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*RESEARCH ARTICLE*

# PHYSICO-CHEMICAL CHARACTERISATION OF 'UTTAMARASASINDURA': A VALUABLE KUPIPAKWARASAYANAFROM 'RASARAJACINTAMANI'

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## Abstract

Ayurveda, literally means the science of life is a comprehensive system of health care of great antiquity, based on experiential knowledge and grown with perpetual additions. Rasashatra(Iatrochemistry)is an integrated part of Ayurveda which is based on *Rasa* (Mercury) and *Rasadravyas*(Mercury related substances). Rasarajacintamani is a compiled textbook of Rasashastra written in Malayalam language by Vadayattukotta K ParameshwaranPillai.*Uttamarasasindura* is explained in this textbook along with different types of *Rasasinduras*. The treatise Rasarajacintamani is reviewed for the specific formulation, *Uttamarasasindura*. Analytical study is needed to evaluate the proper formation and chemical composition of the medicine.Contexual reviewing revealed that *Uttamarasasindura* is a multidimensional remedy of clinical practice.



## Introduction:-

Rasashastrais considered as the ancient science of pharmaceutics.Thebranch has discovered many formulations and wellknownwith theirquicker action in smaller doses and longer stability period. Ayurveda made a land mark in the history of medicine after the development of Rasashatra(Iatrochemistry)by judicious therapeutic administration of Herbo-Mineral preparations with high degree of safety and efficacy.In the classical textbooks of Rasashastra, different forms of mercurial preparations were mentioned.*KupipakwaRasayana*[1] is one among them. *KupipakwaRasayana*or *Sindurakalpana*preparations differs due to their proportion of mercury and sulphur. The concept of *gandhakajarana* [2] (heat treatment given on purified mercury in the presence of purified sulphur) is one of the prime concepts among the *RasashastraAcaryas.Rasarajacinthamani*is a compiled textbook of Rasashastra written in *Malayalam* language by Vadayattukotta K ParameshwaranPillai. This book can be taken as by the virtue of its practical usefulness.*Uttamarasasindura,*also known as*kuppisindura*is explained in this textbook along with different types of*Rasasinduras (eg: Jyothishangarasasindura).* Administration of*Rasasinduras*with different*Anupanas*are also mentioned in this context.The conversion of *'Kajjalirupa Rasa'*(Black sulphide of mercury)to *'Sindura'* is a complex process.Thus occurred product is having action in the deeper*Dhathu*level, also capable offurnishing*Rasayana*effect.*Uttamarasasindura*contain s*Navasadara*(Salammoniac)alongwith*Parada*(mercury)

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and*gandhaka*(sulphur).Three different*Bhavanadravyas*(media of levigation)are also mentioned here.Thus it differs from normal*Rasasindura*. Different*Anupanas*are mentioned for this formulation and explained as*Sakalarogahara*.

For pharmacological and therapeutic assessment of the compound structural and chemical analysis is a necessary part.So in the present study, *Uttamarasasindura* prepared with *AshtasamskaritaParada*was prepared and its physicochemical characterization was carried out by using sophisticated techniques such as X‑ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) ,Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Analysis(EDAX).

## Materials and Methods:-

Test drug *Uttamarasasindura* (URS)[3] was prepared as per the reference from Rasarajacintamani in the departmental laboratory. Raw Material *Parada* (mercury) ,*AmalasaraGandhaka*(yellow sulphur) and *Navasadara*(Sal ammoniac) were purchased from local market, and drugs for levigation like *citrakamoola* (*Plumbagoindica* Linn.),*datura*(*Daturametel* Linn.) and*kumari* (*Aloe barbadensis* Mill) were collected and identified from the Pharmacy of MVR Ayurveda medical college,Parassinikkadavu,Kannur,Kerala.

*Parada* was subjected to *sodhana* by methods of *Ashtasamskara* [4]. *Gandhaka* was processed to *Shodhana* by *kurmaputavidhi* [5] and*navasadara* was prepared as per the classical method of *navasadarasodhana*[6].Processed mercury and sulphur were taken in ratio of 1:1 and triturated in a clean *khalwayantra*.After about 38 hrs of trituration prepared *navasadara*(1/2 part) was added and trituration continued until fine lusterless powder of black sulfide of mercury (*Kajjali*)[7] was obtained.*Kajjali* was levigated with decoction of *citrakamoola* (*Plumbagoindica Linn*.), juices of *datura* (*Daturametel* Linn.) and*kumari* (*Aloe barbadensis* Mill).Levigation was done for 24hrs each with each of the media(Table 01).The fine powder was filled in the specially prepared glass bottle (*LepitaKacakupi*)[8] and heated for 16 ½ hrs. The heat was provided in controlled manner and gradually increasing temperature in conventional *kupipakwa* method using *valukayantra* [9] (Diagram.01). After the desired characteristic features of the preparation was obtained, mouth of glass bottle was sealed, and sand was removed from the neck portion from outsideand allowed for self-cooling. The peak temperature obtained was672.3$^0$ C(Table 02). Sublimed final product was collected from the neck of the glass bottle [Figure 01].It was powdered ,triturated for 3 hrs and used for further analysis [Figure 02] ;labelled as sample URS.Physicochemical analysis were carried out for determination of structural characterization of URS.

**Table 01:-** Showing quantity of ingredients used for preparation of URS.

| Qty. of *Parada:Gandhaka:Navasadara* taken | *Kajjali* obtained | Percentage of Yield | Total qty.of *bhavanadravya* used | *Kajjali*taken for *bhavana* | Qty. after *bhavana* |
|---|---|---|---|---|---|
| 96g :96g :48 g | 236g | 98.3 % | 183 ml | 234 g | 235.5g |

**Table 02:-** Showing details of preparation of URS.

| Qty. of *kajjali*used | Duration of heat given | Peak temperature | Wt.of URS obtained | % of yield |
|---|---|---|---|---|
| 233 g | 16 ½ hrs. | 672.3$^0$ C | 94.5 g | 40.5 % |

## Observations and Results:-

During gradual heating process of *kupipakwavidhi* different stages shown characteristic observations(table 03).The powdered URS was found tasteless, odorless, fine and bright brick red colored powder on physical examination(Table.04).Characterization by instrumental analysis [10] such as X‑ray diffraction (XRD), Fourier transform infrared spectroscopy (FTIR) , Scanning Electron Microscopy (SEM) and Energy Dispersive X-Ray Analysis(EDAX) were carried out.

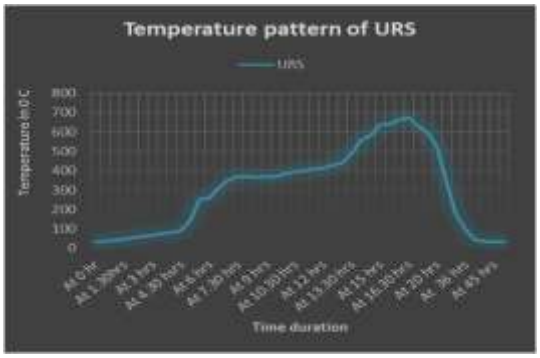
**Table 03:-** showing observations during URS preparation.

| Time duration | Observations |
|---|---|
| At 4 hrs | Pale-white fumes inside the kupi;*kajjali*remained in powder form.;Characteristic smell of *navasadara* arrived |
| At 4.35 hrs | Pale-white fumes came out |
| After 5 hrs | Characteristic odour of *gandhaka* started;melting of *gandhaka* started.confirmed with *seetasalaka*. |
| At 5.50 hrs | Dense white fumes |
| At 6 .10hrs | Yellow coloured fumes with typical rotten egg smell |
| At 6.30 hrs | Mouth of *kupi* blocked with more *gandhaka*;small flames appeared on *taptasalaka* insertion |
| At8 hrs | More blockage and height of the flame increased |
| At 8.15 hrs | Continuos bluish flame from the mouth of *kupi* |
| At 9th hr | Orange-brown deposits at the neck and mouth region with glittering appearance |
| At 10th hr | Boiling *kajjali* found at the base with an orange shade |
| At 10.45 hrs | More Flames and dense yellow fumes on tapta *salaka* insertion |
| At 13th hr | Golden coloured glittering at the neck region |
| At15th hr | Appearance of *balaruna varna*(bright red colour of rising sun) |
| At 16.30 hrs | Fumes and flames Completely stopped,confirmed with copper coin test and corking was done. |

**Table .04:-**showing physico- chemical parameters [11] of URS

| Parameter | Results |
|---|---|
| Colour obtained | Brick red colour(*sinduravarna)* |
| pH value | 7.3 |
| Total ash/Ash value | 10.11 ± 0.08 |
| Acid insoluble ash | 0.48 ± 0.10 |
| Water solubleash | 1.20 ±0.25 |
| Loss On Drying | 7.16 ±0.12 |

**Diagram 01** showing temperature pattern of URS




**XRD analysis**

XRD analysis was done using the instrument RigakuMiniflex 600X-ray diffractometer by using X-ray wavelength, CuK alpha -1.54056 Angstroms and with a power 30kv and15mA.Sample was scanned from 5 degree to 90 degrees (2θ value). Crystallite size (D) is calculated by using Scherer's formula. Red cinnabar (α-HgS), Crystal Structure: Hexagonal, Space group: P3221 is identified by the standard data JCPDS no: 01-080-2192.[table 05,table 06,diagram 02].

Peaks corresponding to (101),(003),(102),(013),(110),(104),(021),(015),(016), (211),(122),(300),(018) planes are obtained for HgS.

**Table 05:-**showing observations and results of XRD analysis for sample URS.

| 2θ (degree) | d value ($A^0$) | Intensity (cps) | Crystallite Size ($A^0$) | | 2θ (degree) | d value ($A^0$) | Intensity (cps) | Crystallite Size ($A^0$) |
|---|---|---|---|---|---|---|---|---|
| 26.45 | 3.36678 | 942.26 | 260 | | 52.72 | 1.73467 | 207.36 | 238 |
| 28.24 | 3.15792 | 312.74 | 224 | | 54.53 | 1.68154 | 247.67 | 221 |
| 31.16 | 2.86844 | 858.96 | 314 | | 58.14 | 1.58526 | 97.11 | 188 |
| 37.69 | 2.38461 | 128.04 | 230 | | 59.31 | 1.55663 | 105.98 | 250 |
| 43.57 | 2.07526 | 316.37 | 252 | | 64.95 | 1.43457 | 101.23 | 258 |
| 44.62 | 2.02911 | 89.03 | 234 | | 69.88 | 1.34486 | 181.19 | 306 |
| 45.78 | 1.97998 | 253.59 | 236 | | 72.28 | 1.30606 | 84.98 | 209 |
| 51.73 | 1.76561 | 238.13 | 283 | | 75.41 | 1.25943 | 91.27 | 275 |

**Table 06:-** Showing identified compound details of sample URS.

| Sample Name | Compound name | Formula | Concentration level | Crystal Structure |
|---|---|---|---|---|
| URS | Cinnabar | HgS | Major | Hexagonal |

**Diagram 02:-** showing XRD data of sample URS

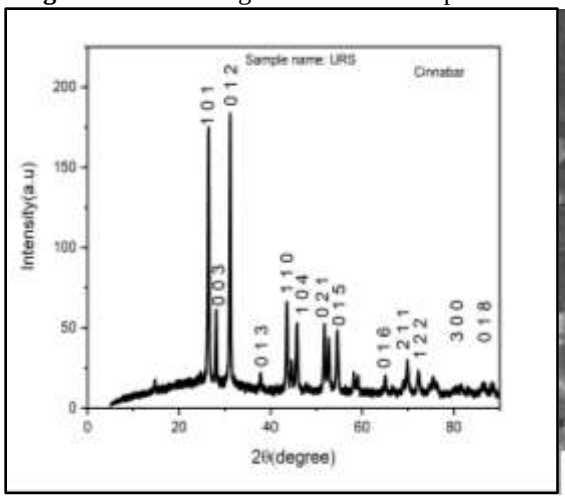
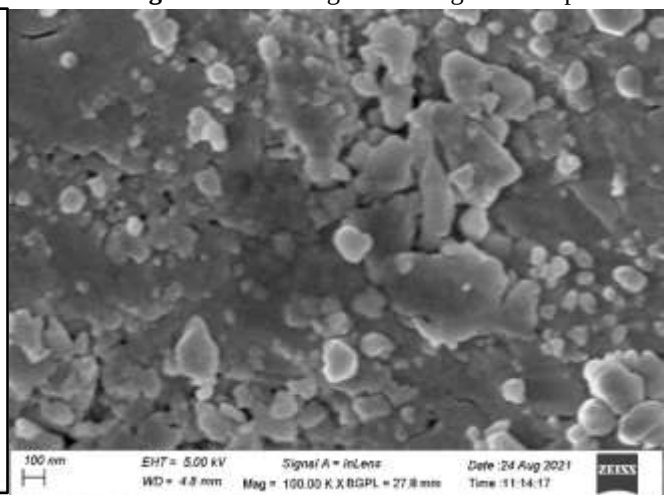



**Figure 03**:-showing SEM-images of sample URS




**SEM Analysis**

The SEM analysis reveals that *Uttamarasasindura* particle size lies in micro-nano sized range and has an average size of 100 nm at 100.00Kx.[Table 07,figure 03]

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**Table 07:-** Showing SEM analysis results of URS sample.

| URS | Magnification | Particle size. | Magnification | Particle size. |
|---|---|---|---|---|
| Area 1 | 50.00K x | 200nm | 2.50Kx | 10 µm |
| | 25.00Kx | 1 µm | 1.00 Kx | 10 µm |
| | 10.00Kx | 1 µm | 500 x | 20 µm |
| | 5.00 Kx | 2 µm | | |
| Area 2 | 100.00 Kx | 200 nm | 5.00 Kx | 2 µm |
| | 75.00 Kx | 200 nm | 2.50 Kx | 10 µm |
| | 50.00 Kx | 200 nm | 1.00 Kx | 10 µm |
| | 25.00 Kx | 1 µm | 500 x | 20 µm |
| | 10.00 Kx | 1 µm | | |
| Area 3 | 100.00 Kx | 100 nm | 5.00 Kx | 2 µm |
| | 50.00 Kx | 200 nm | 2.50 Kx | 10 µm |
| | 25.00 Kx | 1 µm | 1.00 Kx | 10 µm |
| | 10.00 Kx | 1 µm | 500 x | 20 µm |

**EDAX Analysis**

EDAX analysis was carried out in association with SEM.Data shows Mercury and sulphur as the main contents.[Table08,Table09,Table 10,Diagram 03].

**FT-IR Spectroscopy**

Infrared Qualitative Analysis was performed using NICOLET iS50 FT-IR.Used test method was based on ASTM E1252-98(Re-approved 2021) and E573-01(Re-approved 2021).KBr pellet was used for background correction.Sample was mixed with KBr, pelletized and analysed in transmission mode.In the FTIR spectrum of sample URS two peaks are obtained at 3447cm$^{-1}$ and1070cm$^{-1}$.These peaks can be assigned to O-H stretching and O-H deformation.[Diagram 04]

**Diagram 03:-** showing EDAX result of URS sample

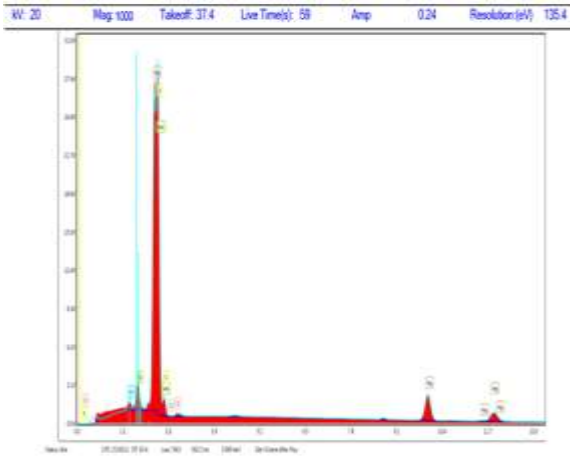


**Table08:-**showing results of EDAX-Area1

| Element | Weight % | Atomic % | Net Int. |
|---|---|---|---|
| Al | 0.73 | 2.83 | 93.13 |
| Si | 0.65 | 2.41 | 101.01 |
| S | 15.92 | 51.68 | 2036.37 |
| Cl | 0.08 | 0.23 | 5.85 |
| Hg | 82.61 | 42.86 | 674.49 |

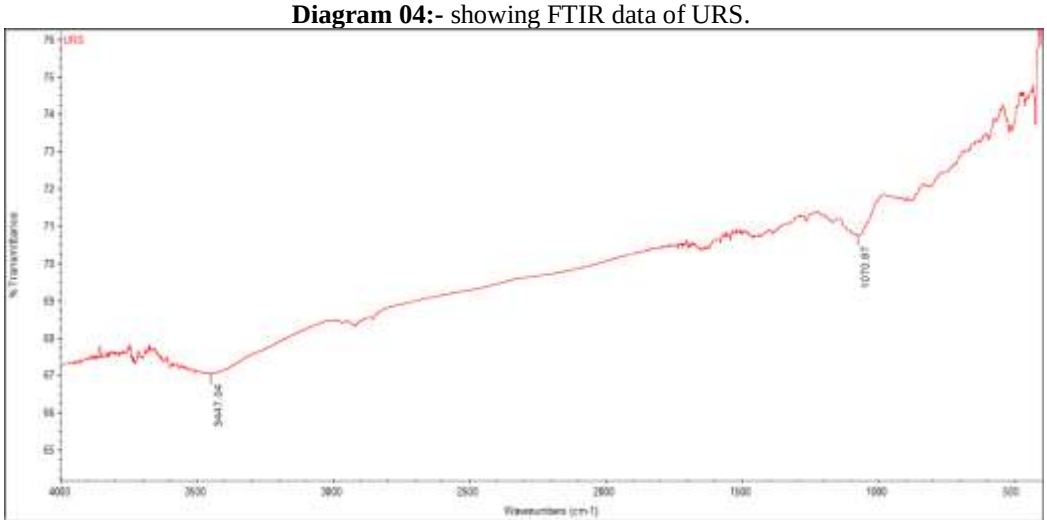
**Table 09-** showing results of EDAX-Area 2

| Element | Weight % | Atomic % | Net Int. |
|---|---|---|---|
| Al | 1.13 | 4.11 | 153.1 |
| Si | 0.92 | 3.2 | 150.8 |
| Mo | 1.96 | 2.01 | 131.12 |
| S | 16.9 | 51.67 | 2293.32 |
| Cl | 0.17 | 0.47 | 13.49 |
| Hg | 78.92 | 38.55 | 674.41 |

**Table 10:-** showing results ofEDAX-Area 3

| Element | Weight % | Atomic % | Net Int. |
|---|---|---|---|
| Al | 1.36 | 4.89 | 178.31 |
| Si | 0.92 | 3.17 | 145.86 |
| Mo | 1.61 | 1.63 | 103.85 |
| S | 16.94 | 51.25 | 2219.37 |
| Cl | 0.34 | 0.94 | 26.47 |
| Hg | 78.82 | 38.11 | 650.22 |

**Diagram 04:-** showing FTIR data of URS.



## Discussion:-

In conceptual study, *Uttamarasasindura* has been described as very effective and as a wide angle remedy.On qualitative test, the *sindura*sample (URS) showed relevant values.Test for acid insoluble ash was carried out to evaluate the percentage of insoluble inorganic content of the sindura in dilute acid. Since a drug must first pass into solution before it can be

absorbed, the acid insoluble ash test is therapeutically very important. It is intended to provide a step towards the evaluation of the physiological availability of the product.The obtained value is 0.48±0.10 which signifies the genuinity of the product. The water soluble ash value was found as 1.20±0.25.

The total ash value has importance in *sindura* preparation because total ash may act as source of trace elements. Physico chemical changes of media during *Shodhana, bhavana* may contribute to this.It is calculated as 10.11±0.08. pH was calculated as 7.3 which is acceptable to body tissues.LOD for the product was carried out.This test is to detect the moisture and volatile content in the sample.Theanalysed value 7.16±0.12 is indicating stability and more shelf life of *Uttamarasasindura*.

SEM analysis of URS shows surface is ultra smooth with auniform morphologyas shown in the result part. The particlesize ranged from 100nm to several µm. The EDAX shows the chemical composition consist of mercury and sulphur. As the particle size was in nanometer, absorption of the drug will be more with quicker action.EDAX refers to energy dispersion X-ray spectroscopy and is employed here as an analysis tool to determine the elemental composition of *Rasoushadhis*.It works by analyzing the spectrum of emitted X-Rays from a sample as a beam of high energy electron is incident upon it. Here EDAX worked in association with SEM.EDAX gives weight % as well as atomic % of the major elements. Selected areas of URS sample were tested.Two elements were observed in major concentration in the tested sample. Observations in Area 1, Area 2 and Area 3 revealed Hg with percentage weight of 82.61%, 78.92% and 78.82% and atomic% of 42.86%, 38.55%, 38.11% respectively. Sulphur (S) with percentage weight of 15.92%, 16.9%, 16.94%and atomic% of 51.68%, 51.67%,51.25% respectively. The atomic% of Hg is less but its mass% is more it is because Hg is heavy metal. Other major elements are Al,Si,Mo and Cl.During*citrakasodhana* the *rakatacitraka* roots were kept immersed in an aluminiumvessel.That may be the reason for presence of aluminium in the sample.Area 2 and area 3 contains Mo.Other mentioned elements are similar in all the three selected areas with slight variations in their values.The element Cl may be a contribution from the *bhavanadravyakumari*.Because the ion content of leaves and sprouts of aloevera contains Cl.Drug*navasadara* also can be a source of Cl.FTIR analysis was carried out for samples of *bhavitakajjali*(BKJ) and the final product *Uttamarasasindura*(URS).The presence of functional group is very much essential in drug absorption as it increase the rate of absorption,on the other hand multiple functional groups may hinder drug absorption by hindering the action of other functional group.In the FTIR spectrum of Sample BKJ-two peaks are obtained at 3148 $Cm^{-1}$ and 1401 $cm^{-1}$.Peak at 3148 $cm^{-1}$can be assigned to N-H bond stretching vibrations.It can be due to presence of ammonium salt or amine formation.Peak at 1401$cm^{-1}$ can be assigned to O-H deformation or C-O stretching.Presenceof peaks together can be considered as an evidence for the formation of ammonium salt of carboxylic acid.In the FTIR spectrum of sample URS, two peaks are obtained at 3447$cm^{-1}$ and 1070 $cm^{-1}$.These peaks can be assigned to O-H stretching and O-H deformation.In the conversion of BKJ to URS, a reduction in number of functional groups has been observed, which inturn reveals the quality of the finally formed *Uttamarasasindura*.The XRD analysis of the samples *kajjali* (KJ), *bhavitakajjali (*BKJ) and *Uttamarasasindura* (URS) showed presence of HgS (cinnabar) in major concentration level.Crystal structure is observed as hexagonal.More than 15 identical peaks of HgS were observed in the final URS sample.KJ and BKJ samples showed peaks of concentration of Chlorine with an orthorhombic crystal structure.About 13 peaks were observed in KJ, BKJ samples. From this results it is clear that the final product formed (URS) is in pure HgSform.Peaks corresponding to (101),(003),(102),(013),(110),(104),(021),(015),(016), (211),(122),(300),(018) planes are obtained for HgS.

## Conclusion:-

*Uttama*rasasindura is a tasteless, odorless, fine and bright brick red coloredsubstance.Chemical characterisation through various techniques shows the final product formed (URS) is in pure HgSform.It contains different fuctional groups .The particle size of the URS ranged from 100nm to several µm.*Uttamarasasindura* has been described as very effective and as a wide angle remedy in the text 'Rasarajacitamani', a book with lot stress on practical applications.

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**Conflicts of interest**
There are no conflicts of interest.

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**Figure 01**-Sublimed final product

**Figure 02**-Powdered sample URS

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