Role of Ghandhak Jarana in the preparation of Malla Sindura - A Kupi pakwa rasayan

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ABSTRACT

Background: Mercurial preparations known by the name of Kupipakwa Rasayan are prepared in a glass bottle by adopting gradual temperature gradient. Aims and Objectives: To elucidate the jarana process (role of temperature and time) process in kupipakwa rasayan. Materials and Methods: Two sample of Malla Sindura was prepared by heating a mixture of Parada (mercury), Gandhaka (sulfur), and Malla (arsenic) in a Kupi (glass bottle covered with mud smeared cloth) placed in an electric muffle furnace at two different temperature and time duration ie 24 and 48 hrs. Final product deposited at the neck of bottle was collected as Malla Sindura. Results and Conclusion: Three batches of each sample of Malla Sindura was prepared by adopting gradient temperature pattern for 24 hrs and 48 hrs i.e., 8 h of mild (120-250°C), moderate (250-450°C) and strong heating (450-600°C) for SDMS and 18 hrs of mild (140-250°C), 22 hrs moderate (250-450°C) and 8 hrs strong heating (450-630°C) for LDMS each. Average yield of Malla Sindura in two samples is 151.33 g (37.83%) and 237.7 g (59.42%) using 400 g Kajjali in each batch respectively.

Keywords: Characterization, Kupipakwa Rasayana, Malla Sindura .

INTRODUCTION

Rasashastra, a branch of Ayurveda has been evolved on the principles of converting metals and minerals into their bio-available forms and thus making them therapeutically viable [1]. Recently, Rasashastra formulations viz. mercurial preparations have been in limelight for concerns over their quality and safety [2]. Mercurial preparations known by the name of Kupipakwa Rasayan are prepared in a glass bottle by adopting gradual temperature gradient. Malla sindura [3], Rasasindura [4], Makaradhwaja [5] and SwarnaVanga [6] are few often used medications prepared by above method. Malla sindura is a well-known drug for its use in various diseases like bronchial asthma, bronchitis, rheumatoid arthritis, diabetes mellitus, etc [7]. As far as treatment with Rasasudhidhis is concerned, along with Rasasindura, it forms an indispensable part of Ayurvedic repertory for physicians. Malla sindura is prepared by heating a combination of Parada (mercury), Gandhaka (sulfur) and Malla (arsenic trioxide) in a glass bottle. Jarana [8] is a one of most important and intermediate process of Kupipakwa Rasayan on which total therapeutic efficacy and safety depends. Historically the detailed description of Gandhaka Jarana and its effects has been found mentioned in Rasendra Chintamani (12th A.D.). Before this a few references regarding Gandhaka Jarana are found in Rasarana, Rasendra Cudamani and Rasa Prakash Sudhakara [9]. Later on a detailed description with regards to importance necessity and affection is found mentioned. It is process in which Gandhika; Abhraka Satva [10] etc should be treated with Parada different means like heating according to different methods preparation in different Yantra like Valuka Yantra, Kacchopa Yantra etc. It is a thirteenth Sanskara amongst Ashhtadasha Sanskara of Parada which are mainly intended for Lohasiddhi or Dehasiddhi. According to methods of preparation when Gandhika is burnt or subjected to treatment in the presence of mercury in Valuka Yantra, Kacchopa Yantra etc are called as Gandhaka Jarana. In the process of Gandhaka Jarana when free sulfur is completely evaporated/burnt then that state is known as Gandhaka Jarana. If sealing is done and extra heat is given then Rasasindura will be prepared. So in the preparation of Kupipakwa Rasayan, first stage is Gandhaka Jarana and next stage is preparation of product. The safty and therapeutic efficacy of the heavy metals specially mercury increases with the process of Jarana, more the time duration and amount of Gandhika [12] more will be
efficacy and safety of the final product i.e. HgS. Temperature also play a vital role in Kupipakwas Rasayan. According to modern science Sulphur reduces the toxic action of mercury\[^{13}\]. According to Goodman, Gillman pharmacology mercury readily forms covalent linkages to Sulphur. When Sulphur is in the form of sulphhydril group, divalent mercury replaces the hydrogen atom to form mercaptides, X-Hg-Sr and Hg(SR)2 where X is an electronegative radical and R is protein. Sulphhydril compounds have long been called Mercaptes because of their ability to capture mercury. The affinity of mercury for thiols provides the basis for treatment of mercury poisoning by such agents as Dimercaprol and Penicillamine. The minerals and metals when processed with addition of Sulphur or Sulphur containing mineral, immediately get converted into sulphides and most of these are non-toxic. Cu, Pb, Sn, Zn, As etc; are highly toxic if they are in oxide form, but their sulphate compounds are nontoxic or least toxic\[^{14}\]. In the Ayurvedic pharmacopoeia on the other hand, mercury predominantly used in the form of sulphides. It is indeed strange that a country, where this metal was first harnessed into the service of medicine, should have chosen an insoluble and possibly an inert salt for therapeutic uses. So, investigations are being carried on to determine whether this salt is at all made soluble under ordinary physiological conditions in the gut and whether the mercury ion liberated from this so called inert combination can be utilized by the tissues.

**MATERIALS AND METHODS**

**Materials**

Raw materials viz. Hingula, Gandhaka and Malla (Somali) including required equipment such as Kupi (glass bottle coated with seven layers of mud-smeared cloth) were collected from Pharmacy, Gujarat Ayurved University, Jamnagar and vertical electric muffle furnace (EMF), iron spoke, and big iron rod were collected from Dept. of Rasashastra and Bhaishajaya kalpana, Gujarat Ayurved University, Jamnagar.

**Method**

Two sample of Malla Sindura was prepared by using the same classical reference ie SDMS (Short duration and upto 600 °C and 24 hrs) and LDMS (Long duration and upto 630 °C and 48 hrs). Both the sample of Malla sindura was divided into three stages, namely, preparatory stage, principal stage and concluding stage.

**Preparatory stage**

Purification of raw materials

Hingula was purified by triturating it with lemon juice to convert into fine powder. This shodhit Hingula\[^{15}\] was further used for Parada Niskasan process to get Hingulottha Parada\[^{16}\]. Gandhaka was purified by subjecting it to Dhalana (melting and pouring in media) process in Godudha (cow’s milk) for 3 times \[^{17}\]. Purification of Malla\[^{18}\] was done by Swedana (Boiling in liquid bath) method in a Dolayanta (heating under liquid media), whereby small pieces of Malla were tied in a four folded cotton cloth and suspended in Dola Yanta containing Cow milk for one Yama (3 hrs). After self-cooling of the Dola Yanta, pieces of Malla were obtained and washed carefully with hot water and dried.

**Preparation of Kajjali (fine black powder)**

Shuddha Parada and Shuddha Gandhaka were taken in 1:1 ratio and triturated in an iron mortar until the whole mixture was converted into Kajjali\[^{19}\]. Then Shuddha Malla was added half the quantity of Parada to the Kajjali and again it was triturated upto uniform mixing. Bhavana of Kumari Swaras was given for 6 hrs to make a uniform black lustreless powder. This Kajjali was filled in the Kupi and placed in the center of EMF in such a way that it could receive equal distribution of heat.

**Principal stage**

Both sample of Malla sindura was prepared in three batches of 400 g Kajjali by following Kramagni\[^{20}\] (gradual intermittent increase in temperature) pattern. For 1\(^{st}\) Sample the temperature pattern adopted was 8 hrs of mild (120-250°C), 8 hrs of moderate (250-450°C), and lastly 8 hrs of intense (450-600°C) heating and for 2\(^{nd}\) sample the temperature pattern adopted was 18 hrs of mild (140-250°C), 22 hrs of moderate (250-450°C), and lastly 8 hrs of intense (450-630°C) heating.

Different observations like mild white fumes, thick yellow fumes, blue flame, etc. were recorded during both the process. Accumulated sulfur at the neck was burnt by rubbing Tapta Shalaka (red-hot iron rod) repeatedly with the neck of bottle. Confirmative tests like cessation of flame, red hot appearance of the bottom, coin test etc. were ascertained before corking (600°C) and the temperature was increased to facilitate sublimation of the final product at the neck. The bottle was removed from the furnace after self cooling.

**Concluding stage**

Kupi was cautiously removed from EMF. The layers of wrapped cloth were carefully scraped, bottle was broken with the help of thread dipped in kerosene oil and the product was collected at the neck. The whole process was repeated for both samples in three batches.

**Analytical Profile**

Chemical characterization of finished sublimated product Malla Sindura was subjected to various organoleptic and quantitative parameters such as texture, color, taste, odor, pH\[^{21}\], ash value\[^{22}\], acid-insoluble ash, water soluble ash, loss on drying\[^{23}\], and percentages of mercury\[^{24}\], sulfur\[^{25}\] and arsenic.

**OBSERVATIONS AND RESULTS**

Different stages during the process like fuming, melting of Kajjali, flaming, confirmative test for completion of product formation (flame disappearance, Shita Shalaka test, copper coin test, red hot appearance of the bottom, etc.) were observed and recorded (Table 1). Malla sindura collected at the neck of the Kupi in both the sample in all three batches were weighed and calculated for the percentage of yield (Table 2). Results of various organoleptic and physicochemical analyses are shown in Tables 3 and 4.

**DISCUSSION**

The Rasashastra texts claim that Parada treated with Gandhaka Jarana process becomes highly potentiated and many pharmaceutical and therapeutic properties. This potentiating depends on the proportion of Sulphur made burnt during Jarana process. According to textual references of Ayurveda Prakasha\[^{26}\] and Rasatarangini\[^{27}\], Sadagunahalijarita parade is claimed to be much more effective than Samguna or Dwi-guna balijarita parade. Not only have this some texts claimed that without sadagunahalijarana, parade does not develop a disease curing capacity. Hence from therapeutic point of view it is essential. Kupipakwa Rasayana is also categorized on the basis of proportion of Gandhaka. Different classics like Rosendrachintamani, Yogaratnakara, Ayurveda Prakash and Rasatarangini have mentioned different preparative procedures and proportion of Gandhaka of Kupipakwa Rasayan specially Rosa Sindura. Proportion of Gandhaka varies between classical text from 1:1/4 to 1:6. According to classical texts, more the amount of Gandhaka taken for Rosa Sindura preparation more potent will be the final product. Similar concept is applied in the classical text for duration of heating for Kupipakwa Kalpa preparation.
Duration of heating ranges from 8 hours (e.g. Rasakarpura) to 168 hours i.e. seven days (e.g. Shadaguna Bali Jariita Rasasinadura). Chemical characterisation of different Kupipakwa Rasayan like Rasa Sindura[32], Malla Sindura[30] shows that, it is mercuric sulphide (HgS). It is a well-known fact that temperature and pressure affect molecular structure and ionization potential of elements[33]. Diamond, graphite and coal are the purest carbon compound with similar chemical composition but they differ in physical as well as chemical properties due to difference in temperature and pressure during their formation. Similarly Kupipakwa Rasayan prepared from different methods and different time duration are basically HgS but they are different in their physical, chemical as well as therapeutic properties along with its safty aspects.

It is understood that different proportion of Gandhaka and different duration of heating definitely have linear effect on the clinical efficacy of Kupipakwa Rasayan. Based on their vast experience the ancients seers of Rasashastra have emphasized on the difference in therapeutic potential of Kupipakwa Rasayan prepared from different methods. According to Rasatarangini, Rasasindura prepared from equal amount of Gandhaka (Samaguna Balijarita Rasasinadura) is able to cure simple disease, Dwiguana Balijarita Rasa sindura possesses potential to cure Maharoga (7 diseases which are difficult to treat), Triguna Balijarita Rasa sindura can cure Kshaya (tuberculosis) and impotency, Chaturguna Balijarita Rasa sindura is said to have influence to improve memory and intellectual power, Panchaguna Balijarita Rasa sindura has therapeutic potential to cure all diseases and Shadaguna Balijarita Rasa sindura is claimed to have amazing effects in treating dreadful ailments.[32] In other word Shadaguna Rasasinadura is best for therapeutic purpose.

Malla sindura is a Kupipakwa Rasayan formulation which is prepared by heating a mixture of few essential materials in a glass bottle with graded pattern of mild, moderate and intense heat in an EMF[33]. Various references of Malla sindura are found in classical texts of Rasashastra. The reference[34] selected for present study is from one of the dedicated books of Drugs and Cosmetics Act, 1945[35]. Furthermore, studies on pharmaceutical standardization and characterization of other Kupipakwa Rasayanaviz. Rasa sindoree[36], Makaradhwaja[37] Swarna Vangal[38] and Malla Sindura[39] have been reported by various scholars. The following classical guidelines, Shodhana (purification) of Hingula and parada Niskasan, Gandhaka Shodhana and Malla Shodhana was done as a preliminary mandatory process. Shodhana adds to therapeutic activity and curtails potential adverse effects of the purified drug[40]. All the ingredients of the formulation were triturated to form an admixture of fine powder, i.e., Kajjali. Ten layers of cloth soaked in clay were wrapped over the bottle to strengthen the bottle and protect it from breaking during the process. Kajjali was filled up to 1/3rd of the bottle which was 400 g, and kept constant for both the sample to all the three batches each.

Graded heating pattern in Kupipakwa is essential aspect to get the therapeutic effect of product. As the transformation of Kajjali to Malla sindura is dependant on time and temperature pattern. The Agni pattern is designed for both the sample as Mridu Agni, Madyamya Agni and Tivra Agni for SDMS and LDMs are 120-250°C, 250-450°C and 450-600°C and 140-250°C, 250-450°C and 450-630°C respectively. There is no major changes observed in the Mridu Agni except fumes. The major chemical reaction occur in Madhayaam Agni stage so the time duration for LDMs is longer. In Tivra Agni stage only product sublime and collected at the neck of the Kupi. The time duration is also design for SDMS sample is 8hrs each but Sample LDMs the duration of time is changed for each stage i.e 18 hrs, 22 hrs and 8 hrs. Flame appears at the neck of Kupi indicating burning of excess sulfur at around 470°C and 525°C respectively. The time duration of flame are 70 mints and 93 mints respectively. Deposition of sulfur around neck of Kupi was cleared by insertion of hot iron rod repeatedly. Once the flame subsides, observations like complete cessation of sulfur fumes, Sheeta Shalaka test (insertion of iron spoke in the bottle to confirm complete burning of free sulphur) were undertaken. When Sheeta Shalaka appears sticky and black, indicates the presence of sulfur, but when it is dry and reddish orange in color, test is said to be positive indicating completion of the process. Copper coin test (copper plate is placed at the mouth to detect the free sulfur or mercury) is done to test the absence of free sulfur. If, mercury particles adhere to copper coin, it decolorizes coin and is said to be positive. Red hot appearance of the bottom is also a confirmative test as an indication of complete formation of compound. Thereafter, corking over the mouth of Kupi is done to ensure the sublimation of product followed by increasing the temperature [Table 1&2]. Optimum heat is necessary at later stages of Kupipakwa because low temperatures reduce chances of sublimation whereas high temperature can lead to dissociation of compound. Arsenic has a boiling point of 465°C. It sublimes at 193°C and melts at 312°C. So by the time complete sublimation of HgS is achieved, most of the arsenic is lost in form of vapors and left in traces in the finished product which is also the optimum quantity of arsenic required for desired therapeutic effect. This not only significantly reduces the toxicity of Malla Sindraba but also make it useful in treating a majority of ailments.

Dark red colored Malla Sindraba collected at the neck of Kupi was collected judiciously after self-cooling of EMF on the following day. The complete process was repeated in two more batches to each sample for the development of standard operating procedure. On an average 37.83% and 59.42% of Malla sindura was obtained in three batches [Table 3]. On trituration, Malla sindura turned into bright red color and was tasteless [Table 4].

CONCLUSION
Two Sample of Malla sindura were prepared by adopting two gradient temperature pattern for 24 and 48 hrs. Average yield in both the sample of three batches are 37.83% and 59.42% respectively. The yield is unexpectedly high by increasing the temperature and duration of Jarana.

REFERENCES
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