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Research Article

PHYSICO-CHEMICAL CHARACTERIZATION OF SIDDHA SASTRIC FORMULATION *ARUMUGA CHENDHURAM*

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ABSTRACT

Arumuga Chendhuram is a herbo-mineral formulation cited in *Siddha Vaidya Thirattu* prepared through the special oxidation procedure involving purified form of minerals processed under herbal juice. It has been practiced frequently for treating inflammation of joints and anorectal disorders at 65-130 mg with the adjuvant Honey. The present study dealt with analyzing the physico-chemical characterization of *Arumuga Chendhuram* to establish standard quality parameters. Two samples of *Arumuga Chendhuram* (Self prepared and Pharmacy Samples) were subjected to analytical tests and quantitative analyzes using Sophisticated Instruments. Analytical analysis showed low level of moisture content and extractive values in both samples but ash values were high. The functional groups analyzed by FT-Raman Spectroscopic study inferred different numbers of peaks among the samples. The particles of both samples were in 1-10 micron size scanned under Electron microscope. Elements found in both samples were in its oxide form at different concentrations which was analyzed through Wavelength dispersive X-Ray Fluorescence. Both samples contain high concentration of ferric oxide. The concentration of trace and heavy metals were analyzed using Inductively Coupled Plasma - Optical Emission Spectrometer showed the presence of low concentration of Mercury, Arsenic and Iron in self prepared *Arumuga Chendhuram*. From the result of above studies, we inferred that *Arumuga Chendhuram* prepared by us was superior to Pharmacy product in quality and that physicochemical fingerprint shall be used as standard.

KEYWORDS: *Siddha, Chendhuram*, *Arumuga Chendhuram*, Standardization, Mineral.

INTRODUCTION

Metallic preparations play an important role in Siddha treatment modules for treating chronic ailments at minimal dosage itself.[1] Metallic preparations are in various forms such as *Parpam, Chendhuram, Urukku, Kattu, Chunnam* and *Chathu*.[2] These forms are varies in their physicochemical properties, dosage forms and therapeutic values. Among metallic preparations, *Chendhuram* form of medicine has shelf life period of 75 years and has significant process that it can be prepared by processing either single or compound metals into red coloured fine powder by the process of burning, frying or insulating or keeping them in specialized *Pudams* by grinding them with decoctions, *Ceyaneers*, juices etc.[3] There are different types of *Chendhuram* mentioned in the Siddha Materia Medica such as *Chendhuram* prepared by burning e.g. *Arumuga Chendhuram, Poorna Chandrodayam, Chendhuram* prepared by roasting e.g. *Ayaveera Chendhuram, Chendhuram* prepared by grinding e.g. *Chandamarutha Chendhuram, Chendhuram* by exposing to sunlight e.g. *Suyamakkini Chendhuram, Chendhuram* prepared by *Pudam* e.g. *Aya Chendhuram, Kaandha Chendhuram*.[3] For the present study, *Arumuga Chendhuram* (ARC) was chosen to establish standard finger print for quality. ARC, a multi-ingredient formulation comprises of six minerals and has been extensively used in the practice for pain management in arthritis. ARC has wide range of therapeutic usage such as *Arai vatham, Virai vatham, Eruvai mulai noikal, Erikunmam, Varatchi, Anal, Veti soolai, Marpuccali, Karpparogam, Veluppu, Uuthal, Kiranthi, Kutal vatham, Kayam, Araiyappu, Malam, Mantai idi, Tontai vali, Kantamaalai, Culai, Vali, Azhal* and *Perun kalichal*.[4] ARC is prepared in three steps viz., *Sutthi4tthal* (Purification of ingredients), *Villai Seithal* (Grinding with juice and making into dry cakes) and *Eritthal* (Burning).[4] Many *Siddha* Pharmacy industries have been preparing ARC but the quality varies among them since variation found in the raw materials and batches.[5] To ensure the uniform reproducible quality, standard operative procedures (SOP's) should be established.[6] This study was done for characterization analyzes among ARC prepared by us in our laboratory, Department of *Gunapadam*, National Institute of *Siddha*, Chennai, Tamil Nadu, India and ARC procured from GMP certified Pharmacy using classical methods and sophisticated instrumental techniques. The outcome of this study produces the fingerprint of ARC and shall be used as standards for accessing the quality and reproducibility of the product.

MATERIALS AND METHODS

Procurement of *Arumuga Chendhuram*

ARC was purchased from GMP certified SKM Siddha Pharmaceutical industry, Erode, Tamil Nadu, India and coded as sample I.

Preparation of *Arumuga Chendhuram*

ARC was prepared by the method cited in *Siddha Vaidya Thirattu*[4] which is found in the First Schedule of Drugs and Cosmetics Act, 1940.

Raw materials

Lingam (Cinnabar), *Kaantham* (Magnetic oxide of iron), *Gandhagam* (Sulphur), *Venkaaram* (Sodium biborate - Borax), *Indhuppu* (Sodium chloride impure – Rock salt) and *Ayam* (Ferrum - Iron) procured from registered *Siddha* Medical practitioner Dr. Murugesan MD *Siddha*, Orathanadu, Tamilnadu, India. The plant material *Katraazhai* (*Aloe vera*) was collected from herbal garden of National Institute of Siddha, Chennai, India.

Authentication of Raw materials

The raw materials for the above formulation (*Arumuga Chendhuram)* have got authentication from Dr. M. Suresh Gandhi, Assistant professor, Department of Geology, University of Madras, Chennai, Tamil Nadu, India after studying its physicochemical properties. The herbs were authenticated by Dr. D. Aravindhan, Associate professor of Medicinal Botany, National Institute of Siddha, Chennai, Tamil Nadu, India.

Preparation of *Valai Rasam*

Valai Rasam obtained during the process of sublimation of *Lingam* (Cinnabar) has been considered as detoxified form of *Rasam.*[2] A sufficient amount of Cinnabar was grinded with lemon juice for three hours using *Kalvam* (Black stone mortar) and then sublimed in the apparatus called *Urddhapatana yantra*. The mercury was deposited within the upper pot of the apparatus as a blackish powder. This was scraped, rubbed with lemon juice and boiled in water, when it is fit for use.

Purification of *Kaantham*

Kaantham was heated in *Kollan ulai* (Traditional heat blower) until it became red hot. Then it was soaked in *Kollu Kudineer* (Horse gram decoction). This process was done for 21 times.[3]

Purification of *Gandhagam*

Maruthonri ilai karkam (Lawsonia inermis poultice was mixed with Cow's milk curd in an earthen pot and closed with the cotton cloth. Gandhagam was placed over the cloth and closed with another appropriate pot and covered the lid with *Seelai mann* (mud pasted cotton cloth) and dried. This set up was kept in the dug ground and subjected to *Pudam* (Traditional method of heating using Cow dung cakes). The *Gandhagam* was melted and settled down in the pot. It was collected and washed with water. This process was done for 7 times.[3]

Purification of *Venkaaram*

Venkaaram was fried in a mud plate until the water contents evaporated.[3]

Purification of *Indhuppu*

Indhuppu was soaked in *Kaadi neer* (Rice Vinegar) for three day and dried well.[3]

Purification of *Ayam*

Ayam was heated in *Kollan ulai* (Traditional heat blower) until it became red hot. Then it was soaked in *Kaadi* (Rice Vinegar), *Ennai* (Gingelly oil), Cow's Urine and*Kollu Kudineer* (Horse gram decoction) respectively. This process was done for 3 times.[3]

Processing of *Arumuga Chendhuram*

5 parts of *Valai Rasam was ground with* 9 parts of purified *Gandhagam* using *Kalvam* and the black powder formed was kept separately. 7 parts of purified *Kaantham,* 8 parts of purified *Venkaaram*, 4 parts of purified *Indhuppu*, 12 parts of purified *Ayam* were powdered separately using *Kalvam* and mixed thoroughly. This mixture was added to the black powder and ground to fine powdered form. To this, *Katralai* juice was added constantly and triturated for 5 days and made into cakes and dried well. The dried cakes were placed in an earthen pot and covered with seven *Seelai mann* (Mud pasted Cotton roll) and dried well. Then, the pot was heated under moderate flame for 24 h and cooled and opened to get finished test drug. This test drug was stored in an air tight sterile glass container and kept in a dark condition.

Qualitative analysis of *Arumuga Chendhuram*

***Siddha* classical method**

The quality of *Arumuga Chendhuram* was accessed by the parameters cited in Siddha Formulary of India. The parameters are follows.

- Red in colour without any shiny appearance
- Tasteless and odourless
- Did not regain luster on heating again at same temperature
- Sample floats on water. Did not immediately immersed in water
- Not translucent
- Impinged in the papillary ridges when the sample rubbed in between Index finger and Thumb

Physico chemical analysis

Physical properties such as colour, taste, odour, Reaction to HCl, Magnetic and fluorescent properties were analyzed. The physico-chemical test was performed at Regional Research Institute of Unani Medicine, Chennai, India. The procedures recommended in protocol for testing (Pharmacopoeial Laboratory for Indian Medicine) were followed to determine loss on drying at 105°C, total Ash, total acid-insoluble ash and solubility in alcohol and water.[7]

Functional group analysis

The study was carried out using BRUKER RFS27 Stand-alone FT-Raman Spectrometer having scan range from 50 to 4000 cm^{-1} done at Sophisticated Analytical Instrumentation Facilities, Indian Institute of Technology - Madras, Chennai, Tamil Nadu, India. The functional groups present among Samples were analyzed by correlating with the standard Raman Spectroscopy data.[8]

Microscopic analysis

Scanning Electron Microscopic study (SEM) on LC was carried out by using Carl Zeiss MA15/EVO 18 High Resolution Instrument done at SAIF, IIT Madras, Chennai-36. The nature of particle distribution and morphology in LC was analyzed.

Quantitative analysis of *Arumuga Chendhuram*

Estimation of oxide forms of elements

The study was performed using Bruker S8 Tiger Wavelength dispersive X-ray fluorescence spectrometer (WDXRF) under vacuum mode.

Estimation of heavy metals and trace minerals

The study was performed using Perkin Elmer Optima 5300 DV Inductively Coupled Plasma – Optical Emission Spectrometer (ICP-OES). For digestion of 100 to 200 mg of LC, mixture of Nitric acid and Perchloric acid (2:1) was used. After the completion of digestion, the content was cooled and heated for the removal of acid. Then the solution was made into 50 mL using deionized water.

RESULTS AND DISCUSSION

Qualitative analysis

The table 1 shows both samples satisfies all the quality parameters mentioned in the literature and proves that both samples were well finished and fit for therapeutic usage.

Table 1: Quality observations of *Arumuga Chendhuram*

S.No	*Siddha* classical method	Sample I	Sample II
1	Red in colour without any shiny appearance	Positive	Positive
2	Tasteless	Positive	Positive
3	Odourless	Positive	Positive
4	Did not regain luster on heating again at same temperature	Positive	Positive
5	Sample floats on water. Did not immediately immersed in water	Positive	Positive
6	Not translucent	Positive	Positive
7	Impinged in the papillary ridges when the sample rubbed in between Index finger and Thumb	Positive	Positive

There was no difference observed among the samples in their physical characters (Table 2) except in their colour. The literature cited that a well finished *Arumuga chendhuram* should possess bright colour resemble as the colour of *Punica granatum* flower. Sample II had bright colour appearance which satisfies the better quality and also its fineness was more. Both samples were free from radioactive contents and metals possessed magnetic property. Both samples were highly stable due to no reaction with HCl.

Table 2: Physical characters of *Arumuga Chendhuram*

Parameter	Sample I	Sample II
Colour	Dark red	Bright red
Taste	Nil	Nil
Odour	Nil	Nil
Touch	Soft	Soft and fine
Magnetism	Nil	Nil
Reaction to HCl	No effervescence	No effervescence
Luminescence	Non fluorescent	Non fluorescent

The water soluble extractive value was low in sample II on compared with sample I inferred that sample II contains more stable components. Both samples had low moisture content implies having better stability. Both samples had high ash value and inferred that more inorganic components were seen. High insoluble ash value was observed in Sample II and high water soluble extract was observed in Sample I (Table 3).

Table 3: Analytical Parameters of *Arumuga Chendhuram*

Parameter	Sample I	Sample II
Loss on drying at 105°C	2.19±0.11	2.63±0.06
Total Ash Value	92.44±0.22	98.26±0.11
Acid Insoluble ash	62.47±0.12	94.48±0.16
Alcohol soluble extract	0.23±0.07	0.29±0.09
Water soluble extract	22.23±0.13	1.79±0.14

Values are expressed as Mean±S.D.

The results of table 4 shows the functional groups present in the samples. By Raman Spectroscopy of Sample I, Raman shifts have been observed at 12 peaks (Fig. 1). Among 12 peaks, prominent band at 292.71 cm^{-1} was observed representing strong υ(Xmetal-O) group and has other bands representing the presence of lattice vibrations in crystals indicating storage of heat in the form of oscillatory energy, strong υ(S-S), strong υ(C-S) aliphatic and medium υ(C-O-C). Eighteen peaks were observed in Sample II (Fig. 2) having prominent band at 299.40 cm^{-1} representing strong υ(Xmetal-O) group. The other bands in Sample E indicate the presence of lattice vibrations in crystals, strong υ(S-S), strong υ(C-Br) and strong υ(C-S) aliphatic groups, medium υ(C-O-C), strong υ(CC alicyclic group with aliphatic chain vibration. strong υ(C=S) ,

strong υ(C-(NO2)) and strong υ(O-H). Sample I shows low content of functional groups having more inorganic materials rather than organic. Sample I shows high number of peaks at the functional group showing metals in oxide form.

Table 4: Analyzes of Functional groups present in *Arumuga Chendhuram*

Functional Group/Vibration	Region	Sample I* (Band cm^{-1})	Sample II* (Band cm^{-1})
Lattice vibrations in crystals	10-200 cm^{-1}	71.64, 100.36, 116.90, 135.74	75.74, 191.52
υ(S-S)	430 -550 cm^{-1}	-	455.54
υ(Xmetal-O)	150-450 cm^{-1}	163.40, 225.68, 260.10, 292.71, 420.33	299.40, 352.85, 405.57
υ(C-Br)	500 - 700 cm^{-1}	-	578.78, 664.69
υ(C-S) aliphatic	630 - 790 cm^{-1}	691.36	728.16
υ(C-O-C)	800 -970 cm^{-1}	811.53, 839.39	835.03
υ(CC) alicyclic, aliphatic chain vibrations	600 - 1300 cm^{-1}	-	796.13
υ(C=S)	1000 - 1250 cm^{-1}	-	1059.78, 1093.09, 1131.63, 1196.98
υ(C-(NO2))	1340 - 1380 cm^{-1}	-	1364.54
υ(O-H)	3100 - 3650 cm^{-1}	-	3142.84, 3233.77

SEM images of both samples showed difference in size from 1 – 10 µm and agglomeration of the particles (Fig. 3-4). Repeated grinding for 5 days and heating for 24 h during the preparation process of ARC made agglomeration of the particles. Micron level particles size had increased dissolution rate which enhance better absorption of ARC in the alimentary tract on suspended with adjuvant.[9]

Quantitative analysis

The concentration of elements in oxide form was observed by Wavelength dispersive X-ray fluorescence Spectroscopic study was shown in the table 4. Ferric oxide found major content in both samples. The concentration of trace elements including heavy metals were observed by Inductively Coupled Plasma Optical Emission Spectroscopic study was shown in the table 5. Sample II has low concentration of Mercury, Arsenic and Iron on compared with Sample I.

Table 4: Elements in oxide form of *Arumuga Chendhuram*

S. No	Elements in oxide form	Sample I Concentration (%)	Sample II Concentration (%)
1	Ferric oxide	44.33	41.95
2	Sulphur trioxide	18.54	16.36
3	Sodium dioxide	16.95	10.35
4	Silicon dioxide	13.43	11.44
5	Chlorine	3.83	0.39
6	Aluminium trioxide	0.83	2.00
7	Calcium oxide	0.67	2.63
8	Potassium oxide	0.47	11.22
9	Magnesium oxide	0.44	2.87
10	Manganese monoxide	0.16	-
11	Mercury	0.13	0.04
12	Phosphorus pentoxide	0.08	0.56
13	Lead monoxide	0.05	0.15
14	Cupric oxide	0.05	0.02
15	Chromium (III) Oxide	0.04	0.02

Table 5: Elemental Concentration in *Arumuga Chendhuram*

S.No	Element	Wave length (nm)	Sample I		Sample II	
			(mg/L)	(ppm)	(mg/L)	(ppm)
1	Arsenic(As)	188.979	3.124	3.127568555	0.124	0.124141646
2	Cadmium(Cd)	228.802	BDL	BDL	BDL	BDL
3	Iron(Fe)	238.204	82.530	82.624274267	42.530	42.578582147
4	Mercury(Hg)	253.652	04.105	4.109689154	03.125	3.128569697
5	Sodium(Na)	589.592	134.510	134.663651177	24.310	24.337769386
6	Lead(Pb)	220.353	BDL	BDL	BDL	BDL
7	Sulphur(S)	180.731	87.224	87.323636237	51.524	51.58285602
8	Vanadium(V)	313.07	BDL	BDL	BDL	BDL

CONCLUSION

Arumuga Chendhuram (ARC) is a herbo-metallic formulation has been prepared by many Siddha Pharma industries. For the study, we optimized the best variety of ARC among the self-prepared ARC and ARC procured from GMP certified Pharmacy. ARC procured from the industry was coded as Sample I and ARC prepared as per the

method cited in the 1940 drug and cosmetic act authenticated literature *"Siddha Vaithiya Thirattu"* was coded as Sample II. The both samples were analyzed for qualitative and quantitative estimation. Preliminary physical parameters such as total ash, moisture content and extractive values were analyzed. The functional groups were analyzed by FT-Raman Spectroscopic study. The content of lead and cadmium were analyzed using Atomic Absorption Spectroscopic study. The concentration of elements in oxide form was analyzed through Wavelength dispersive X-Ray Fluorescence. The concentration of trace and heavy metals were analyzed using Inductively Coupled Plasma - Optical Emission Spectrometer. On analyzing the results of above studies, it can be concluded that Sample II (Prepared at Gunapadam Lab, National Institute of Siddha) is better in quality than Sample I (Procured from SKM Pharmacy).

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