ayurvedic literature to check whether the final product, Jasad bhasma has been formed or not. If not, then the sample was triturated again with lemon juice followed by aloe vera gel treatment and calcined in the same

manner. Each time a fresh crucible was used. The procedure was repeated till the sample showed all the tests of the bhasma (as mentioned in Table 1) positive. The intermediates obtained after each calcination process and the final product (JB-4) were isolated separately and designated as, JB-1, JB-2, JB-3and JB-4 respectively.

Table 2

Sample details

Sample code	Description
M-1	Mercury obtained after garlic paste treatment
S-1	Sulfur obtained after ghee treatment
K-1	Kajjali obtained by mixing M-1 and S-1
Z-1	Granules obtained after 21 milk treatments
JB-1	Sample obtained after first calcination
JB-2	Sample obtained after second calcination
JB-3	Sample obtained after third calcination
JB-4	Sample obtained after fourth calcination

2.2.4. Characterization of Jasad Bhasma prepared using Kajjali

2.2.4.1. Chemical analysis

The spot tests on the intermediates and the final product revealed the

presence of Zn, Fe, Ca, Mg, Sn, Pb, Mn, Co, Cr and Cu in the samples. The presence of metals other than zinc in the Jasad bhasma can be explained by the results of the spot tests performed on the raw materials (Table 3). The source of Ca, Mg, could be milk used during the process of Shodhan. Aloe vera could be the source of Ca, Mg and Mn in the samples. Presence of iron is due to the iron container used for synthesis. The raw material zinc contains Sn, Pb, Co and Cu as impurities.

Table 3

Sources of various metals in the product Jasad bhasma

Metals	Probable source
Ca, Mg	Milk, Aloe vera
Sn, Pb, Co, Cu	Zinc metal used as the source of zinc
Mn	Aloe vera
Fe	Iron container in which zinc was melted

The complexometric analysis of the final product Jasad bhasma shows that zinc is present as the major component (79.83%). Table 4 gives the elemental analysis of the intermediates and the final product determined by EDAX. EDAX results of the bhasma show that it contains zinc as the major element. Ca, Mg, Sn and Pb are present in low concentrations, while Fe is 1.69 %. Trace metal analysis

of the bhasma using ICP shows that concentrations of Mn and Cu are 10.6 and 2 ppm respectively, whereas concentrations of Co and Cu are less than 0.5 ppm. The intermediates JB-1, JB-2 and JB-3 show presence of free sulfur. The chemical analysis of these samples shows that they have free sulfur as well as sulfide anion. Sample JB-3 shows presence of sulfate anion also. The final product JB-4 does not contain free sulfur, sulfide and sulfate anions. The source of sulfur in the intermediates is the Kajjali used for the synthesis of the bhasma.

EDAX results show that the percentage of sulfur is decreasing with successive heat treatments. The intermediates JB-1, JB-2, JB-3 and the final product JB-4 do not show the presence of *mercury* even at ppb level (not detected by ICP). This indicates that all the mercury from the Kajjali was vaporized during the first calcination process itself. The final product Jasad bhasma does not contain mercury *even at trace levels.*

Table 4

Elemental analysis data obtained by EDAX

Elements	JB-1	JB-2	JB-3	JB-4
Zn	72.05	81.95	94.07	95.08
S	24.52	15.00	1.25	0.00
Ca	0.35	1.05	1.75	1.82
Fe	0.97	0.84	1.64	1.69
Mg	0.00	0.00	0.92	1.00
Sn	0.10	0.15	0.25	0.27
Pb	2.00	1.01	0.12	0.14

(Atomic percentages normalized to 100%)

2.2.4.2 Particle size distribution

Fig. 2 gives the particle size distribution patterns of the intermediates and the final product. The data for the particle size distribution of the samples is given in Table 5. The particle size distribution of JB-1 is quite broad; the particles are distributed between 5 and 50 µm. However, about 90% particles have a size less than 34 µm. The particle size distribution of JB-2 is similar to that of JB-1. The distribution of JB-3 is divided into two regions. 96% particles are distributed in the region of 0.1 to 10 µm where as 4% particles are distributed in the region of 30 to 100 µm. Thus, the size of some of the particles seems to have increased while there is a drastic decrease in the size of a majority of the particles. In JB-4 the particle size has reduced. 90% particles have a size less than 10 µm. The distribution is

mainly in the range of 5 to 10 μm and thus has become narrower. It appears that the particle size of the sample decreases from JB-1 to JB-4 with successive aloe vera gel treatment followed by lemon juice treatment and the calcination process. Also the particle size distribution becomes narrower with the treatments.

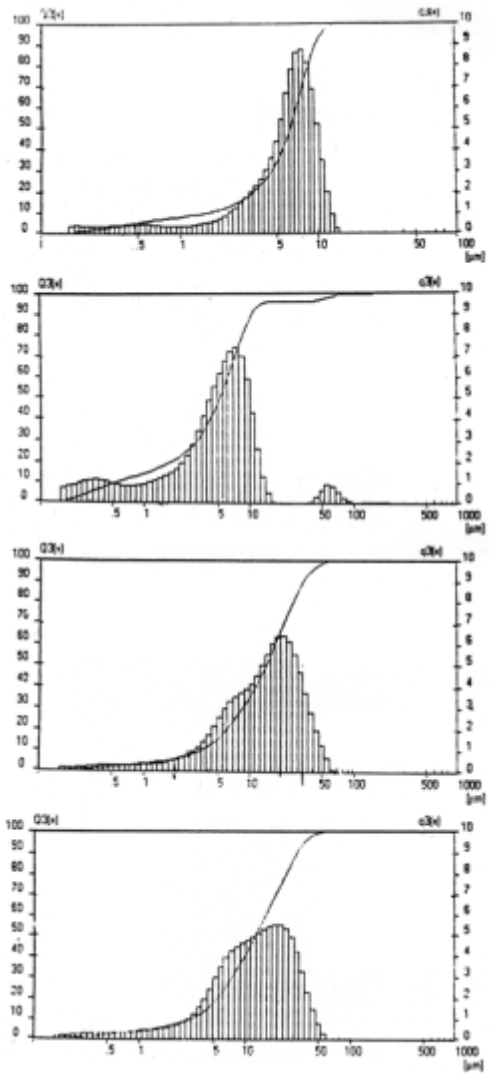


Fig. 2 The particle size distribution patterns of the intermediates and the final product.

(a) JB-1, (b) JB-2, (c) JB-3, (d) JB-4

Table 5

Particle size distribution of different samples

% of particles	Particle size less than (microns)			
	JB-1	JB-2	JB-3	JB-4
10	3	3	1	2
20	6	5	2	3
30	9	8	3	5
40	12	10	4	5
50	15	13	5	6
60	18	16	6	7
70	22	20	7	8
80	26	24	9	8
90	33	31	11	10
99	52	48	70	12

2.2.4.3. Powder XRD studies

Fig. 3 shows the powder XRD patterns of the raw materials Z-1 and K-1 used for the synthesis of Jasad bhasma. The XRD pattern of Z-1 indicates that it is a mixture of hexagonal zinc metal (88%) and hexagonal zinc oxide (12%). Thus during the 21 milk treatments on the metallic zinc, the metal was partly converted

into zinc oxide. However, the major change was observed in the physiological form of the sample. The metallic sheets of zinc were converted into a fine granular form, which could now be easily mixed with the Kajjali.

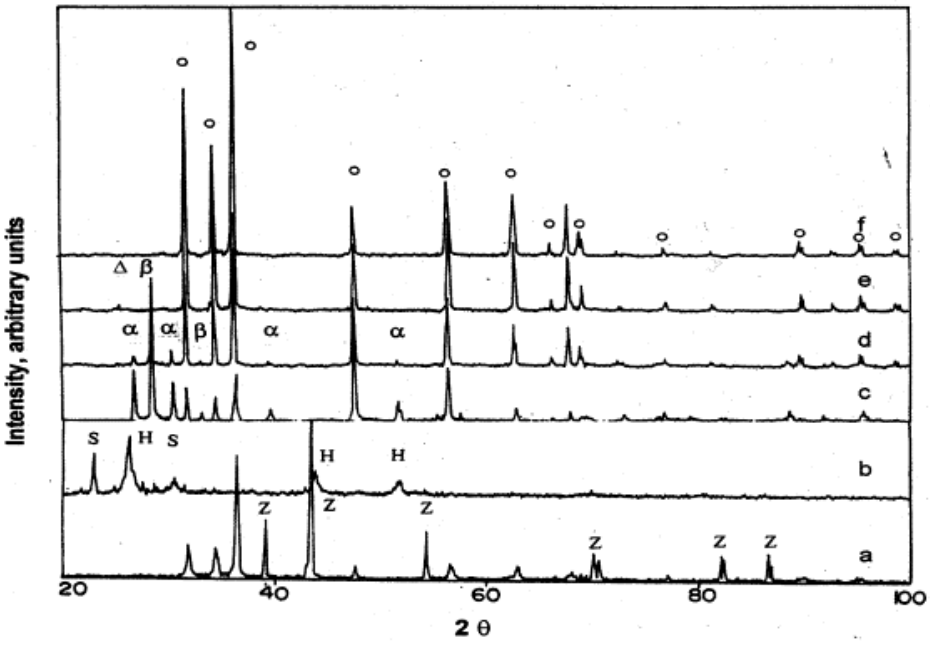


Fig. 3 Multiple plot of the Powder XRD patterns of the intermediates and the final product. (a) Z-1, (b) K-1, (c) JB-1, (d) JB-2, (e) JB-3, (f) JB-4 (o) ZnO, (α) hexagonal ZnS, (β) cubic ZnS, (Δ) $ZnSO_4$, (H) HgS, (S) Sulfur, (Z) Zinc metal

The XRD pattern of Kajjali shows that it is a mixture of hexagonal mercuric sulfide and free sulfur. This indicates that when the samples M-1 and S-1 are mixed together to prepare Kajjali, mercury reacts with sulfur to form mercuric sulfide in the hexagonal form. The XRD pattern of JB-1 shows that it is a mixture of three phases containing 53% cubic zinc sulfide, 21% hexagonal zinc sulfide and 26%

hexagonal zinc oxide (Table 6).

Table 6

Relative phase composition (%) of the phases present in the intermediates and the final product obtained from the Powder XRD patterns

Sample	ZnS (Cubic)	ZnS (Hexagonal)	ZnO (Hexagonal)
JB-1	53	21	26
JB-2	4	23	73
JB-3	-	-	99.4
JB-4	-	-	100.00

Although JB-2 is also a mixture of these three phases, yet there is a predominant increase in the concentration of hexagonal zinc oxide (73%), at the expense of cubic zinc sulfide (4%). The concentration of hexagonal zinc sulfide does not change much (23%). The sample JB-3 that is obtained after the third calcination contains predominantly only one phase i.e. hexagonal zinc oxide (99.4%). The zinc sulfide in the first two calcinations has gone from a higher symmetric cubic zinc sulfide to lower symmetric hexagonal structure and is finally converted into hexagonal zinc oxide. The chemical analysis of this sample shows the presence of sulfate anion. The powder XRD pattern of this sample shows the

presence of three peaks at d = 3.494Å, d = 2.636 Å and d = 2.543Å with intensities 1.7%, 2% and 1%, respectively, which could be due to the presence of orthorhombic zinc sulfate. These peaks are not present in the XRD patterns of any other samples. The EDAX results of the intermediates JB-1, JB-2 and JB-3 and their chemical analysis show presence of free sulfur. Kajjali could be the only source of sulfur, which is found in these samples. The XRD pattern of Kajjali shows presence of free sulfur. However, the XRD patterns of JB-1, JB-2 and JB-3 do not show the presence of free sulfur. This suggests that the crystallite size of the sulfur present in the samples is very small and is XRD amorphous. This suggests the conversion of crystalline sulfur from Kajjali into XRD amorphous sulfur during various treatments followed by calcination processes. The XRD pattern of JB-4 matches exclusively with hexagonal zinc oxide. The XRD patterns and the chemical analysis of this sample do not show the presence of free sulfur, sulfide or sulfate anions. This suggest the complete conversion of metallic zinc into hexagonal zinc oxide during the number of calcination processes and that the final product, Jasad bhasma has been formed.

Table 7 provides the cell dimensions data of the different phases present in the intermediates and the final product. The data indicate an expansion of the lattice of hexagonal zinc oxide in the final product JB-4. This could be due to the presence of calcium, iron and lead in the zinc oxide lattices.

Table 7

Lattice parameters of the different phases present in the intermediates and the final product obtained by Powder XRD

Phase present	Lattice parameters	JB-1	JB-2	JB-3	JB-4	ASTM reference data
ZnS	a (Å)	5.410	5.387			5.406
Cubic	V($Å^3$)	158.33	156.36			157.99
ZnS	a (Å)	3.824	3.822			3.820
Hexagonal	c (Å)	6.264	6.241			6.260
	V ($Å^3$)	70.304	78.953			79.103
ZnO	a (Å)	3.249	3.249	3.250	3.251	3.249
Hexagonal	c (Å)	5.188	5.202	5.207	5.209	5.205
	V ($Å^3$)	47.427	47.541	47.631	47.686	47.581

The chemical analysis, EDAX results, XRD patterns and the relative phase

composition analysis of the intermediates and the final product suggest the following probable mechanism involved in the synthesis of Jasad bhasma. The granules containing metallic zinc and zinc oxide when treated with Kajjali (a mixture of mercuric sulfide and sulfur), in presence of aloe vera gel and lemon juice are first converted into hexagonal zinc sulfide. Then with successive heat treatments, cubic zinc sulfide is first converted into hexagonal zinc sulfide and finally into hexagonal zinc oxide.

2.2. STUDY ON JASAD BHASMA PREPARED USING AGHARA AND NEEM LEAVES

2.3.1. Preparation of Jasad bhasma using Neem leaves

2.3.1.1 Shodhan

During Shodhan, zinc metal was melted in an iron vessel. The molten metal was poured into limewater and filtered. The residue was again heated to melt and the molten metal was again poured into limewater. This was repeated 8 times, each time fresh limewater was used. The product obtained after the Shodhan process was in the form of metallic mass, which could be ground into a powder easily. The molten mass had lost the metallic luster.

2.3.1.2. Maaran

During Maaran, Jasad obtained after the Shodhan procedure was again melted in an iron container. In this process all the heat treatments were carried out by using burning coals as the heat supplier. To the melt was added a fresh Neem leaf and the melt was continuously stirred with an iron rod. After the leaf burnt completely, another Neem leaf was added. This addition of Neem leaves was continued till the metal was converted into yellow colored fine powder (JB-N1). Then this powder was heated for another 4 hours over the burning coals to obtain the powder with a reddish tinge (JB-N2). The resultant sample was mixed with aloevera juice and triturated for 8 hours. This mixture was then triturated with lemon juice for 8 hours to obtain a finer powder. The mixture was sealed in an earthen crucible and calcined in the furnace as described in the above procedure. At each stage, the sample was tested for the formation of Jasad Bhasma as per the tests described in Table 1.

2.3.2. Preparation of Jasad bhasma using Aghara leaves

The bhasma using Aghara leaves was synthesized exactly in the same manner, as the bhasma using Neem leaves was prepared. However, in this method Aghara leaves were used instead of Neem leaves to obtain a yellow colored fine powder (JB-A1), which was heated to obtain a reddish powder (JB-A2). The mixture was then calcined to obtain the bhasma (JB-A).

2.3.3. CHARACTERIZATION OF JASAD BHASMA

2.3.3.1 Chemical Analysis

The Jasad bhasma samples (JB-N and JB-A) were prepared using the Aghara and the Neem leaves as per the method mentioned in the Ayurvedic text, Rasaratnasamucchhaya. The chemical analysis of the Bhasma samples indicated the presence of Zn, Sn, Pb, Fe, Ca, K, Mg Cu, Mn and Na in the samples. The raw materials used during the preparation were also analyzed to find out the probable source of the metals in the bhasma. The concentrations of the various metals in the bhasma samples are given in Table 8 along with their probable sources. The data indicates zinc as the only metal in major concentration, 93.72 and 93.84% for the JB-N and JB-A samples, respectively. The samples contain Sn, Pb, Fe and Ca in lower concentrations while Ca, K, Mg Cu, Mn and Na were detected in trace quantities. Fe and Ca were in higher concentrations in both the Bhasma samples as compared to the other metals. Presence of iron could be due to the iron vessel in which the heat treatments were carried out, while the source of Ca could be the lime

water used during the Shodhan processes. The intermediate samples JB-N1 and JB-2 showed presence of sulfide ion (S^{--}) in small amounts during the chemical tests.

Table 8

Chemical composition obtained from chemical analysis (Metal concentrations normalized to 100 %)

Metal	**JB-N**	**JB-A**	**Probable source of the metal**
Zn	93.72 (%)	93.84 (%)	Used as the raw material
Sn	0.43 (%)	0.38 (%)	Zinc metal used as the source of zinc
Pb	0.28 (%)	0.31 (%)	Zinc metal used as the source of zinc
Fe	1.76 (%)	1.82 (%)	Iron container in which bhasma was prepared
Ca	1.98 (%)	2.17 (%)	Lime water used during the Shodhan process
Mg	0.96 (%)	0.75 (%)	Aloe Vera and lemon juice
K	0.49 (%)	0.31 (%)	Aloe Vera and lemon juice
Na	0.38 (%)	0.42 (%)	Aloe Vera and lemon juice

Mn, Cu	< 10 ppm	< 10 ppm	Iron container in which bhasma was prepared

2.3.3.2. Powder XRD study

The powder X-ray diffraction patterns of the intermediate samples and the final products of the bhasma samples prepared using Aghara and Neem leaves are given in Fig. 4 (a) and (b), respectively. The powder XRD patterns of the samples prepared from Neem leaves suggest that the zinc metal is being converted to zinc oxide with hexagonal structure during the Maaran procedure. The observation is same for the samples prepared using the Aghara leaves. All the samples, JB-N1, JB-N2, JB-A1 and JB-A2 show a mixture of both, zinc metal as well as the zinc oxide phase. It can be observed that the relative concentration of zinc metal in the samples decreases with the heat treatment. The sharp peaks in the powder patterns indicate a high crystallinity of the zinc oxide phase in the sample. The lattice parameters for the zinc oxide phase in all the samples are given in Table 9. The data indicate that the unit cell dimensions ‘a’ and ‘c’ and hence the unit cell volume ‘V’ increase with the treatments during the Maaran process. This indicates the incorporation of the metal ions into the zinc oxide lattice causing an increase in the

unit cell dimensions.

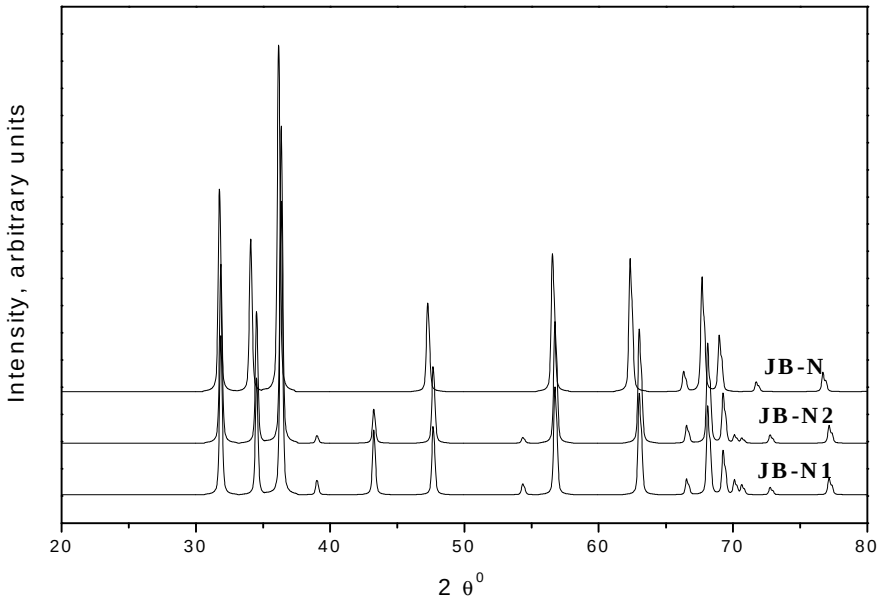


Fig. 4(a) The powder X-ray diffraction patterns of the intermediate samples and the final products of the bhasma samples prepared using Neem leaves

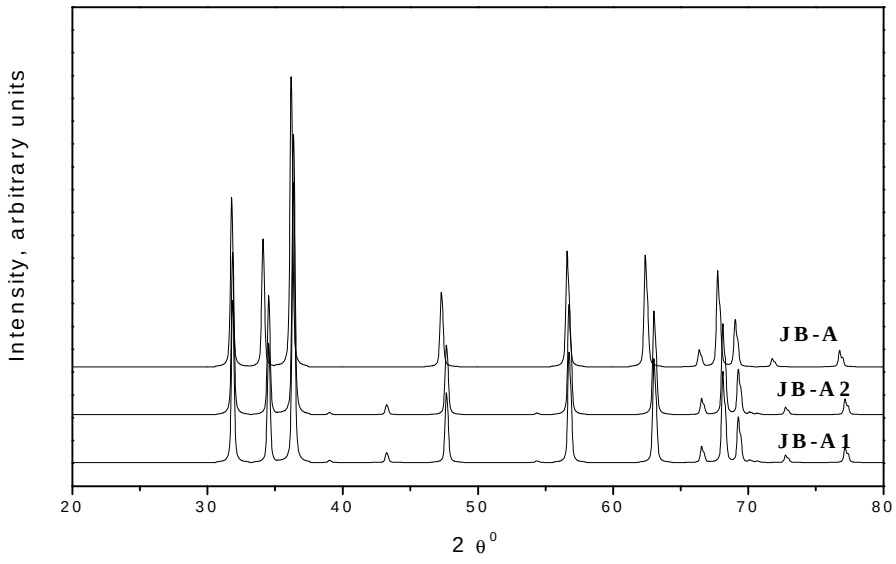


Fig. 4 (b) The powder X-ray diffraction patterns of the intermediate samples and the final products of the bhasma samples prepared using Aghara leaves.

Table 9

Structural information for the Jasad Bhasma samples prepared using Aghara and Neem leaves

Sample	Zn % Composition	ZnO % Composition	ZnO (Lattice parameters)		
			‘a’(	‘c’(	‘V’(^{3})
JB-N1	10	90	3.252	5.208	47.697
JB-N2	4	100	3.263	5.217	48.103
JB-N	-	100	3.263	5.217	48.103
JB-A1	8	92	3.254	5.211	47.783
JB-A2	2	98	3.259	5.214	47.957
JB-A	-	100	3.261	5.214	48.016

2.3.3.3. Particle size distribution

Fig 5 (a) and (b) give the particle size distribution of the Bhasma samples JB-N and JB-A. JB-N has the particles distributed over a wide range from 0.1 μm to 100 μm. Further, the distribution does not appear to be homogeneous. JB-A

also has particles with wide distribution of sizes varying from 0.1 μm to around 200 μm with a large amount of particles between 100 and 200 μm range.

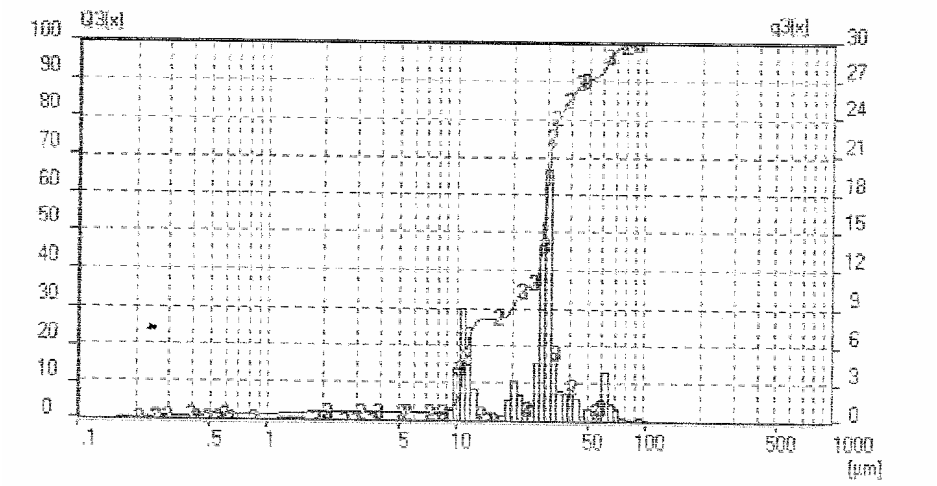


Fig. 5 (a)

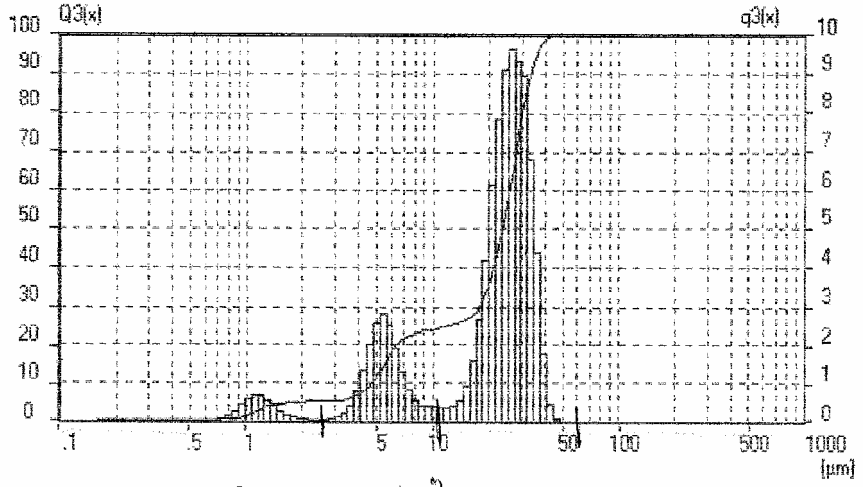


Fig. 5 (b)

Thus, the overall procedure involved in the preparation of Jasad Bhasma by using Neem leaves and Aghara leaves indicates conversion of metallic zinc into zinc oxide with the hexagonal crystal structure. While metallic zinc is converted into a brittle form during the Shodhan procedure, the combustion of the plant leaves during the Maaran procedure supplies the requisite heat for the oxidation of zinc to zinc oxide in presence of atmospheric oxygen.

2.4. CHARACTERIZATION OF JASAD BHASMA SAMPLES AVAILABLE IN THE MARKET

After studying the preparation and characterization of the bhasmas of Jasad prepared using different routes and the intermediates obtained during their synthesis using various analytical tools and we have determined the probable mechanism involved in the formation of the Jasad bhasma. We have analyzed the Jasad bhasma samples marketed by various pharmaceutical companies and compared it with the Jasad bhasma sample that we have prepared. The samples have been subjected to chemical analysis to determine their chemical compositions. The samples have been characterized by powder X-ray diffraction technique and the particle size analyzer. A comparison of the various commercial bhasmas has been done with our sample based on their physico-chemical properties.

After studying the mechanism involved in the formation of Jasad bhasma, we have tried to understand the variations in the physico-chemical properties of the

bhasma samples. An attempt has been made to establish the sensitivity of the basic requirement of a scientific study on these materials, with respect to the manufacturers of bhasmas, Ayurvedic practitioners as well as the end-users.

2.4.1. Chemical analysis

The seven Jasad bhasma samples prepared by the method involving the use of Kajjali (as quoted in the different Ayurvedic texts) showed a remarkable variation in the appearance. They varied in the color from white to dark gray. The colors of the samples and the chemical analysis data of the samples have been tabulated in Table 10. The data indicates that zinc is the only metal in major concentration in all the samples ranging form 74.33 % to 96.39 %. Sn, Pb, Fe, Ca, Mg, K, Cu and Na are present in lower concentrations in all the samples. The metals Co, Mn are present in all the samples in very low concentrations. In sample F iron is as high as 2.04 %. Sample A showed presence of Hg in very low concentration (26 ppb) while the concentration of Hg in sample D is 870 ppb. Both the samples showed presence of sulfide ions (S^{--}) in the qualitative tests. Carbonate ion (CO_3^{--}) was detected in Sample B when a large amount of sample was used for the test. The data indicates that there is a huge variance in the relative concentration of the metal atoms in the bhasma samples.

Table 10

The colors of the samples and the chemical analysis data of the samples

Metals	**A**	**B**	**C**	**D**	**E**	**F**	**G**
Col or	White	Gray	Off-white	Green	Yellow	Brick red	Yellow
Zn (%)	74.33	93.06	94.22	93.87	96.39	93.19	95.79
Sn (%)	21.36	1.06	0.67	0.93	0.62	1.16	1.01
Pb (%)	0.32	0.97	0.91	1.52	0.84	0.72	0.48
Ca (%)	1.20	1.28	1.09	1.78	0.58	1.24	1.06
Fe (%)	1.49	1.08	1.71	1.29	1.05	2.04	1.21
K (%)	0.42	0.92	0.78	0.51	0.26	1.00	0.10
Mg (%)	0.86	1.62	0.59	0.09	0.24	0.61	0.33
Cu (%)	0.02	0.01	0.03	0.01	0.02	0.04	0.02
Metals in ppm level	Co, Mn	Co, Mn	Co, Mn	Co, Mn	Co, Mn	Co, Mn	Co, Mn
Metals < 1ppm	Hg 26 ppb			Hg 870ppb			

Anions detected	S^{--}	CO_3^{--}		S^{--}

(the metal concentrations have been normalized to 100)

2.4.2. Particle size distribution

The particle size distribution of the bhasma samples is given in Fig. 5. Table 11 presents the particle size distribution for various samples as obtained from particle size analysis. The data indicate the distribution of particles over the wide

e sample A shows the particle

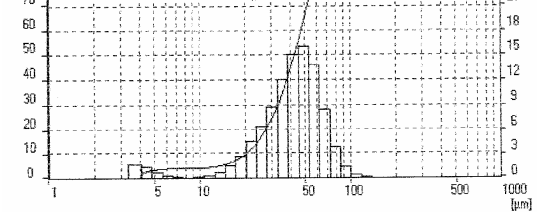
distributed

particle size ranges in a sample suggests formation of different types of particle during the heat treatments involved in the synthesis.

Table 11

Particle size distribution of the Jasad Bhasma samples marketed by various pharmaceuticals

Sample	**10 % particles < (μ**	**50 % particles < (μ**	**90 % particles < (μ**
A	20.6	42.5	67.6
B	34.5	162.7	235.2
C	10.7	29.0	49.3
D	1.0	6.9	29.4
E	2.7	10	21.3
F	4.7	23.6	33.6
G	0.8	5.6	8.0


70
60
50
40
30
20
10
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18
15
12
9
6
3
0
1
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50
100
500
1000
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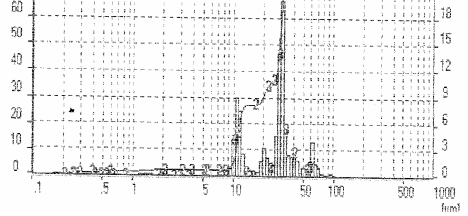
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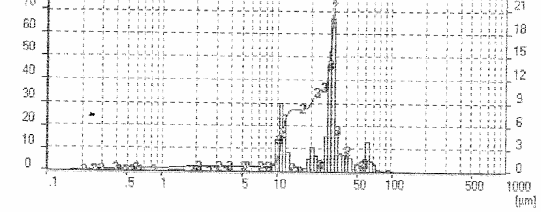

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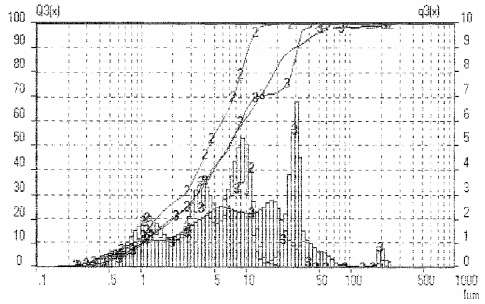

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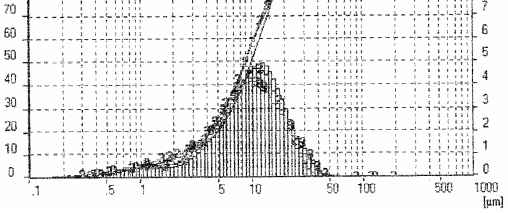
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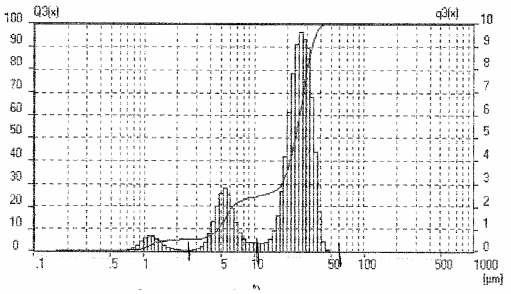
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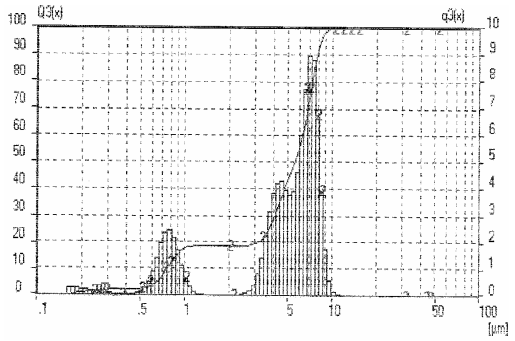

G

Fig. 5 A-G give the particle size distribution of the bhasma samples A to G

2.4.3. Powder XRD studies

Fig. 6 (a-g) gives the multiple plot of the powder X-ray diffraction patterns of the Jasad Bhasma samples. The sharp peaks in the powder patterns for all the samples suggest the high crystallinity of the materials. The phases present in the samples determined by powder XRD are given in Table 12. Sample A is a mixture of zinc oxide as the major phase with small concentrations of tin oxide (tetragonal). Sample D is a mixture of zinc oxide and zinc sulfide in the hexagonal symmetry. Sample G is a mixture of zinc oxide with a small amount of impurity phase that could not

showed presence of zinc metal in low concentrations. The XRD profiles for samples C, E and F indicate that the materials are single-phase zinc oxide in the hexagonal symmetry.

Table 12

The phases present in the samples determined by powder XRD

Sample	Crystallographic phases detected by XRD
A	ZnO (hexagonal), SnO_2, Zn metal
B	ZnO (hexagonal), Zn metal
C	ZnO (hexagonal),
D	ZnO (hexagonal), ZnS
E	ZnO (hexagonal)
F	ZnO (hexagonal)
G	ZnO (hexagonal), unidentified phase

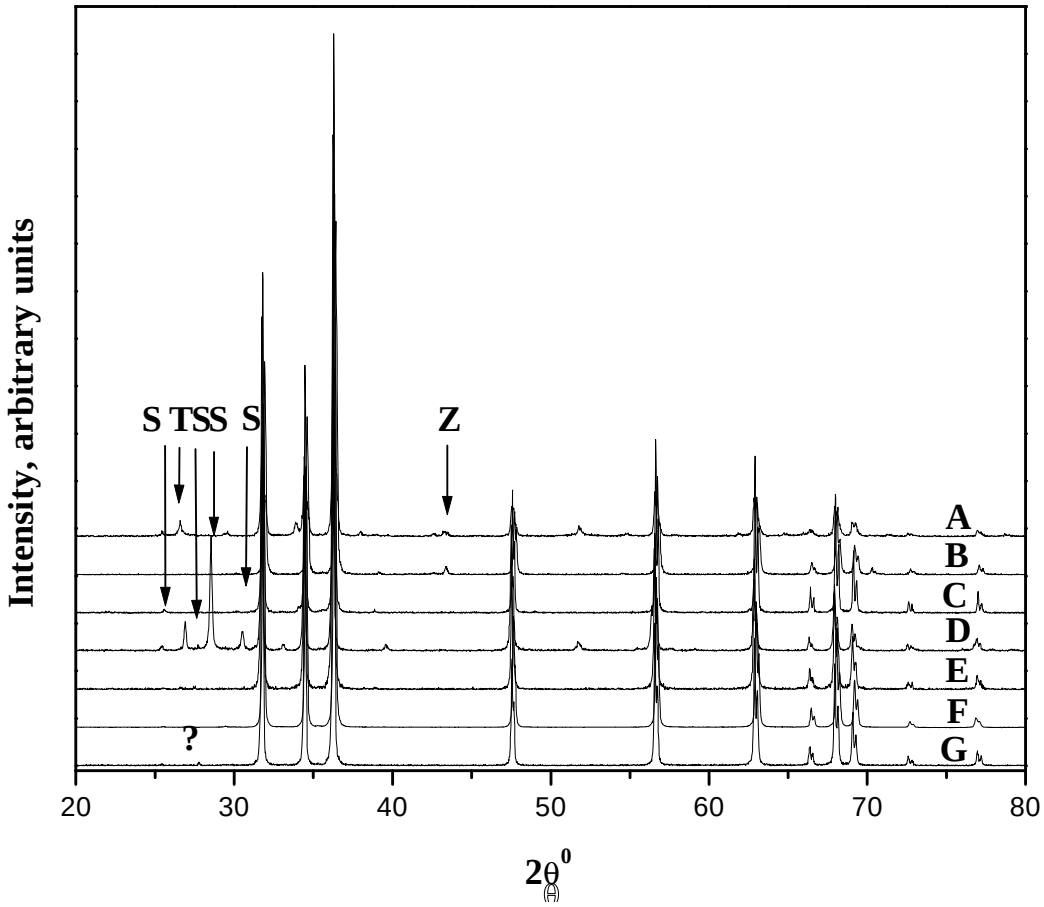

Fig. 6 The multiple plot of the Powder X-ray Diffraction patterns of the Jasad Bhasma samples A to G. Peaks for zinc metal, tin oxide, zinc sulfide and the unknown phase have been marked as '**Z**', '**T**', '**S**' and '**?**'

All the Jasad bhasma samples used in the study have been prepared using the same method employing use of Kajjali as mentioned in the various Ayurvedic texts. In our earlier work, we had reported the preparation of Jasad bhasma using the same method employing Kajjali[13]. The data obtained from the chemical analysis, powder XRD technique and the particle size analyzer are given in Table 13. The Jasad bhasma was characterized as zinc oxide in the hexagonal phase

with a variety of metals including Fe, Ca, Sn, Pb in the zinc oxide lattice. The analysis of the raw materials used for the preparation of the bhasma helped us to determine the probable sources of these metal atoms. We could establish the formation of Jasad bhasma as a conversion of zinc metal into zinc oxide in the hexagonal form with various metal atoms in the zinc oxide lattice. The conversion was enabled by the reaction of zinc metal with Kajjali (HgS) resulting in formation of zinc sulfide in both cubic and hexagonal forms. Oxidation of these intermediates during the heat treatments lead to the formation of zinc oxide. It was observed that the particle size of the sample reduced with the heat treatments and the distribution became narrower. The data help in understanding the analysis of the marketed samples. The data obtained on the marketed samples clearly indicate that some of the samples are still in an intermediate stage and the bhasma is still not formed. Presence of zinc sulfide in samples A and D thus suggest that more number of calcination steps is still required. The presence of Hg in these two samples is also an indication of inadequate heat treatment. While presence of SnO_2 as a second major phase in sample A and unidentified phase in sample G suggests that proper analysis of raw materials (the zinc metal which is used as a source for Jasad) is important before their use for the preparation of the bhasma. Presence of zinc metal in sample A and B also indicate improper treatment of Kajjali or inadequate heat treatment. The significance of each rigorous step involved in the preparation of the bhasma during the Shodhan and the Maaran procedures is quoted in literature and no deviations from the procedures can lead to formation of such intermediates.

Zinc oxide is known to exhibit a variety of colors when doped with various metal atoms. The color of zinc oxide is also dependant on its particle size. The variation in color of the bhasma samples could be due to the different metal atoms present in the samples in various concentrations and the wide range of particle size distribution shown by the bhasma samples.

Various tests mentioned in the Ayurvedic literature to check the formation of the Jasad bhasma are given in Table 1. These tests are well designed to confirm that zinc does not remain in a metallic form in the sample and that the bhasma has a small particle size. However, it cannot detect the Hg present in the samples in such low concentrations. Neither can these tests detect presence of tin oxide and other impurities in the bhasma samples. These tests were designed with the limitations of the analytical support in the olden days. Today with the availability of excellent analytical techniques, the appropriate use of these techniques for characterization and standardization of the bhasma is required.

The medicinal potentials of the bhasmas have been well established with their successful routine use since a few centuries. However, the pharmaceutical companies need to ensure that the products marketed are the bhasmas and not the intermediate samples. These products have to be standardized and tested for the presence of toxic elements like Hg or presence of zinc in metallic form. The pharmaceutical companies can routinely use these analytical techniques as quality control tools. A proper use of the techniques shall ensure the uniformity in the samples marketed by the manufacturers. A routine use of such scientific techniques

will ensure standardization of the products to a certain extent and would definitely help in promoting the use of bhasmas for medication.

2.5. CONCLUSIONS

Jasad Bhasma prepared using Kajjali

The treatment of zinc metal 21 times with milk is a pre-requisite step to obtain zinc in powdered form. This powdered mixture of zinc metal and zinc oxide is converted first into cubic zinc sulfide, then into hexagonal zinc sulfide and finally into hexagonal zinc oxide during the treatments with aloe vera gel and lemon juice followed by calcination procedure described. Zinc sulfate could also be an intermediate formed during the synthesis. Jasad bhasma prepared using Kajjali is thus zinc oxide in the hexagonal form containing metals like Fe, Ca, Mg, Sn, Pb, Mn, Co, Cu and Cr in low concentrations. The final product does not contain mercury even in trace levels. The particle size of the sample decreases with various treatments and the distribution becomes narrower. The Jasad bhasma has the particle size distributed between 5 and 10μm.

Jasad Bhasma prepared using Aghara and Neem leaves

The overall procedure involved in the preparation of Jasad Bhasma by using

Neem leaves and Aghara leaves indicate conversion of metallic zinc into zinc oxide with the hexagonal crystal structure. While metallic zinc is converted into a brittle form during the Shodhan procedure, the combustion of the plant leaves during the Maaran procedure supplies the requisite heat for the oxidation of zinc to zinc oxide in presence of atmospheric oxygen.

Analysis of the Bhasma samples available in the market

Most of the Jasad bhasma samples used in the study are still in an intermediate stage of the preparations and the bhasma can be obtained with further heat treatments. Zinc oxide in the hexagonal form is the major phase in all the samples prepared by using Kajjali. A variety of metals including Sn, Pb, Fe, Ca, Mg, K, Cu, Co, Mn and Na are present in different concentrations in the samples. T

analytical techniques like powder XRD can be a useful tool for quality control for various pharmaceutical companies.

2.6. REFERENCES

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// SAMUDRAPANCHAK //

3.1. INTRODUCTION TO SAMUDRAPANCHAK

Ayurved believes that every material with a natural origin has some medicinal value. Popularity of Ayurved is growing even outside India not only because many of these medicines show no noticeable side effects but also because of the fact that most of them are of herbal or natural origin[1]. Although Bhasmas too, form an important class of Ayurvedic medicinal system that are inorganic in nature (being metal preparations), the Bhasmas involving use of calcium sources are not of 'mineral' origin. Bhasmas containing calcium are prepared using different calcium sources of marine origin like natural pearls, corals, corn shells and pearl shells and are labeled as 'Samudrapanchak'[2]. These Bhasmas are of bioinorganic origin and these also differ from each other in their medicinal properties, depending on the source of calcium used during synthesis. The group Samudrapanchak consists of the Bhasmas of Kapardika, Shankha, Shimpla, Paval and Moti[3]. The Bhasmas included in the Samudrapanckak group, their sources and their medicinal properties are listed in Table 1. All these Bhasmas have a common method of preparation. However, they are known to differ in their medicinal properties.

Kapardika Bhasma is one of these bhasmas and is administered to patients suffering from high fever. It is prepared by using Kapardikas (corn shells) as the source of calcium[4].

Table 1

Samudrapanckak group, their sources and their medicinal properties

Bhasma	**Source**	**Medicinal properties / Use as a drug in case of**
Kapardika Bhasma	Sea shells	High fever
Shankha Bhasma	Conch Shells	Acidity
Shimpla Bhasma	Pearl Shell	Cosmetics
Paval Bhasma	Coral	Digestive problems
Mauktik Bhasma	Pearl	Power enhancing

The general procedures for preparation of bhasmas involve treatment of the source of the metal with various herbal or animal products and calcination of the mixture in a sealed earthen pot used as the crucible in a traditional furnace described in literature[3-5]. This process is repeated till the bhasma is obtained. The metal is said to be completely converted into the bhasma when it complies with the tests and observations described in the Ayurvedic texts (Table1-Chapter-1). In case of Samudrapanchak the ‘taste of the Bhasma’ also constitutes an important test in deciding the formation of the Bhasma. An itching sensation to the tongue

indicates incomplete formation of the Bhasma[6]. The intermediate product is then subjected to additional treatments with herbal products followed by calcination till the itching sensation of the sample is lost. However, the test is highly subjective. With the advent of sophisticated instrumentation, it has now become necessary to develop tests that would not only throw light on the changes taking place during formation of the bhasma, but would also help to establish some quality control methods for standardization of the synthetic procedure of the bhasma and the final product.

Researchers have reported the analysis of some of the Bhasmas belonging to the Samudrapanchak family[7-9]. These reports describe the study on Bhasmas limited to the chemical analysis of the raw materials and the final product. Further, the reported literature is based on the study of the bhasmas marketed by different pharmaceutical companies. No reports could be found that throw light on the various processes involved in the preparation of the bhasma and the intermediates obtained at the various steps. In the present work, we have prepared the Kapardika bhasma strictly according to the method quoted in the ancient Ayurvedic text Rasaratnasamuchhaya under the supervision of an Ayurvedic expert. The raw material, intermediates and the bhasma have been characterized by various techniques described in Chapter I to understand the mechanism involved in the formation of the Bhasmas.

3.2 PREPARATION OF KAPARDIKA BHASMA

3.2.1. Shodhan of Kapardika

Kapardikas, each weighing about 2 to 3 g were chosen as the raw material for the preparation of the Kapardika Bhasma, as per the specifications given in Rasaratnasamucchaya. Kapardikas were first cleaned with hot water. Freshly extracted lemon juice was diluted four times with water. The cleaned Kapardikas (K0) were immersed in this diluted lemon juice. After 4 hours the lemon juice was filtered. A white powder coating (KLJ) was formed on the surface of the Kapardikas. An effervescence was observed while the Kapardikas were immersed in the lemon juice.

3.2.2. Maaran of Kapardika

The Kapardikas along with the coating were then transferred to an earthen pot (acting as the crucible). The pot was covered with an earthen lid and sealed with mud. This crucible was ignited in the traditional furnace using cow dung cakes. The sample obtained after the first calcination (KB1) was subjected to the tests described in Table1 (Chapter-I) to check weather the bhasma has been formed or not. It was then treated with Aloe vera gel and triturated for 8 hours. The mixture was dried and mixed with equal quantity of lemon juice. The resultant mixture was triturated for 8 h. This mixture was calcined in a similar fashion to obtain KB2.

The procedure was repeated to obtain the final product Kapardika bhasma (KB3). The samples obtained after the first (KB1), second (KB2) and the third (KB3) calcination processes were isolated.

3.3. CHARACTERIZATION

3.3.1. Chemical Analysis

The raw material, intermediates and the final product were analyzed qualitatively for the presence of metals other than calcium and the anions by routine chemical tests. Calcium was estimated from the final product volumetrically by titrating its solution with a standard EDTA solution using Pattern and Readers indicator and ammonium chloride-ammonia buffer. Calcium and the other metals in lower concentration were determined by EDAX on a JOEL-JSM-5200 Scanning Electron Microscope with Energy Dispersive X-ray analysis facility (Kevex). Qualitative analysis of the Kapardika bhasma shows the presence of metals like Mg, Al, K, Fe, Zn and Na other than calcium. Carbonate is the only anion present. EDAX results showed that the relative atomic percentages of metals other than calcium are less than 1% whereas calcium is the only major metal (96%). Volumetric estimation of calcium in the bhasma showed presence of 39%calcium

corresponding to about 98% calcium carbonate.

3.3.2. Powder X-Ray Diffraction Analysis

Fig.1 shows the powder X-ray diffraction patterns of the raw materials, intermediates and the final product. The pattern of KB0 shows that the raw material Kapardika is in the aragonite form of calcium carbonate. The diffraction pattern of KB1 shows the presence of two phases, calcite form of calcium carbonate and calcium hydroxide. Thus during the first heat treatment, the crystalline form of calcium carbonate changes from aragonite to calcite. The peaks for the calcite phase are strong and sharp. The calcium hydroxide peaks are broader and reflect smaller crystallite size. Hydroxide phase is the major one. The diffraction pattern of KB2 shows presence of three phases, calcite, calcium hydroxide and calcium oxide. Calcite is the predominant phase. The peaks for the calcite phase are sharp, while that for the hydroxide phase are broader. Calcium oxide phase is in low concentration. The final product, Kapardika bhasma (KB3) was obtained by treating this sample with Aloe Vera juice and igniting it in a similar manner. The XRD pattern of the bhasma shows presence of only one phase, calcite form of calcium carbonate. This indicates complete conversion of the hydroxide and oxide phases into calcium carbonate. However, the peaks are even sharper than those of the KB2 sample and reflect high crystallinity of the final product. The lattice parameters of the calcite phase in various samples are listed in Table 2.

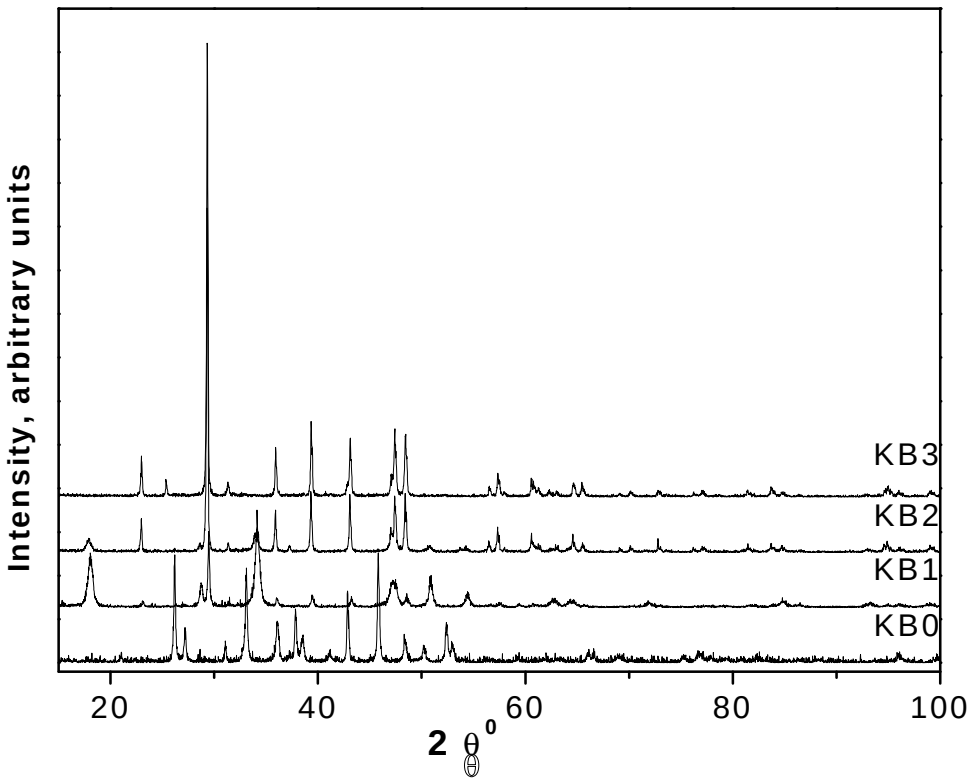

Fig. 1 Multiple plot of the powder XRD patterns of the Kapardika, the intermediates and the Kapardika Bhasma.

Table 1

The phase analysis obtained from with the help of powder XRD data

Sample	Phase present
KBO	$CaCO_3$ (Aragonite)
KB1	$CaCO_3$ (Calcite) + $Ca(OH)_2$
KB2	$CaCO_3$ (Calcite) + $Ca(OH)_2$ + CaO
KB3	$CaCO_3$ (Calcite)
KB4	$CaCO_3$ (Calcite)

Table 2

Lattice parameters and the crystallite size of the calcite phase present in the samples

Sample	Unit cell parameters of the calcite phase		
	a (Å)	c (Å)	V $(Å)^3$
ASTM data (Calcite)	4.989	17.062	367.77
KB1 (Calcite)	4.9904	17.0759	368.27
KB2 (Calcite)	4.9939	17.0775	368.78
KB3 (Calcite)	4.9877	17.0952	368.29

3.3.3. Thermal studies

Thermograms of the raw materials, intermediates and the final products are shown in Fig.2 while the differential thermograms are shown in Fig. 3. The temperature range for the various decomposition steps and the percentage wt. losses at each step are presented in Table 3.

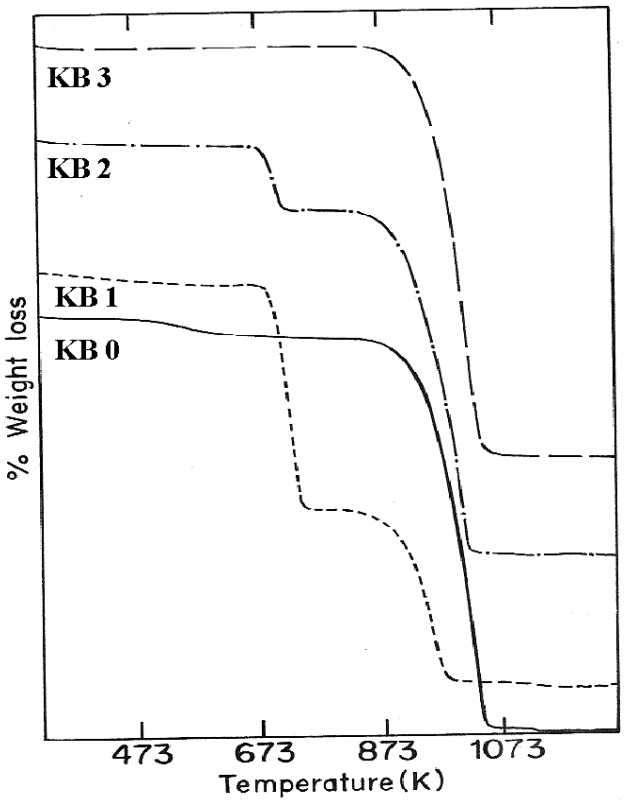

Fig. 2 Multiple plot of the thermograms of the raw material (kapardikas) , intermediates and the final product.

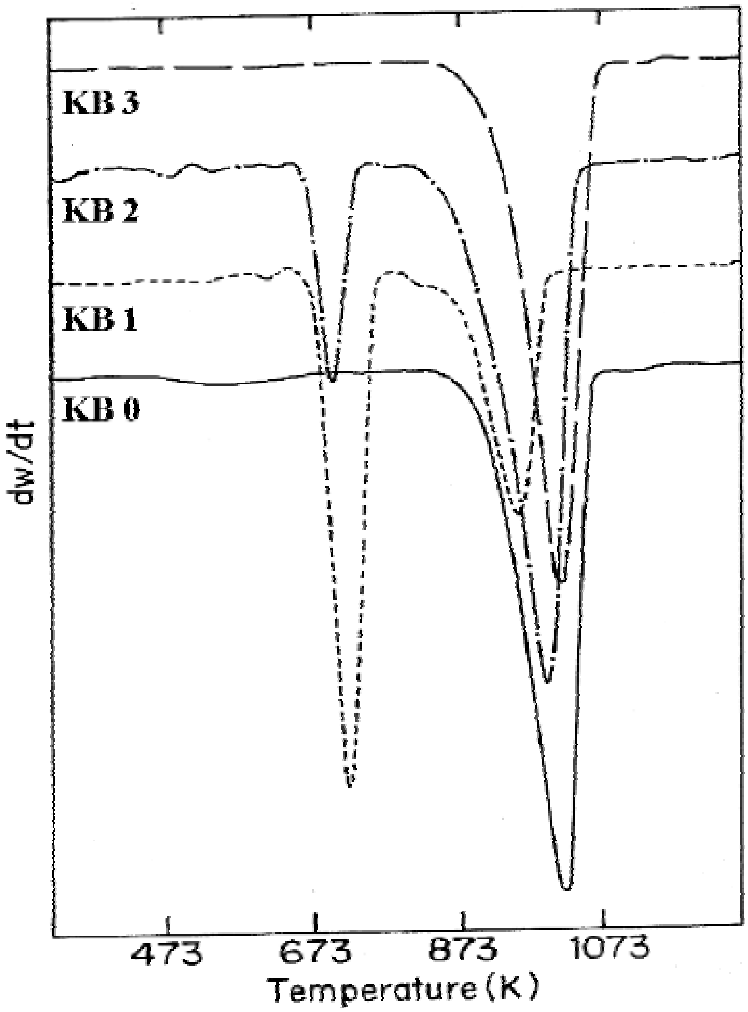

Fig. 3 Multiple plot of the differential thermograms of the raw material (kapardikas), intermediates and the final product.

The thermogram of KB0 shows a major decomposition in the temperature region from 861 to 1142 K, likely due to the decomposition of calcium carbonate in the aragonite form into calcium oxide as confirmed by powder XRD. 41.9% weight loss corresponds to 95.2% calcium carbonate in the Kapardikas used as raw

materials for preparing the bhasma. The remaining part of the Kapardikas (about 5%) could be trapped moisture and organic matter in the Kapardikas.

Table 3

Decomposition temperature ranges and relative percentages of the phases present in the samples as obtained from the TGA-DTA studies.

Sample	Temp. Range	%wt.loss	Phase present	% of the phases
KB0	588-869	41.856	Aragonite	95.13
KB1	388-553	16.266	$Ca(OH)_2$	66.87
	568-791	12.79	Calcite	29.07
KB2	372-486	2.789	$Ca(OH)_2$	11.47
	578-867	36.101	**Calcite**	82.45
KB3	631-864	43.300	Calcite	98.41

The thermogram of KB1 shows decomposition of the sample in mainly two regions. The first decomposition is in the range of 841 to 1099 K and corresponds to the decomposition of calcium hydroxide. 16.3% weight loss in this region suggests that the sample contains 66.9% hydroxide phase. The second decomposition corresponding to 12.8% weight loss is in the range of 841 to 1064 K

and is due to the decomposition of calcium carbonate to calcium oxide. The amount of calcium carbonate present in the sample is 29.1%. Thermogram of KB2 also shows decomposition in the two regions similar to that of KB1. However, the percentage weight losses in the two regions indicate that calcium hydroxide is 11.5% and calcium carbonate is 82.5%. Thermogram of KB3 shows decomposition in only one region from about 902 to 1150 K. 43.3% weight loss in the KB3 sample indicates presence of about 98% calcium carbonate in the final sample, Kapardika bhasma.

3.3.4. Infrared Analysis

The IR Spectra of the raw material Kapardika show bands at 2497 (w), 2520 (w), 1786 (m), 1473.5 (s), 862 (m) and 711 (m) resembling the reported spectrum of the aragonite form of calcium carbonate. The extra bands at 3398 (wb), 2962, 2923 and 282 cm^{-1} correspond to the frequencies of methyl and methylene groups from the organic matter in the kapardikas. The bands at 1261, 1024, 804 (w), 700 (w) cm^{-1} are due to the bending vibrations of organic material that could be due to hydrocarbons present in the raw material. A possible organic impurity has been confirmed by the thermal analysis also. The IR spectrum of KB1 shows a strong band at 3641 cm^{-1} due to the hydrate molecules. Powder XRD pattern of this sample confirms presence of calcium hydroxide as the major phase. There are no absorption bands corresponding to the organic material suggesting complete

combustion of the organic phase during calcination of the KLJ sample. The bands at 1793, 1412, 875 and 714 cm^{-1} show the presence of calcite form of calcium carbonate.

The IR spectrum of KB2 is similar to that of KB1. However, the intensity of the peak corresponding to calcium hydroxide is less than that observed in the KB1 sample. The calcite peaks are prominent. The IR spectrum of KB3 shows presence of peaks corresponding to only calcium carbonate in the calcite form.

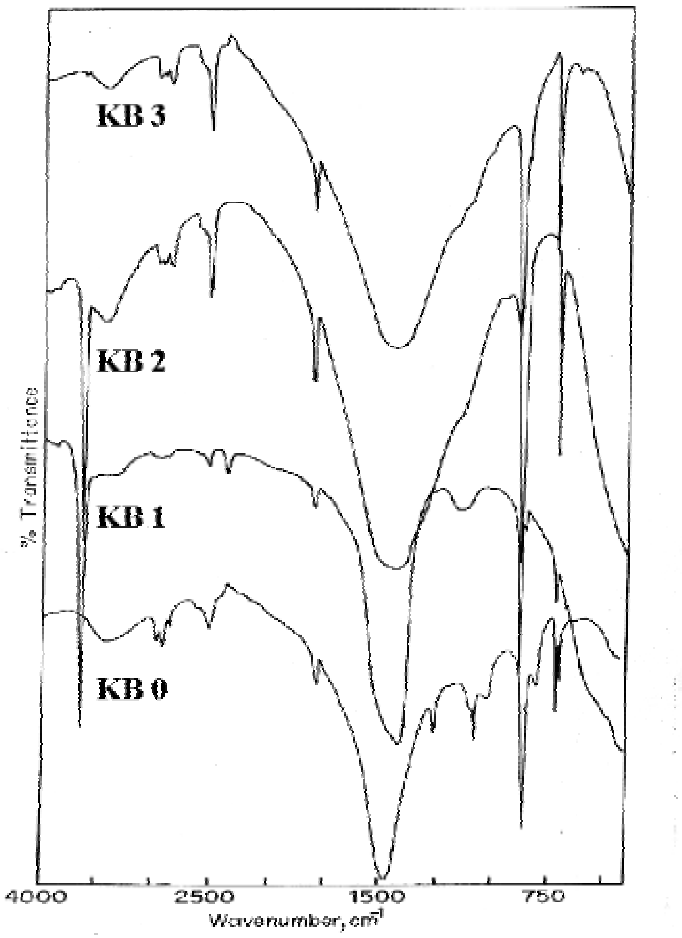

Fig. 4 Multiple plot of the FTIR spectra of the raw material (kapardikas), intermediates and the final product

3.3.5. pHmetric studies

The pH of the samples was determined using a pHmeter (Labindia) with a gel filled combined electrode. Kapardikas were immersed in lemon juice and the pH of the mixture was determined after a regular interval of one hour for four hours to study the reaction between the Kapardikas and the lemon juice. Also, the samples obtained after each calcination processes were suspended in distilled

water and the pH of the water was measured in a similar way to determine the nature of the sample obtained.

The aim of pH measurements is to assess the feasibility of using simple instruments like pHmeter in characterization of the Bhasmas. The presence of calcium hydroxide and calcium oxide in the sample should form a basic solution in water, indicating incomplete formation of the Kapardika Bhasma. Presence of calcium carbonate alone in the sample should give a neutral solution in water. The pH of the lemon juice with the Kapardikas immersed in it was monitored at an interval of one hour. A gradual increase in pH was observed (from 2.26 to 4.12) which suggests that the reaction of the calcium carbonate from the Kapardikas with the lemon juice is slow. Table 4 shows the pH studies of the suspensions of the raw material and the samples obtained after each calcination process in water.

Table 4

pH of the suspensions in water of samples obtained after each calcination

Time interval	00 min	15min.	30 min.	45 min.	60 min.	75 min.
KB0	7.00	7.95	9.12	9.99	10.03	10.10
KB1	7.75	10.90	11.22	11.35	11.35	11.37

KB2	7.61	8.71	9.67	10.28	10.97	11.05
KB3	7.00	7.20	7.32	7.3	7.3	7.32

The pH of the suspension of KB0 in water shows that it is neutral. However, the gradual increase in the pH could be due to the low solubility of calcium carbonate in water (1x 10^{-3} at 25°C). The Suspensions of KB1 and KB2 in water are basic indicating the presence of calcium hydroxide and calcium oxide in the samples as confirmed by XRD and IR. The suspension of KB1 is more alkaline than KB2 indicating a higher percentage of calcium hydroxide in KB1 as compared to KB2. This is supported by the results obtained by thermal analysis with presence of 30% and 80% calcium carbonate in the two samples, respectively. Suspension of KB3 is neutral and suggests the absence of calcium hydroxide or oxide in the sample, i.e. complete conversion of the sample into the Bhasma.

As a part of the experimentation, Kapardikas were directly calcined without the lemon juice treatment. The sample obtained after calcination was only calcium oxide. Further treatment of this sample according to the authentic procedure could not yield the Bhasma. This indicates that lemon juice treatment given in the authentic procedure is a requisite step in the formation of the Kapardika bhasma. Thus, calcium citrate is also an important intermediate formed during the synthesis of the bhasma. Sane et al[9] have reported the presence of hydroxide impurity in the bhasma (obtained from the market), which was not found in the raw material.

However, no explanation for the same was given. Reports indicating the presence of hydroxide impurity in the bhasma prepared in the muffle furnace are also available[8]. In the authentic procedure, the bhasmas are prepared by carrying out the calcination process in the traditional furnace described in Ayurvedic literature. The sample taken in a sealed earthen pot is calcined in a pit by burning cow dung cakes. The porous nature of the earthen pot allows the passage of the carbon dioxide obtained during the decomposition of calcium carbonate out of the crucible. There is carbon dioxide and carbon monoxide atmosphere outside the crucible due to the burning of the cow dung cakes. Thus, the chances of the loss of carbon dioxide from the crucible are less. This is not so in case of a muffle furnace and so is the presence of the hydroxide impurity due to the lack of carbon dioxide required for recarbonation of the calcium hydroxide and oxide intermediates. After characterizing the intermediates obtained during synthesis of the bhasma, it can be stated that the reports about the presence of the hydroxide impurity could be due to the analysis of the intermediate and not the final product.

3.4. CONCLUSIONS

The Kapardika bhasma obtained by using the synthesis procedure mentioned in the Ayurvedic literature is the calcium carbonate in the calcite form

with other metals in low concentration. The analysis of the results shows that the overall process of the formation of Kapardika bhasma involves decarbonation of calcium carbonate in the aragonite form and reformation of calcium carbonate in the calcite form. This transformation occurs via formation of calcium hydroxide and calcium oxide as the intermediates. The characterization techniques like powder XRD, TG-DTG, Infra-red spectroscopy have been used to study the process of formation of the bhasma which can be used as the quality control methods for characterization of the samples by the industry. However, even simple quality control methods like pHmetric studies can be adopted to identify the completion of the process of the formation of the Bhasma.

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// Vanga Bhasma //

4.1 INTRODUCTION TO VANGA BHASMA

Vanga Bhasma involves use of tin metal for medication. Vanga Bhasma forms an important Ayurvedic drug commonly used for disorders related to the reproductive system in human beings. Vanga Bhasma is also used in combination with the Jasad Bhasma (Bhasma of zinc) and Nag Bhasma (Bhasma of lead), popularly known as Trivanga Bhasma (the combination of three). The formulation is also used for various ailments related to the disorders caused due to diabetes.

4.2 PREPARATION OF VANGA BHASMA

Several procedures for the preparation of Vanga bhasma are reported in the texts Rasaratnasamucchaya. The processes differ from each other with respect to the materials used during the Shodhan of Vanga and in the Maaran procedure. Freshly prepared juice from the plant nirgundi or limewater is commonly used during the Shodhan process. The Maaran processes involve use of Kajjali or Haartal or herbal products for the treatments. In the present work, we have prepared Vanga Bhasma by using lime water during the Shodhan and herbal products during the Maaran step.

4.2.1. Shodhan of Vanga

In the Shodhan process, 50 g of limestone was dipped in water and stirred. The mixing was exothermic. On cooling, the mixture was filtered and the filtrate (Liquid A) was used for treatments with the tin metal during the Shodhan process. About 5 g of tin metal (Vanga) was heated in an iron vessel over burning coals. The molten tin was poured into 50 ml of liquid A and filtered on cooling. The filtered tin pieces were again heated to melt and poured into the filtrate. This step was repeated seven times. Each time fresh quantity of Liquid A was used. It was observed that with the increasing number of steps, lesser amount of the tin was getting melted. Also, the tin metal pieces were losing the luster, and formation of a white coating on the metal pieces could be observed. These metal pieces obtained at the end of the seventh step were subjected to the process of Maaran.

4.2.2. Maaran of Vanga

The herbal products used for treatments during the Maaran process include 1.25 g each of *Ova* (O), *Jeera* (J), *Haldi* (H), *Pimpali* (P), *Chincha* (C). The above materials were dried and powdered separately. The product obtained after the process of Shodhan was again heated in the iron vessel. Some part of the tin pieces melted. To the hot mixture of molten tin was added small quantity of Ova with continuous stirring. The powder Ova burnt and the formation of white powder could be observed. Thus tin metal was getting converted into a white powder because of the local burning due to the combustion of Ova. Similarly 1.25g of Ova was added with continuous stirring, small quantity at a time. In the same way,

Jeera, Haldi, Chincha churna and Pimpal churna were added to the hot metal. The powders were added in small quantities at a time with continuous stirring one after the other. With the addition of the powdered herbal products and a continuous heat treatment, tin metal was converted into a fine powder (VB1). The powder was covered partially with an iron lid and heated for four hours on the Bunsen burner to obtain sample (VB2). Then the powder was cooled and mixed with an equal quantity by weight of lemon juice. The mixture was triturated for hours till a fine powder was obtained. To the resultant mixture was added equal quantity of aloe vera gel. Again the mixture was triturated. Finally, the mixture was transferred to an earthen pot, which was sealed (it serves as the crucible) and calcined in a traditional furnace described in the Ayurved literature. The sample obtained after the heat treatment (VB3) was subjected to the tests mentioned in the Ayurvedic literature to check whether the bhasma has been obtained. The treatments with lemon juice and aloe vera gel followed by calcination were repeated twice to obtain sample VB4 and the final sample Vanga bhasma (VB5).

4.3. CHARACTERIZATION OF VANGA BHASMA

The Vanga Bhasma sample was obtained by following the method involving treatment of the molten tin metal with herbal products including ova, jeera, haldi, chinch churna and pimpali churna. During the preparation it was observed that as these herbal products were added to the molten metal, they caught fire and burnt. A

local reaction could be observed near the burning mass resulting in the conversion of metallic tin into a fine white powder. The herbal products burnt partially leaving behind gray ash (carbonaceous matter).

4.3.1. Chemical analysis

Table 1 gives the relative metal concentrations obtained by chemical analysis of the intermediates and the final product. The chemical analysis of the raw materials enabled the determination of the probable source of these metals in the samples. It can be observed that tin is the only metal in major concentration in all the samples. The final Vanga bhasma sample (VB5) contains Pb and Zn in small concentration (2.4 and 1.58 % respectively) the source of which could be tin metal used as the raw material for the preparation of the Vanga Bhasma that contained both Pb and Zn.

Presence of iron in the bhasma sample could be due to the container in which the heat treatments were given, while calcium could be from the limewater used during the Shodhan process. The bhasma sample contains K, Na and Mg in concentrations of less than 1 % whose presence could be attributed to the various herbal products used during the heat treatments of the Maaran procedure. It can be observed that the concentrations of these metal atoms increase gradually with the different treatments. In addition to these metal atoms, the bhasma sample contains Co, Cu, and Mn in very low concentration (108ppm, 450 ppm and 60 ppm,

respectively).

Table 1

Relative metal concentration as obtained from AAS and ICP analysis

Metals	VB1	VB2	VB3	VB4	VB5	Metal Source
Sn	92.98	92.68	91.92	91.58	91.52	Sn metal used as the source for Vanga Bhasma
Zn	2.31	2.34	2.34	2.34	2.34	Sn metal used as the source for Vanga Bhasma
Pb	1.60	1.59	1.59	1.59	1.58	Sn metal used as the source for Vanga Bhasma
Fe	0.26	0.26	0.80	0.94	0.94	Iron container in which the heat treatments were carried out
Ca	1.23	1.23	1.24	1.24	1.24	Lime water used during the Shodan process
Na	0.42	0.52	0.54	0.57	0.59	Herbal products used during the Maaran process
K	0.72	0.81	0.84	0.84	0.87	Herbal products used during the Maaran process
Mg	0.48	0.57	0.73	0.89	0.92	Herbal products used during the Maaran process

Atomic concentrations have been normalized to 100 %.

4.3.2. Carbon analysis

The burning of the herbal products results in the formation of ash. The carbon content in the samples is given in Table 2. Sample VB1 obtained after the treatment of tin metal with all the five herbal products contained about 8.28 % carbon. This indicates partial combustion of the herbal products during the heat treatments resulting in the formation of ash (carbonaceous products). On further heating of this sample, the carbon content decreased to 1.74 % in VB2 due to its combustion in presence of atmospheric oxygen.

Table 2

Carbon content in various samples obtained from the carbon analyzer

Sample	Carbon content (%)
VB1	8.28
VB2	1.74
VB3	0.26
VB4	0.07
VB5	0.00

The calcination of this sample in the furnace further resulted in the removal

of this carbon to as low as 0.26 percent (sample VB3). The sample VB5 contained no carbon at all. The successive heat treatments remove all the carbon from the sample.

4.3.3. Powder X-ray diffraction studies

The samples were analyzed by the powder X-ray diffraction technique to determine the structural changes taking place in the sample with heat treatments. Fig. 1 gives the multiple plot of the powder X-ray diffraction patterns of the intermediate samples and the final product. The relative phase composition of the samples is listed in Table 3.

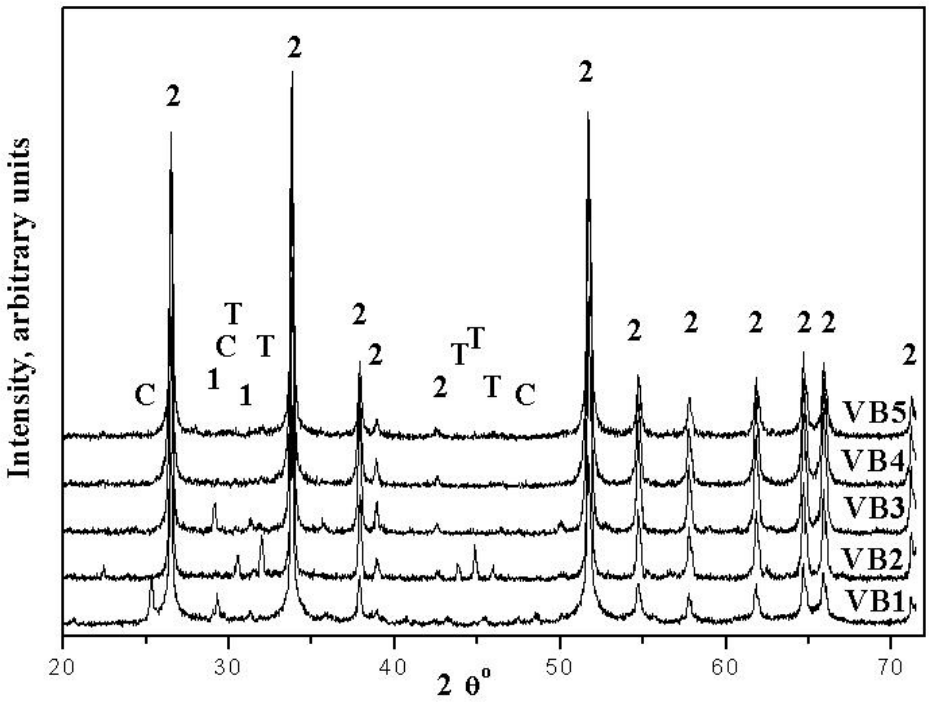

Fig. 1 Multiple plot of the powder X-ray diffraction patterns of the intermediates and the Vanga Bhasma. (2) = SnO_2, (1) = SnO, T = Tetragonal tin, C = Cubic tin

It can be observed that the sample VB1 is a mixture of tin (IV) oxide as the major phase with small concentration metallic tin. Sample VB2 also contains SnO_2 as the major phase with small concentration of SnO and metallic tin. The sample VB3 contains a mixture of SnO_2 and SnO. The samples VB4 and VB5 are single phase of SnO_2.

Table 3

Structural information obtained on the intermediates and the final product Vanga

Bhasma, by powder XRD

	Relative % phase composition		
Sample	SnO_2	SnO	Sn
VB1	96	-	4
VB2	93	5	2
VB3	90	10	-
VB4	100	-	-
VB5	100	-	-

The probable mechanism of the formation of the bhasma that can be proposed on the basis of the results obtained from the powder XRD data and the carbon analysis data are given below:

The Shodhan process ‘softening of the metal’ by converting tin into brittle, thin sheets that can be cut into finer particles. During the process of Maaran, metallic tin (obtained from the Shodhan process) is converted into SnO_2 powder during the various heat treatments. The heat generated by the combustion of the herbal products assists the oxidation of metallic tin into tin (IV) oxide with the help of atmospheric oxygen. However, only that part of the sample gets oxidized which comes in contact with the herbal product due to the local heating of the burning material. Hence, continuous stirring of the sample is required to ensure thorough mixing. An incomplete conversion of the sample can result in the remainder of tin metal in the sample. The partial combustion of the herbal products leaves behind

carbonious mater in the sample.

Table 4

Structural information of SnO_2 phase present in the intermediate samples and the final product, Vanga Bhasma, obtained from powder XRD

	Tin (IV) oxide (SnO_2)	
Sample	Crystallite size (nm)	Unit cell volume ($Å^3$)
VB1	104	71.511
VB2	102	71.596
VB3	105	71.631
VB4	103	71.639
VB5	104	71.642

Table 4 gives the structural properties of the samples as determined from the X-ray profiles. The data confirms nanocrystalline nature of the powder samples. It can be observed that all the samples exhibit a crystallite size of about 105 nm. It is worth noting that the crystallite size of the SnO_2 phase remains unaltered in spite of the heat treatments given to the samples. The lattice parameters (unit cell volume) seem to increase with the various treatments. This could be due to the presence of various metal ions in the SnO_2 lattice there by causing lattice expansion.

4.4. ANALYSIS OF THE MARKETED SAMPLES

The Vanga Bhasma sample prepared by us is compared with the Vanga Bhasma obtained from various pharmaceutical companies prepared employing similar procedures.

4.4.1. Chemical Analysis

Table 5 gives the elemental analysis of the various samples determined using AAS and ICP spectroscopic techniques. It can be seen that the concentration of Sn in the Vanga Bhasma varies between 88.86 and 94.55 % in the Vanga Bhasma samples. While all the samples contain Pb, Zn, Ca, Fe, K, Na, Mg, their relative concentrations vary over a wide range from sample to sample.

Table 5

Relative metal concentrations as obtained from AAS and ICP analysis

Metals	A	B	C	D	E	VB5
Sn	92.67	88.86	91.07	94.55	90.87	91.52
Zn	1.78	2.12	0.68	1.58	1.83	2.34
Pb	2.35	3.14	1.64	1.10	1.29	1.58
Fe	1.62	1.52	0.94	0.12	2.94	0.94

Ca	0.12	1.31	2.31	1.15	1.16	1.24
Na	0.19	0.23	1.29	0.49	0.62	0.59
K	0.86	0.43	1.62	0.34	0.58	0.87
Mg	0.41	0.27	0.45	0.67	0.71	0.92

Atomic concentrations have been normalized to 100 %.

Sample A and B contain Pb in higher concentrations than the other samples. It could be due to the presence of higher concentration of Pb in the tin metal used as the raw material for the preparation of the Bhasma. The higher concentration of Fe in sample E indicates higher inclusion of Fe from the iron container used during the heat treatments. Sample C contains higher concentration of Na, K, and Mg (as a whole) indicating more number of herbal treatments being given to the samples (as the source of these metals are likely to be the herbal products).

4.4.2. Carbon Analysis

It is desired that the Bhasma samples do not contain any carboneous matter.

The carbon analysis data shows that the samples A, B D and E contain carbon. This could be due to insufficient heat treatments or calcinations steps which would have ensured the complete combustion of the organic matter.

Table 6

Carbon content in various samples obtained from the carbon analyzer

Sample	Carbon content (%)
A	0.82
B	5.03
C	0.00
D	1.35
E	2.63
VB5	0.00

4.4.3. Powder XRD studies

Fig. 2 gives the powder X-ray diffraction patterns of the samples. The sharp reflections in the powder patterns reflect high crystallinity of the samples. The Fig. 2 indicates that the powder patterns for samples A and C contain peaks for tin (IV) oxide as well as metallic tin, while that of sample B contains a mixture of SnO_2 and SnO. The relative phase composition data obtained from the powder XRD patterns of the commercial samples is given in Table 7. Sample A and D contain tin (IV) oxide as the major phase (97% and 96% respectively) with small concentration of metallic tin.

Sample B is a mixture of 98% SnO_2 and 2% SnO. It can be seen that sample

C and E contain SnO_2 as the only crystallographic phase. The presence of metallic tin in the samples indicates insufficient herbal and heat treatments. The presence of carbonaceous matter in the sample could result in the reduction of SnO_2 to SnO as seen in Sample B indicating insufficient calcinations steps.

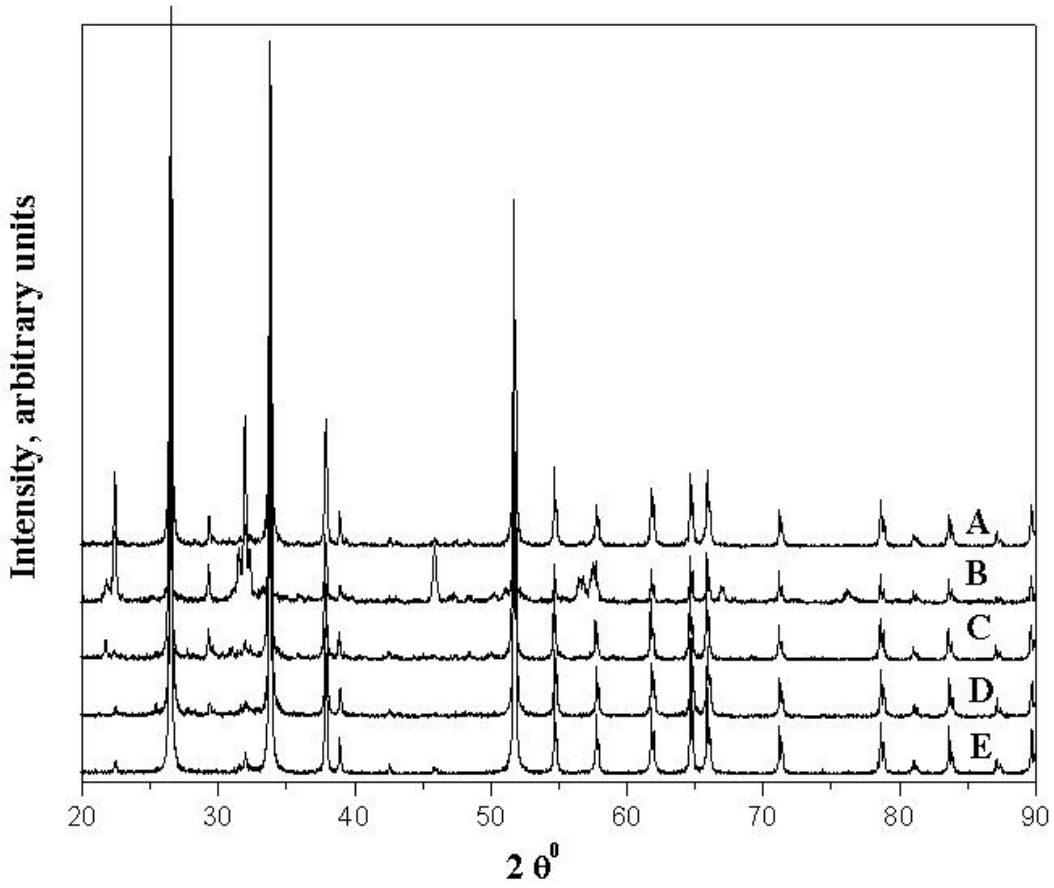

Fig. 2 Multiple plot of the powder X-ray diffraction patterns of the Vanga Bhasma marketed by different pharmaceutical companies

Table7

Structural information about the marketed Vanga Bhasma samples as obtained by powder XRD

	Relative % phase composition		
Sample	SnO_2	SnO	Sn
A	97	-	3
B	98	2	-
C	100	-	-
D	95	-	5
E	100	-	-
VB5	100	-	-

4.5. CONCLUSIONS

Vanga Bhasma obtained as per the procedure mentioned in the Ayurvedic

text Rasaratnasamucchaya is tin (IV) oxide with the tetragonal crystal structure. The formation of the Bhasma involves conversion of metallic tin into tin (IV) oxide with tin (II) as a probable intermediate. The conversion takes place in the presence of the heat provided by the combustion of herbal products and atmospheric oxygen. The Vanga bhasma has a range of metal ions including Pb, Zn, Ca, Fe, K, Na, Mg in small concentrations in the tin oxide lattice. Insufficient heat treatments lead to presence of the carbonaceous materials in the sample, which on further heating may lead to the reduction of SnO_2 to SnO or tin metal. The analysis of the marketed samples indicates the presence of SnO_2 as the major phase in all the samples with carbon and SnO as impurities in some.

4.6. REFERENCES

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// AYURVED IN THE 21ST CENTURY //

5.1. Understanding the Mechanism of the formation of the Bhasmas

The thorough physico chemical characterization of the raw materials, intermediates obtained during the preparations of the Bhasmas and the final products, Bhasmas helped to establish the probable mechanisms involved in the conversion of the metals into the Bhasmas. The various processes involved in the preparation of the Bhasmas could be explained as below, based on the analysis done.

5.1.1. The Shodhan process

To understand the process of Shodhan, the metals were subject to the

respective Shodhan process. The metal was chemically analyzed before and after the shodhan process. Table 1 (a) gives the chemical analysis data of the metal samples before and after the Shodhan process was performed for zinc metal (Jasad Bhasma).

The data indicate that there is no noticeable decrease in the concentration of the metals present in the zinc metal sample used as the source for the preparation of Jasad Bhasma even after the Shodhan process was performed on the metal sheet. The increase in the concentration of Ca could be from milk and that of C from the carbonaceous left behind due to the local combustion of milk when the hot metal is poured into the milk.

Table 1a

Elemental analysis of the Jasad sample before and after the Shodhan process

Elements	Element concentration (%) Before treatment	Element concentration (%) After treatment

Zn	97.1021	94.2173
Sn	0.13	0.09
Pb	2.10	2.13
Fe	0.58	0.93
Ca	0.02	1.18
Mg	0.005	0.0059
Co	0.0017	0.0011
Cu	0.056	0.051
Mn	0.0052	0.0047
C	-	1.29

Name of the Bhasma	Jasad
Metal source for the Bhasma	Zn
Material used during the process of Shodhan for treatments	Cows milk

Similar chemical analysis data obtained for the Vanga Bhasma and Kapardika Bhasma is shown in Table 1 (b) and Table 1 (c) respectively.

Table 1b

Elemental analysis of the Vanga sample before and after the Shodhan process

Elements	Element concentration (%) Before treatment	Element concentration (%) After treatment

Sn	95.71	94.72
Zn	2.13	2.07
Pb	1.52	1.43
Fe	0.19	0.21
Ca	-	1.16
Mg	0.45	0.41

Name of the Bhasma	Vanga
Metal source for the Bhasma	Sn
Material used during the process of Shodhan for treatments	Lime water

Table 1c

Elemental analysis of the Kapardika sample before and after the Shodhan process

Elements	Element concentration (%) Before treatment	Element concentration (%) After treatment

Ca	76.30	41.89
Fe	-	0.16
Mg	0.32	0.47
K	0.21	0.35
Na	0.27	0.41
C	22.90	56.72

Name of the Bhasma	Kapardika
Metal source for the Bhasma	Ca
Material used during the process of Shodhan for treatments	Lemon juice

The chemical analysis data obtained for the sources of the Vanga Bhasma and Kapardika Bhasma before and after the Shodhan process also indicate that the process does not involve purification of metal. The Shodhan process mainly involves converting the metal into its physical form that can be further treated during the Maaran step. The process is similar to the process of ‘quenching’ with which the big metal sheets or blocks can be converted into a fine powder or brittle thinner sheets by dipping them into various solutions when they are hot. The process of Shodhan has been interpretated by many Ayurvedic researchers as the process of purification of the metals (as the name suggests, *shudh* = pure, in Marathi) . However the chemical analysis data does not indicate the purification of the metals during the Shodhan process.

The Ayurved literature prescribes the Shodhan process as a ‘requisite’ step before the metal can be further subject to the process of Maaran. To understand the Shodhan process further, in another set of experiments, the preparation procedures were followed as in the literature, while the Shodhan process was skipped. It was observed that the sample did not get converted into the Bhasma even after the appropriate steps of the Maaran process were performed on the virgin metal (untreated by Shodhan process). The Bhasma could not be obtained even after the steps in the Maaran were repeated a number of times that as prescribed in the texts. Table 2 gives the results obtained form these experiments. The significance of the Shodhan process can be well understood from all the above experiments.

Table 2

The data obtained from the analysis of samples which were directly subject to the

Maaran procedure

Bhasma	Maaran process	Result
Jasad	Trituration of zinc metal sheets with Kaajali, followed by lemon juice and aloe vera and calcinations of the mixture	Zinc-mercury amalgam was obtained.
Vanga	Melting tin metal followed by addition of herbal products with constant stirring.	The Bhasma was obtained after continuous heat treatment for 24 hours with four times the material (herbal products)
Kapardika	Calcinations of Kapardikas in the traditional furnace	Calcium oxide was obtained which could not undergo any change with further treatments.

In the preparations involving the Shodhan process, the sample obtained after the Shodhan process were characterized by various analytical techniques. The results obtained from these experiments are discussed in Table 3. From the data we can conclude that products obtained after the Shodhan process play a crucial role in the formation of the Bhasmas.

Table 3

The data obtained from the analysis of the samples subject to Shodhan process

Bhasma	**Shodhan process**	**Products obtained from the Shodhan process**	**Probable role of this intermediate during the Maaran process**
Jasad	Dipping the molten metal into cows milk	Fine powder containing a mixture of zinc metal and zinc oxide	Facilitates easy conversion into zinc oxide when reacted with Kajjali
Vanga	Dipping the molten metal into lime water	Brittle thin metal sheets with a metal oxide coating on the surface	Easy combustion of the organic mater over the hot metal surface
Kapardika	Dipping Kapardikas into lemon juice	A mixture of calcium carbonate and calcium citrate	Presence of citrate ensures that $CaCO_3$ does not decompose totally into Cao, facilitating carbonation during further treatments.

5.1.2. The Maaran process

This is the main step in the preparation of the Bhasma. Rasashastra aims at converting the metal into the form, which can be easily assimilated into the human body. This conversion of the metal is done by treating the metal obtained from the Shodhan process with a variety of processes to obtain the Bhasma. The process of Maaran also aims at obtaining the Bhasmas which have small particle size and which do not have any component in the 'metallic' form (Maaran = to kill, in marathi). The characterization of the various intermediates obtained during the preparation of the Jasad, Vanga and the Bhasmas of Samudrapanchak and the final products indicates conversion of Zn and Sn into their respective oxides, while Ca is obtained as calcium carbonate in the calcite form during the calcination steps in the Maaran process. It is expected that a material sinters during calcination and an increase in the crystallite size as well as the particle size can be expected. However, it the particle size of the Bhasma samples does not grow during the calcination processes. This could be due to the effect of Aloe Vera gel which contsing maleic acid and other carboxylic acids that act as etching agents. Also, the gaseous atmosphere formed due to the combustion of the organic mater could prevent the union of particle as observed in the process of combustion synthesis commonly used for preparation of oxides. Table 3 shows the details of the changes taking place in the sample during the Maaran process. The mechanisms involved in the formation of the Jasad, Vanga and Kapardika Bhasma have been summarized in the

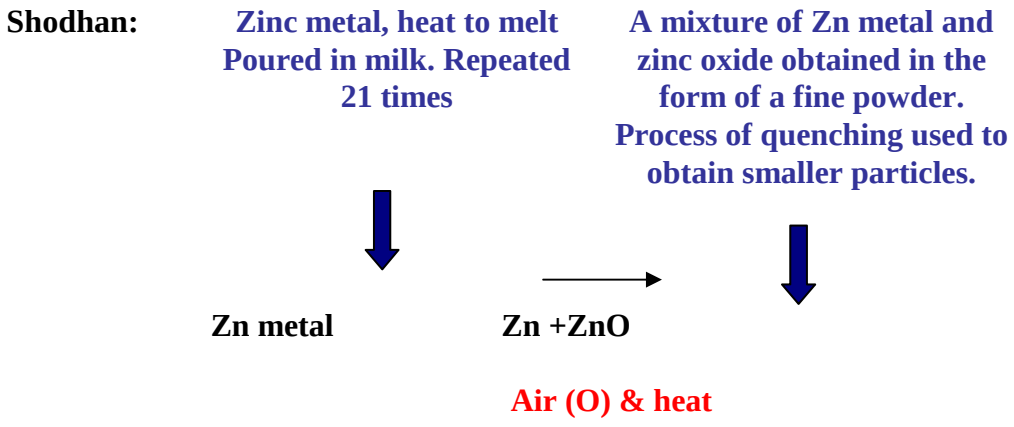
flow charts given in Fig1 (a), (b) and (c) respectively.

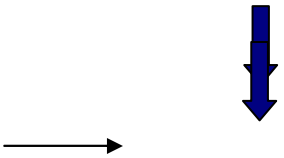
Table 3

The data explaining the significance of the Maaran procedure

Bhasma	**Maaran process**	**Significance**
Jasad	Trituration of zinc metal (after Shodhan) with Kaajali, followed by lemon juice and aloe vera and calcinations of the mixture	Kajjali (HgS) facilitates formation of Zns. Further oxidation of ZnS to ZnO is easy by calcinations at high temperature.
Vanga	Melting tin (after Shodhan) followed by addition of herbal products with constant stirring.	The combustion of herbal products (organic mater) with the hot tin metal results in oxidation of Sn to SnO_2 combustion of the small organic matter ensures complete conversion of Sn to its oxide
Kapardika	Calcinations of Kapardikas ((after Shodhan) in the traditional furnace	The presence of carbonaceous mater obtained from lemon juice and aloe vera ensures the recarbonation of the intermediates to obtain calcite at elevated temperature.

Flow chart for Jasad Bhasma





ZnS **ZnO +S ↑**

Air (O) & heat

Above mixture +Aloe Vera + lemon juice
Calcination

Oxidation of ZnS to ZnO

ZnO **ZnO (smaller particle size)**

Air (O) & heat + carboneous mater

Flow chart for Vanga Bhasma

Shodhan:

Tin metal, heat to melt Poured in lime water. Repeated 8 times

Thin foil of tin metal with metal oxide coating obtained. Process of quenching results in formation of thin foils/ sheets

Sn metal → **Sn +SnO_2 coating**

Air (O) & heat

Maaran:

Tin oxide (obtained from Shodhan process) +heated to melt + Ova, jeera, haldi, chinch and pimpli churna

Sn reacts with oxygen from air to form SnO_2., Burning of the organic mater provides the heat required for this oxidation.

Sn + SnO_2 + organic mater → **SnO_2 + C +CO↑**

CO2↑ $_2$O ↑

Air (O) heat

Above mixture +Aloe Vera + lemon juice Calcination

Conversion of Sn into SnO_2 with removal of carbon as CO and CO_2

SnO_2 + C **SnO_2 +Sn / SnO + CO↑**
CO2↑

Air (O) & heat

Above mixture +Aloe Vera + lemon juice Calcination	**Complete conversion of Sn into SnO_2**

SnO_2 +Sn / SnO → **SnO_2**

Air (O) & heat + carboneous mater

5.2. Standardization of the Bhasmas

While the basic aim of this research work was to understand the intricacies involved in the preparation of these Bhasmas and to understand these materials in depth, the study has also helped us to standardization of the procedures to certain extent. While the basic step towards the standardization could be the use of various analytical techniques that have been used in this investigation so as to monitor various changes and the progress of the reactions. Use of simple techniques like pHmetry for the Kapardika Bhasma, carbon analysis (carbon content determination) for Vanga Bhasma and powder XRD for the Jasad Bhasma for quality control during the preparations could be the first step towards the standardization of the products and the processes in general.

The chemical analysis of the raw materials followed by appropriate choice of the raw materials and various reactants is necessary as these materials govern the presence of various metals in lower concentrations in the Bhasmas (metals other that Zn in Jasad Bhasma, other than Sn in Vanga Bhasma and other than Ca in Kapardika Bhasma).

5.3. Chemistry and Ayurved

Although the first impression that one can gather on Ayurvedic texts is that the preachers and practitioners of Ayurved were highly knowledgeable medical practitioners, a detailed investigation of these texts reveal that they were even more efficient chemists. They were well versed with the art of synthesis of materials and the production of the materials at various scales. The details of the processes including extractions of metals from ores, their physical properties, identification parameters, purification methods, have been well documented. Processes of quenching, trituration, calcination, and distillation were commonly used during the synthesis of materials. This wealth of information about the materials, their properties and their use, the chemistry involved in their preparations is being explored since a period, which dates before the Alchemists era.

The use of the Bhasmas as potential drugs is facilitated mainly because of two reasons. Firstly because of the fact that these materials are being routinely used as effective drugs for centuries and secondly due to the fact that these drugs do not

show any noticeable side effects with the recommended doses. Our study involving the characterization of the Bhasmas prepared by us as well as the Bhasma samples available in the market also suggests that the Bhasmas shall be effective drugs without any noticeable side effects if they have been synthesized strictly as per the methods mentioned in the Ayurvedic literature. It needs to be ensured that the Bhasmas that are being administered to the patients are not the intermediates (whose effect on the patients is not known) but the final products, which do not contain the metal in the metallic form even at low concentrations.

The main question that can be raised after understanding these drugs is regarding the specifications mentioned in the lengthy procedures for the preparation of the Bhasmas. It is realized that the specifications of the number of steps to be followed (related to the trituration and calcinations) which often make these procedures lengthy need to be followed to ensure the complete conversion of the intermediates into the final products.

The importance of the various metals to human body was well known to the Ayurvedic researchers. They also knew that the metals cannot be administered to the patients as such, being toxic and so they need to be converted to a non-toxic form which can be used as the drug. Their sound knowledge about the chemistry of these materials reflects in the methods that they prescribed for preparation of the Bhasmas. The possible routes to convert the metals into the respective forms (compounds) were explored. Although various chemical processes were routinely used during the preparations, it seems that they lacked the knowledge of

precipitation. Hence the common methodology used was the conversion of the metals into oxides by reacting with mercuric or arsenic sulfide, which takes place readily at elevated temperatures, followed by further oxidation to their respective oxides. Other methods involved use of the combustion of herbal products, which brings about the oxidation. This method is comparable to the combustion synthesis procedures carried out using a variety of organic fuels to obtain metal oxides.

5.4. The holy metals!

The bulk of living mater consists of hydrogen, nitrogen, oxygen and sulfur. The concentrations of these elements in the human body can be expressed in grams per kilogram. These elements are required in gram amounts per day as essential nutrients in humans. The macrominerals sodium, magnesium, potassium, phosphorous, magnesium, chlorine and calcium serve as the essential constituents in tissues and body fluids. Although their requirement for human body is comparatively in lesser concentration, still it can be expressed in terms of fractions of kilograms per day. Other then these main components, there is a list of metals including manganese, zinc, iron, vanadium, chromium, which have been, recognized as essential elements in various concentrations.

Rasashastra has explored the use of various metals available, which are required for human body as nutrients or as drugs for various disorders. Other than Ca, Sn and Zn (whose Bhasmas we have studied), a variety of metals are routinely

used in the form of Bhasmas as drugs. Bhasmas of gold, silver, lead, copper, iron, mica, diamond, and brass are commonly used. Their preparation procedures involve use of formulations containing mercury (Kajjali) and arsenic (Haratal) extensively. The use of these metals as drugs and the toxic metals during the preparations raises alarms elsewhere in the world. Unless the chemistry behind the Bhasmas is known, they are truly 'Holy metals' in disguise!

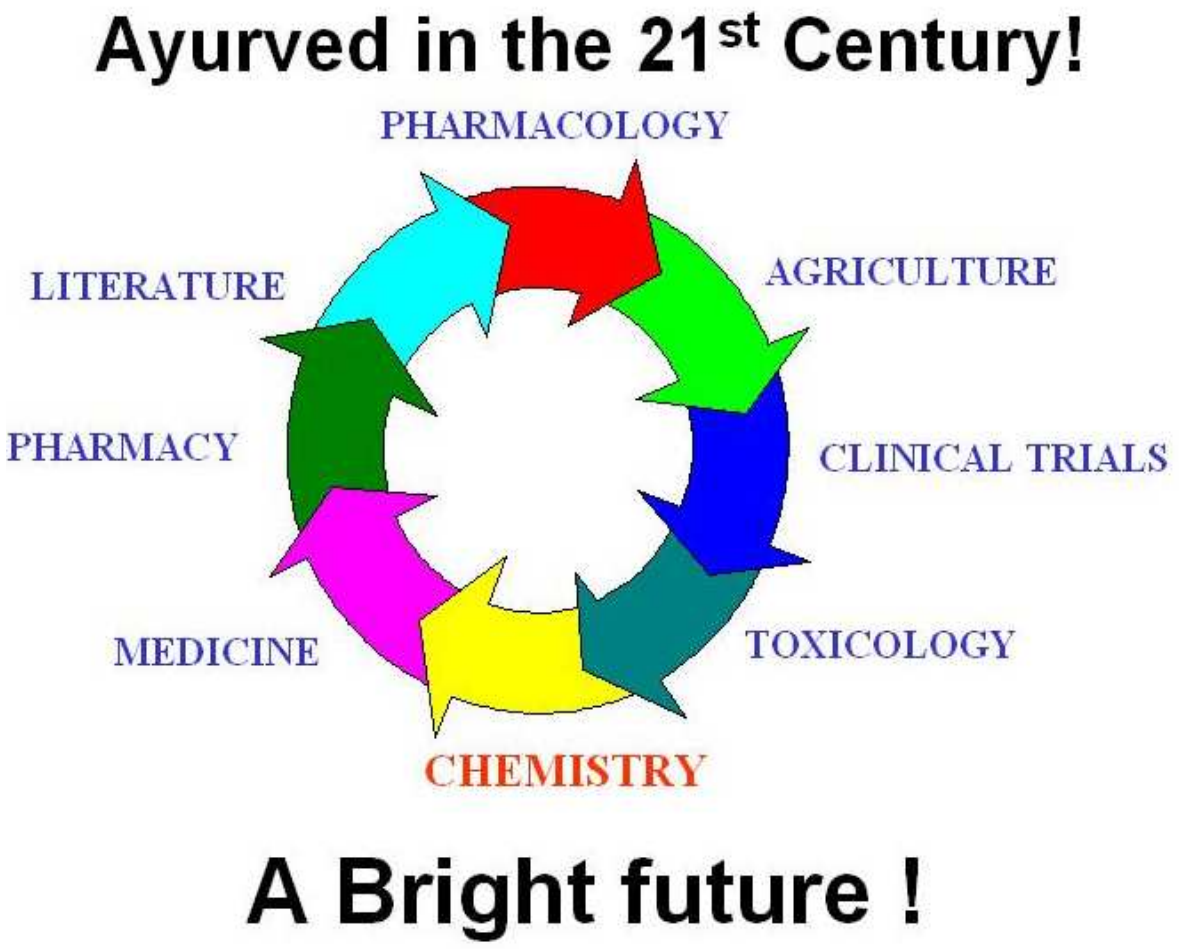
5.5. The road ahead…

A similar kind of systematic study on the other Bhasmas would help in knowing those materials in depth. After the detailed characterization of the Bhasmas, the obvious question that arises is that can these materials be prepared in a chemical laboratory by simpler modern techniques which would enable to make their preparation cost effective and less time consuming.

The characterization of the Bhasmas needs to be followed by evaluation of the toxicity levels of these drugs. This can be followed by clinical investigation of the patients who are being administered these drugs. The study is not required to prove the medical efficacy of these drugs that are been routinely used for centuries but to take them forward to the world the way modern medicines are. A systematic physico-chemical study of the Bhasmas followed by the toxicity measurements and clinical investigation would help in protecting the rich heritage of India and

establish a solid platform in the global market.

5.6. Ayurved in the 21st Century



A systematic study on Ayurved demands a multidisciplinary approach. With various branches of science contributing in the study, Ayurved will definitely have a bright future.

Database Management

- A proper documentation of the ancient Ayurvedic texts in Sanskrit and regional languages.
- Appropriate translation of the texts to English.
- Forming a database of the research reports available on Ayurved from various streams.

Agriculture and botany

- Cultivation of medicinal plant on a large scale to ensure availability of raw materials.
- Identification of the correct species of herbs and study of their chemical and physical properties.
- A detailed study on the bio-diversity in plants.

Pharmacology

- Evaluation of various physico-chemical properties of the materials from the medicinal point of view as described in Indian and British Pharmacopeia.
- Comparative study of the drugs with the similar drugs used in modern medicine.
- Setting up quality control measures for these drugs from using pharmacological parameters.

Toxicology studies and clinical trials

- Toxicological studies to establish the lethal doses for the medicines.
- The effect of drug administration on the various organs can be monitored.
- Clinical trials can establish the efficacy of the drug by monitoring the parameters as done by modern medicine.
- A comparative study of the Bhasmas with those parallely used in the modern medicine.