ORIGINAL RESEARCH ARTICLE EXPERIMENTAL

Preparation and characterization of mercury-based traditional herbomineral formulation: *Shwas kuthar rasa*

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ABSTRACT

Background: *Shwas kuthar rasa* is a prestigious and potential herbomineral formulation of Ayurveda tested on 100 years of time scale for the treatment of asthma, allergy, and other respiratory problems. However, there is a lack of scientific work on *Shwas kuthar rasa.* **Objective:** To prepare and physicochemically evaluate mercury-based *Shwas kuthar rasa* herbomineral formulation of Ayurveda for asthma and allergy. **Materials and Methods:** *Shwas kuthar rasa* was prepared as per Ayurvedic text and characterized by various modern analytical techniques, viz., transmission electron microscopy (TEM), X-ray diffraction (XRD), far infrared (IR) spectroscopy, fourier transform IR spectroscopy, energy dispersive X-ray analysis, and inductively coupled plasma-mass spectroscopy. **Results:** Study clearly revealed that prepared *Shwas kuthar rasa* formulation shows several crystallites agglomerate into a single particle. It yields submicron size particle structure (1.22 μ) with TEM analysis. The usage of mercury in the formulation found in the form of mercuric sulfide (HgS) and reaching to nanocrystalline (31–56 nm) size by XRD analysis. **Conclusion:** The present study indicates *Shwas kuthar rasa* is nanocrystallite with submicron size particle. Trituration of *Kajjali* helps in the formation of HgS and increases the crystallinity in the formulation.

Key words: Formulation, herbomineral, mercury, *Shwas kuthar rasa*

INTRODUCTION

Rasa Shastra means the science of mercury but also refers to the preparation of minerals/metals suitable for the body so that they can be used as medicines.[1,2] Minerals such as mercury and arsenic are considered toxic[3] but with proper *shodhana* (detoxification) process, they can be turned into therapeutic medicines. The formulation was prepared and processed properly, the mercury balances all three *Doshas*, has a soothing effect and protects the body from diseases and aging process. It nourishes all vital body parts and increases the strength of the eyes.[4,5] It is a *vrishya* (aphrodisiac), *balya* (tonic), *rasayana* (rejuvenating), *vrana shodhana, ropana* (wound cleaner and wound healer), and *krimighna* (anthelmintic and antimicrobial).[6] Mercury compounded with herb, the mercury heightens the medicinal potential of the particular herb. Mercury is said to give a firm physique, a stable mind, and considered to be the destroyer of diseases.[7] Herbs were used in the combination with *rasa*, the efficacy spectrum of herbs increases to a

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great extent enabling them to treat complicated diseases. *Rasa Shastra* has been placed with great importance in Ayurveda.

Shwas kuthar rasa a reputed preparation of Ayurveda valued for the treatment of asthma and allergy is a herbomineral formulation contains herbs, purified *Aconitum ferox* (*Aconite*), *Piper longum* (long pepper), *Piper nigrum* (black pepper), and *Zingiber officinale* (ginger), and minerals that is, *parada* (mercury), *gandhaka* (sulfur), *tankana* (borax), and *manahsila* (arsenic disulfide) in purified form as per Ayurvedic text.[8] *A. ferox* inhibited the biosynthesis of leukotriene B4 in bovine polymorphonuclear leukocytes.[9] *P. longum* have a traditional claims of Ayurveda for antiallergic and antiasthmatic activity[10] *P. nigrum* suppressed and reduced the infiltration of eosinophils, hyper responsiveness, and inflammation in mice.[11] *Z. officinale* are capable of inhibiting allergic reactions and is useful for the treatment and prevention of allergic diseases[12] Review of literature revealed that *Shwas kuthar rasa,* apart from treating asthma and allergy, is used for the cure of cough, laryngitis, tuberculosis, unconsciousness, mental disorders, comma, chest burn, and heart diseases.[8] None of the work presents the elemental and structural characterization of herbomineral formulation which is an essential requirement to discuss the therapeutic value of mercurial preparations. The present study aims to study the composition and the structure of *Shwas kuthar rasa* a herbomineral formulation using various techniques, viz., transmission electron microscopy (TEM), X-ray diffraction (XRD), far infrared spectroscopy (FIR), fourier transform infrared spectroscopy (FTIR), energy dispersive X-ray analysis (EDAX), and inductively coupled plasma-mass spectroscopy (ICP-MS).

MATERIALS AND METHODS

Plant materials

The dried fruit of *P. longum and P. nigrum*, root of *A. ferox,* and rhizome of *Ginger officinale* were obtained from Amruth Kesari Herbs, Bangalore in February. The drugs were identified by Prof. K. Prabhu, Botanist, S. C. S. College of Pharmacy, Harapanahalli (Karnataka), India. The herbs voucher specimens (No: SCSCOP/Ph.cog/Herb-652, 717, 691, and 572, respectively) were deposited in the botany herbarium of the institute.

Preparation of *Shwas kuthar rasa*

Shwas kuthar rasa was prepared using herbal and mineral ingredients as prescribed in Ayurvedic text,[13] initially detoxifying the *parada* (mercury), *manahsila* (arsenic disulfide), *gandhaka* (sulfur), *tankana* (borax), and *A. ferox* (vastanabha) as per given Ayurvedic text. Equal quantities of *shodhit* (detoxified and pure) *parada* and *gandhaka* were taken (1:1) in a stone mortar in reference amount, triturated for 40 h or until it attained the required *Kajjalabha* (blackish appearance) and *Nishchandra* (lusterless) state, that is, shining of *parada* is lost. This state of formulation is called *kajjali*.[14] *Kajjali* was then triturated, with the reference amount of powdered *manahsila*, *vastanabha, tankana,* and *trikatu* (equal part of black pepper, long pepper, and ginger), for 72 h to obtain fine powdered herbomineral formulation *Shwas kuthar rasa* and it was allowed for drying and stored in glass jar.

Structural and physicochemical characterization

Shwas kuthar rasa was subjected to evaluation for physicochemical characters such as loss on drying, ash value, and acid insoluble ash[15] followed by TEM (Philips, CEM, CM-12) was used to study the particle size of *Shwas kuthar rasa* whereby a beam of electrons transmits through an ultrathin specimen, interacting with the formulation. As the beam passes through, an image was formed from the interaction of the electrons transmitted through the specimen, the image was magnified and focused on fluorescent screen, on a layer of photographic film (SAIF, Chandigarh, Punjab).

The powder XRD patterns of the *Shwas kuthar rasa* were recorded on X'pert pro panalytical X-ray diffractometer with CuKα radiation (λ =1.5406 A°) operating at 45 KV and 40 mA for the angle (2θ) ranging from 5° to 50° at a scanning rate of 3°/s. A representative portion of *Shwas kuthar rasa* was placed in an alumina crucible, and the temperature was varied from 40°C to 400°C. EDAX (EDAX Inc., Mahwah, NJ, USA) attached to TEM (CEM, CM-12) was used for the detection of various elements in *Shwas kuthar rasa* (SAIF, Chandigarh, Punjab).

For the quantitative determination of heavy metals in *Shwas kuthar rasa* in parts per million (ppm), an ICP-MS, PerkinElmer ELAN-6000 was used. The infrared (IR) spectrum in the low- frequency region (50–400/cm) was recorded on a Bruker IFS 66 V/S vacuum Fourier transform interferometer; whereas the spectra from 400 to 4000/cm region were recorded using FTIR spectrophotometer (Spectrum RXI, PerkinElmer). For IR spectra, powdered samples were mixed in KBr to make translucent pellet and spectrum was recorded (SAIF, Chandigarh, Punjab).

RESULTS

Evaluation of *Shwas kuthar rasa* for organoleptic characters revealed that preparation possess no metallic sound, it is black colored, tasteless, and odorless with no coarse particle. The data of loss on drying, ash value, and acid insoluble ash values were recorded [Table 1].

Modern analytical techniques were used to observe the effect of the procedure employed in processing of *Shwas kuthar rasa*. Particle shape and size from the TEM photograph of *Shwas kuthar rasa* shows spongy structure with the irregular particle size lying in the submicron range [Figure 1]. From the image, it is clear that nanosize crystallites are agglomerated giving rise to micro sized particles with the loss of grain boundaries. These studies confirm that *Shwas kuthar rasa* is nanocrystallite with submicron size particle (1.22 μ).

XRD pattern of *kajjali* [Figure 2a] shows the peaks due to the presence of free sulfur, mercuric oxide, and mercuric sulfide (HgS) (Joint Committee on Powder Diffraction Standards [JCPDS] file number-20-1227, 01-0896, and 02-461, respectively) while the XRD pattern of *Shwas kuthar rasa* [Figure 2b] shows the peaks due to major presence of HgS (JCPDS File number-02-461), mercuric oxide (JCPDS File number-01-0896), and very low intensity of sulfur (JCPDS file number-20-1227). No extra diffraction peaks were observed in case of final *Shwas kuthar rasa* formulation confirming that while in the initial stages of the processing of the formulation free sulfur is present in significant amount; however, after trituration process, the major amount of HgS and mercuric oxide remains in the product. The diffraction peaks in the XRD pattern of *Shwas kuthar rasa* corresponding to HgS become sharper and intense compared to *Kajjali* sample as well as some new peaks appeared due to HgS, which were not present in the *Kajjali* sample. This observation confirms that the trituration of *Kajjali* helps in the formation of HgS and increases the crystallinity in the sample. The crystallite size was calculated from XRD pattern following the Scherrer equation $t = \lambda \times 0.9/(\beta \times \text{Cos}\theta)$. Here "t" is the crystallite size for (h k l) plane, λ is the wavelength of the incident X-radiation (CuKα [λ =1.5406 A°]), β is the full width at half maximum (FWHM) in radians, and θ is the diffraction angle for (h k l) plane.[16] It is notable, here, the FWHM in case of *Kajjali* is high in comparison to the finally prepared *Shwas kuthar rasa* confirms that the size of the crystallite increases. It is obviously due to the trituration process of the *Kajjali* sample. Thus, the XRD study concludes the presence of nanocrystalline structure of the drug.[17]

In addition, the metal Hg and As used as ingredients, other metals like calcium is also expected in the drug that enters in it during its trituration process while carryout the detoxification of individual herbs and metals. EDAX has been used to detect the presence of elements in considerable amount,[17] whereas ICP-MS was used to detect elements Hg and As in trace amount. Chemical compositions of *Shwas kuthar rasa* using EDAX and trace metal composition of *Shwas kuthar rasa* using ICP-MS have been listed [Table 1]. Abundance of C (31.24%), N (12.40%), and O (42.63%) in the drug was observed which is obviously from the herbs used in the preparation of the formulation. Ca (1.62%) conducive to healthy metabolism and preventive for stomach lesions was also found to be present in the final *Shwas kuthar rasa* product.

Table 1: Physicochemical characters of *Shwas kuthar rasa*

Parameters	*Shwas kuthar rasa*
Loss on drying (%)	0.73
Ash value (%)	98.29
Acid insoluble ash (%)	44.58
Particle size (μm)	1.22
Phase identification	HgS and HgO
Element contents (weight %)	C - 31.24, S - 0.89, N - 12.40, O - 42.63, Na - 2.37, Ca - 1.62, Cl - 3.46, H - 4.65.
Heavy metals (ppm)	Hg - 0.94, As - 8.78
Organic macromolecules	9 sharp peaks

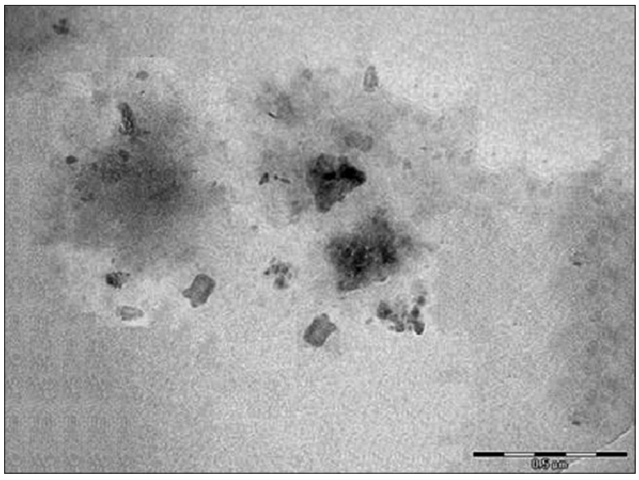

Figure 1: Transmission electron microscopy image of *Shwas kuthar rasa*

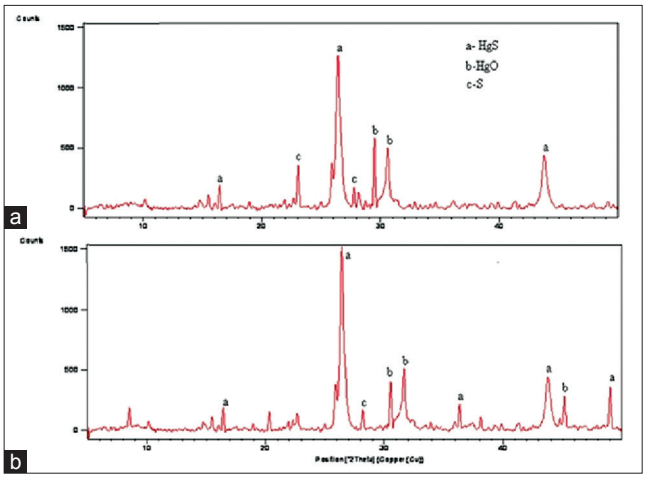

Figure 2: X-ray diffraction pattern of (a) *Kajjali* and (b) *Shwas kuthar rasa*

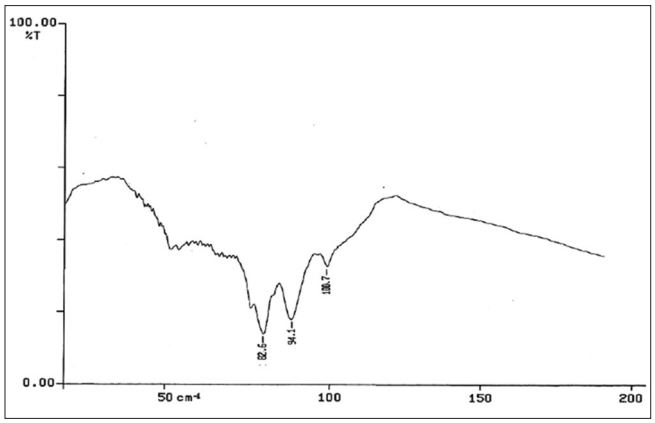

Figure 3: Far infrared spectroscopy spectrum of *Shwas kuthar rasa*

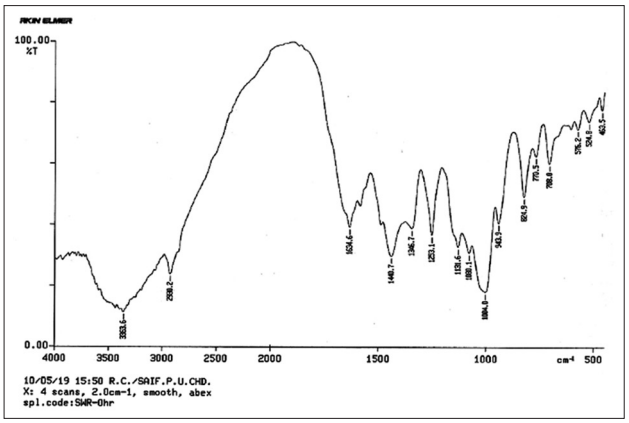

Figure 4: Fourier transform-infrared spectrum of *Shwas kuthar rasa*

Na (2.37%) needed for maintaining normal fluid balance is also present in the final product [Table 1]. These elements (Ca, Na, and S) act as additional supplement improving the curative properties of the formulation.[18] Other elements such as H (4.65%) and Cl (3.46%) were also found in the formulation. Concentration of heavy metals was found 0.94 ppm for Hg and 8.78 ppm for As, which were well within the safe limits recommended by WHO.[19,20] Thus, the additional element present in the drug is clearly due to the botanical origin. It is notable that the proportion/concentration of mercury in *Shwas kuthar rasa* does not seem to follow a consistent trend, and some of mercury is certainly lost during the preparation through direct trituration process. This raises the safety concerns pertaining to the use of mercury and may require additional work on the processing technique employed in the preparation of *Rasas*.

FIR spectrum of *Shwas kuthar rasa* in the region from 50 to 400/cm was studied [Figure 3]. Crystalline HgS is known to have absorption at 83, 92, and 100/cm and their presence in the present FIR spectra indicate that *Shwas kuthar rasa* is essentially HgS. This observation supports the XRD analysis. FTIR spectrum of *Shwas kuthar rasa* in the region from 400 to 4000/cm is shown [Figure 4]. There are fairly sharp peaks at 708, 1080, 1131, 1253, 1346, 1440, 1634, 2930, and 3363/cm which indicate the presence of the organic compounds in the formulation. These arise probably from the usage of the source of herbs. The presence of appreciable concentrations of C, H, O, and N [Table 1] also suggests the presence of organic molecules in the drug. It would not be unexpected if the organic molecules also play an important role in the medicinal properties of these drugs.

DISCUSSION

Macro particle size of the preparation may be attributed to the trituration of detoxified metals, nonmetals, and herbs for a long duration which causes the change in the chemical nature of materials. FT-IR analysis shows the possibility of organic matter in the formulation. This could be due to the formation of organometallic complexes in the drug sample that can sustain even at the high processing temperature of herbomineral drugs. Several significant possibilities and future prospects of the drug could be debated with these results. The macro particle size of the drug matches well with the colloidal size and this suggest the possibility that these colloidal particles are get attached to the human intestine and provide a large surface area thereby increasing the absorption of other nutrients and drugs, which are added to it during the process of preparation or prescribed to the patient along with them.[18] Further, metal ingredients act as the carrier of the herb derived organic matter used during the pharmaceutical processing. In short, metals as a carrier for the organic contents from *A. ferox, P. nigrum, P. longum,* and *Z. officinale* are known to be useful in the treatment of asthma, allergy, cough, inflammation, etc.[21-24] From XRD studies, *Shwas kuthar rasa* concluded that HgS in nanocrystalline range (31–56 nm), in association with organic molecules probably plays an important role in making it biocompatible and nontoxic at therapeutic doses. Other elements present in *Shwas kuthar rasa* act as additional supplement and possibly help in increasing the efficacy of the formulation.

CONCLUSION

Shwas kuthar rasa herbomineral formulation revealed the presence of crystalline HgS associated with several organic macromolecules derived from the herbs as basic ingredients of formulation. In addition, several elements were also found in varying concentrations, which bioavailability enhanced and thus responsible for therapeutic value addition of *Shwas kuthar rasa.*

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Conflicts of interest

There are no conflicts of interest.

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