



Research Article

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Role of *Gandhak Jarana* in the preparation of *Malla Sindura* - A *Kupipakwa rasayan*

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ABSTRACT

Background: Mercurial preparations known by the name of *Kupipakwa Rasayana* 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 *kuipakwa 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 Rasayana* are prepared in a glass bottle by adopting gradual temperature gradient. *Malla sindura*^[3], *Rasasindura*^[4], *Makaradhwaya*^[5] and *SwarnaVanga*^[6] are few often used medicaments 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 *Rasaushadhis* 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 *Rasarnava*, *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 *Gandhaka*; *Abhraka Satva*^[10] etc should be treated with *Parada* different means like heating according to different methods preparation in different *Yantra* like *Valuka Yantra*, *Kacchapa Yantra* etc. It is a thirteenth *Sanskara* amongst *Ashtadasha Sanskara* of *Parada* which are mainly intended for *Lohasiddhi* or *Dehasiddhi*. According to methods of preparation when *Gandhaka* is burnt or subjected to treatment in the presence of mercury in *Valuka Yantra*, *Kacchapa 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^[11]. The safety and therapeutic efficacy of the heavy metals specially mercury increases with the process of *Jarana*, more the time duration and amount of *Gandhak*^[12] more will be

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efficacy and safety of the final product i.e. HgS. Temperature also plays a vital role in *Kupipakwa 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 sulphhydryl 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. Sulphhydryl compounds have long been called Mercaptans 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(Somal)* including required equipment such as *Kupi* (glass bottle coated with seven layers of mud-smear 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 samples of *Malla Sindura* were prepared by using the same classical reference i.e. SDMS (Short duration and upto 600 °C and 24 hrs) and LDMS (Long duration and upto 630 °C and 48 hrs). Both the samples of *Malla sindura* were 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 *Godugdha* (cow's milk) for 3 times^[17]. Purification of *Malla*^[18] was done by *Swedana* (Boiling in liquid bath) method in a *Dolayantra* (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 samples of *Malla sindura* were prepared in three batches of 400 g *Kajjali* by following *Kramagni*^[20] (gradual intermittent increase in temperature) pattern. For 1st 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 2nd 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 processes. 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 samples 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 pharmacological 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 *Rasatarngini*^[27], *Sadagunabali jarita* parade is claimed to be much more effective than *Samguna* or *Dwi-guna bali jarita* parade. Not only have these some texts claimed that without *sadagunabali jarana*, 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 *Rasendrachintamani*, *Yogaratanakara*, *Ayurveda Prakash* and *Rasatarangini* have mentioned different preparative procedures and proportion of *Gandhaka* of *Kupipakwa Rasayan* specially *Rasa 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 *Rasa Sindura* preparation more potent will be the final product^[28]. 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. *Rasakarpara*) to 168 hours i.e. seven days (e.g. *Shadaguna Bali Jarita Rasasindura*). Chemical characterisation of different *Kupipakwa Rasayan* like *Rasa Sindura*^[29], *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^[31]. 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 safety 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 ancient 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 Rasasindura*) is able to cure simple disease, *Dwiguna 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 Rasasindura* is best for therapeutic purpose.

Malla sindura is a *Kupipakwa Rasayana* 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 scheduled books of Drugs and Cosmetics Act, 1945^[35]. Furthermore, studies on pharmaceutical standardization and characterization of other *Kupipakwa Rasayanas viz. Rasasindura*^[36], *Makaradhwaja*^[37] *Swarna Vanga*^[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 i.e. *Kramagni* in *Kupipakwa* is essential aspect to get the therapeutic effect of product. As the transformation of *Kajjali* to *Malla sindura* is dependent on time and temperature pattern. The *Agni* pattern is designed for both the sample as *Mrudu Agni*, *Madhyama 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 *Madhyama 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 mins and 93 mins 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 Sindura* but also make it useful in treating a majority of ailments.

Dark red colored *Malla Sindura* 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*.

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