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PHYSICOCHEMICAL CHARACTERISATION OF *SAMAGUNA BALIJAARITA KAJJALI* AT DIFFERENT DURATIONS OF GRINDING

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ABSTRACT

Kajjali is a *khalweeya rasayana* that can be used as a medicine and also is a base for many mercurial preparations. It is an example of *sagandha niragni murchana* of *parada. Shodhita parada* and any other *dhatus* like *gandhaka* are taken in the prescribed ratio and ground together till they attain specific characteristics like fineness and jet-black colour like collyrium. In this paper, the work done on pharmaceutical and analytical aspects of *samaguna balijaarita kajjali* at different grinding durations is detailed. The pharmaceutical study included *shodhana* of *parada* and *gandhaka* and grinding them together for 100 hours. *Vareetaratvam* was attained by 20 hours, *rekhapoornata* by 40 hours, and *nishchandratvam* by 70 hours. 5 samples were collected 20 hourly and were subjected to X-ray diffraction analysis (XRD), Particle size analysis (PSA), and Thermogravimetric analysis (TGA). All 5 samples were identified as a mixture of sulphur and metacinnabar in XRD. PSA showed decreased particle size with an increase in the duration of grinding and reached 480nm after 100 hours of grinding. TGA of *Kajjali* implied that thermal stability improves with the duration of grinding. From the study, it can be concluded that as the duration of grinding increases, the particle size of *Kajjali* decreases, and thermal stability improves. These properties contribute to a better therapeutic action of the *Kajjali* as medicine itself or as an intermediate product of any mercurial formulation.

Keywords- *samaguna balijaarita kajjali*, metacinnabar, *parada*, *gandhaka*, XRD, PSA, TGA

INTRODUCTION

Ayurveda is a traditional system of medicine with historical roots in the Indian subcontinent. On studying the history of Ayurveda, we can see that the post-samhita period witnessed the evolution of *Rasa sastra*. *Rasa sastra* means 'science of mercury'. *Rasoushadhis* constitute mineral and herbo- mineral drugs but *rasa* is given prime importance as the name of the science indicates. *Rasoushadhis* are considered superior to all other medicines as they have instant effectiveness in very small doses for various diseases from acute to chronic. Also, they can cure *asadhya rogas* while herbal drugs are effective only for *sadhya rogas*[1]. The ultimate aim of *Rasa Sastra* is to attain *jeevan mukti* i.e., to attain salvation before death. For this, *sthira deha* with high longevity is a necessity. It can only be attained by using something indestructible and *sthira* itself. And *rasa* is the only such *dravya* [2].

Rasa being a highly toxic and unstable substance, should be converted to any of the 4 *rasa kalpas* viz *khalveeya rasayana, parpati, pottali*, *or kupipakwa rasayanas* to make it suitable for human consumption. *Kajjali* is a *khalweeya rasayana* that can be used as a medicine and also is used as a base for the majority of herbo-mineral formulations mentioned in classics. Avoiding the use of elemental mercury by combining it with sulphur is the uniqueness of *Rasasastra* and indicates the deep knowledge in toxicity and drug designing, centuries ago. *Shodhita parada* and any other *shodhita dhatus* like *gandhaka* are taken in the prescribed ratio and ground together till they attain specific characteristics like fineness and jet-black colour like collyrium [3]. Even though many ratios of *kajjali* are seen in *yogas*, *samaguna balijaarita kajjali* is the most prepared and used one. *Kajjali* is also one of the prime ingredients in about 80 formulations mentioned in Ayurvedic Formulary of India which have a wide range of therapeutic utility [4]. In the current scenario, where the safety of *rasa* drugs is a global concern, integration of classical and contemporary studies is really important to deeply understand the different pharmaceutical preparations explained centuries ago. The present study concentrates on preparing *samaguna balijaarita kajjali* prepared using equal quantites of *parada* and *gandhaka*, and studying its physico-chemical characteristics at different grinding durations through Ayurvedic parameters like

nishchandratvam, slakshantvam, kajjalabha etc along with contemporary analytical tools like XRD, PSA and TGA.

MATERIALS AND METHODS

Pharmaceutical study

1. Collection of raw materials
2. *Shodhana* of *parada*
3. *Shodhana* of *gandhaka*
4. Preparation of *Kajjali*

Collection of raw materials

99.9% pure distilled mercury and sulphur powder were purchased from Nice Chemicals, Ernakulam.

Shodhana of *parada*

Parada shodhana method mentioned in Rasendra Sara Sangraha was followed [5].

Mardana of 450g of *parada* was done in *rasona swarasa*, *nagavalli swarasa,* and *triphala kwatha* for 12 hours each. After each *mardana*, it was washed with warm *kanjika* to regain mercury.

Table no. 1: Observations during *parada shodhana*

Initial weight of *parada*	Mardana medium and amount	Duration of mardana	Weight after mardana
450g	*Lasuna swarasa* - 100 ml	12 hours	448.7g
448.7g	*Nagavalli swarasa* - 100ml	12 hours	438.18g
438.18g	*Triphala kwatha*- 100ml	12 hours	422.088g

Total loss- 27.92g

Precautions-

Mask and gloves should be worn throughout the process.

Care should be taken to avoid spillage of mercury while grinding and washing.

Figure no. 1: Shodhana of parada in different media

Lasuna swarasa Nagavalli swarasa Triphala kwatha

Shodhana* of *gandhaka

Gandhaka shodhana method mentioned in *Rasa Tarangini* was followed [6].

900g of Sulphur was heated in a vessel with *goghrita*. Once melted completely, it was poured into a vessel containing *godugdha* through a cloth. The process is repeated 3 times.

Table no. 2: Observations during *Gandhaka shodhana*

Initial weight of *gandhaka*	Weight of *ghrita*	Amount of milk	Weight after *dalana*
900g	400g	1500ml	880g
880g	350g	1500ml	850g
850g	300g	1500ml	808g

Total loss- 92g

Precautions

Mask and gloves should be worn throughout the procedure.

Low flame should be maintained throughout the heating process

Complete drying should be ensured before storing

Figure No.2: Shodhana of Gandhaka

Preparation of *Kajjali*

Samaguna balijarita kajjali was prepared according to the reference in Rasa Tarangini[7]. A *khalwa yantra* made of tamda stone of 25inches length was used.

Table no.3: Ingredients of *Samaguna balijaarita kajjali*

Ingredients	Quantity
Shudha *parada*	350g
Shudha *gandhaka*	350g

Grinding was done for 100 hours manually. Every 20 hours, 5g samples were collected for analysis.

Table no. 4: Observations during the preparation of *Kajjali*

Duration of grinding	Observations
10 min	Large mercury globules disappeared
45min	Grey coloured mixture
1 hour	Blackish grey coloured mixture
2 hours	Blackish grey coloured mixture
4 hours	Mixture turned more blackish and finer, spilling of *Kajjali* noted
20 hours	*Sookshmatvam* and *vareetaratvam* attained.
40 hours	*Rekhapoornata* attained
60 hours	Shining considerably reduced, jet black colour
70 hours	*Nishchandratvam* attained (when rubbed with a drop of water, no shining observed)
80 hours	*Kajjali* was very soft and fine.
100 hours	*Kajjali* attained all *Samyak lakshanas*

Precautions

Gloves and masks should be worn throughout the grinding

Care should be taken to avoid spillage of *Kajjali*

Uniform pressure should be maintained while grinding

Figure No.3: Preparation of *Kajjali*

ANALYTICAL STUDY

Ayurvedic parameters of kajjali samples collected every 20 hours of grinding were tested. *Vareetaratvam* was attained by 20 hours, *rekhapoornata* by 40 hours, and *nishchandratvam* by 70 hours. None of the samples showed whitish discolouration when rubbed over a copper foil.

Figure no 4: Ayurvedic parameters of *kajjali* collected every 20 hourly

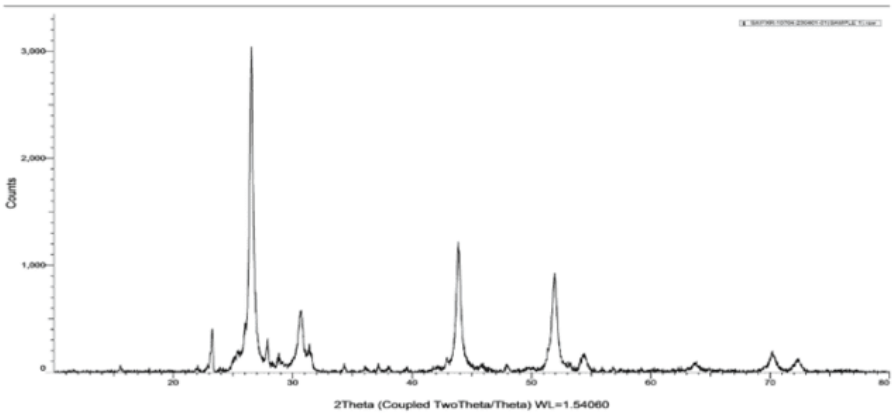
Duration	Rekhapoornata	Vareetaratvam	Nishchandratvam	Copper test
20 hours				

40 hours				
60 hours				
80 hours				
100 hours				

XRD

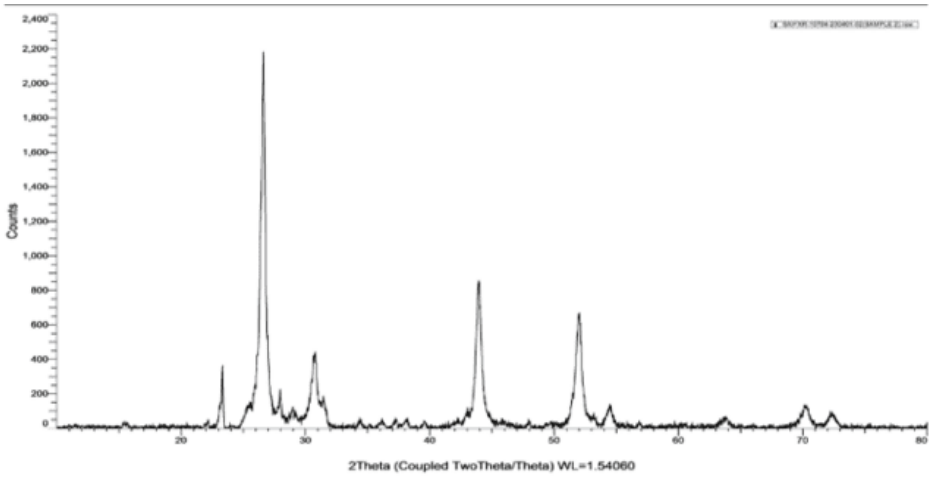
Powder X-ray diffraction analysis (qualitative)was conducted at STIC, CUSAT to determine the crystallographic structure.

Graph no.1: XRD of *Kajjali* sample after 20 hours of grinding



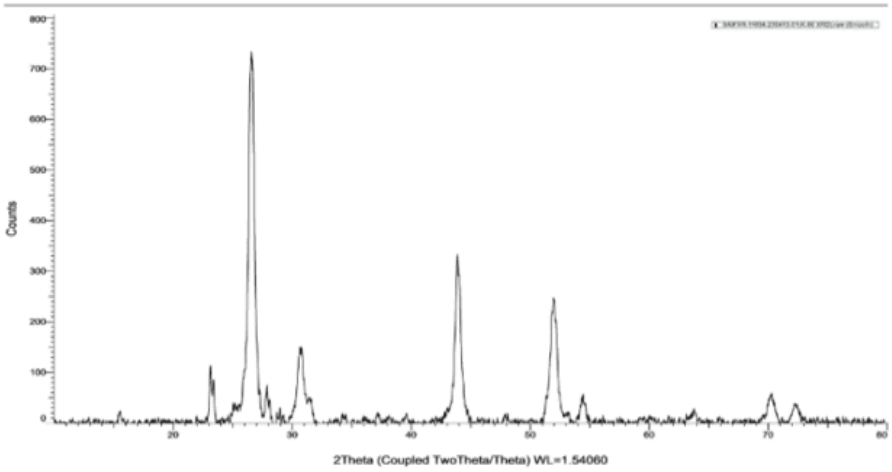
The XRD pattern obtained matches that of metacinnabar (100.0%) with a cubic lattice system. Peaks were obtained at 2θ values of 23.218º, 26.574º, 27.844º, 28.861º, 30.691º, 31.403º, 43.909º, 51.891º, 54.433º, 70.193º, 72.326º. Crystallite sizes range from 195.2 angstroms to 574.5 angstroms.

Graph no.2: XRD of *Kajjali* sample after 20 hours of grinding



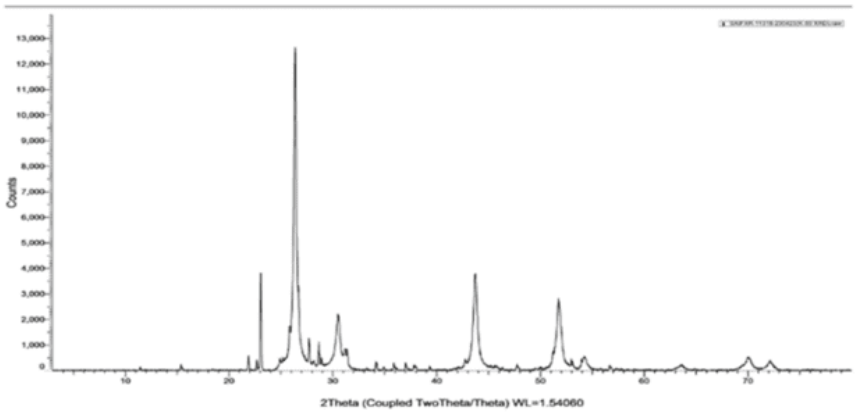
The XRD pattern obtained matches that of metacinnabar (100%) with a cubic lattice system. Peaks observed at 2θ values of 23.324º, 26.618º, 27.957º, 28.992º, 30.750º, 31.332º, 34.358º,43.941º, 51.980º, 54.462º, 56.851º, 63.849º, 70.219º, 72.315º. The crystallite sizes range between 188.1 angstroms and 632.6 angstroms.

Graph no.3: XRD of *Kajjali* sample after 60 hours of grinding



The XRD pattern obtained matches with metacinnabar (85.9%) and sulphur 14.1%). Metacinnabar had a cubic lattice system while sulphur had an orthorhombic lattice system. Major peaks were obtained at 2Θ values of 23.133°, 26.560°, 30.706°, 43.882°, 51.967°. The crystallite sizes range between 158.9 angstroms and 194.8 angstroms.

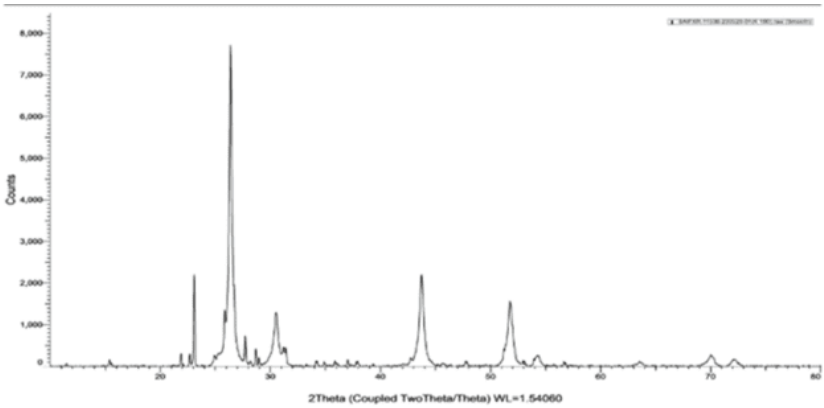
Graph no. 4: XRD of *Kajjali* sample after 80 hours of grinding



The XRD pattern obtained matches with meta cinnabar – 20.6% and sulphur 79.4%. Meta cinnabar had a cubic lattice system while sulphur had an orthorhombic lattice system.

Maximum peaks were obtained at 2Θ values of 23.058º, 26.353º,43.712ºand 51.768º. The crystallite size ranges from 208.7 angstrom to 823.8 angstrom.

Graph no.5: XRD of *Kajjali* sample after 100 hours of grinding

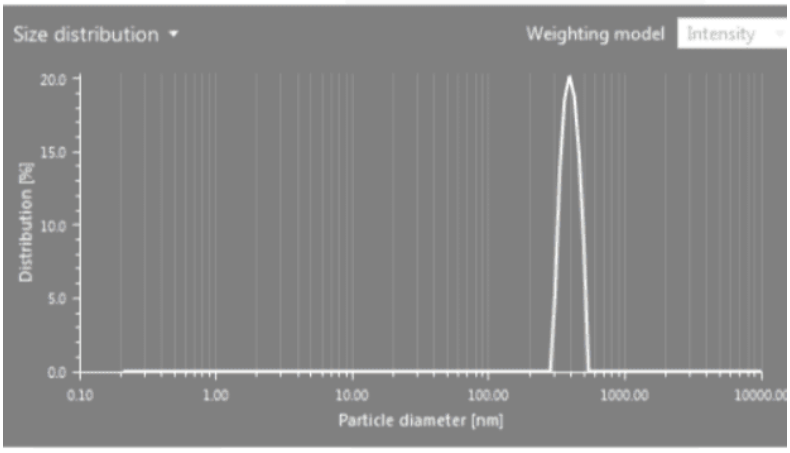


The XRD pattern obtained matches with metacinnabar (83.6%) with a cubic lattice system and sulphur (16.4%) with an orthorhombic lattice system. Maximum peaks were obtained at 2Θ values 23.069º, 26.371º,30.521º, 31.247º,43.735ºand 51.768º. The crystallite sizes of the particles are between 196.9 angstrom to 846.5 angstrom.

Particle size analysis

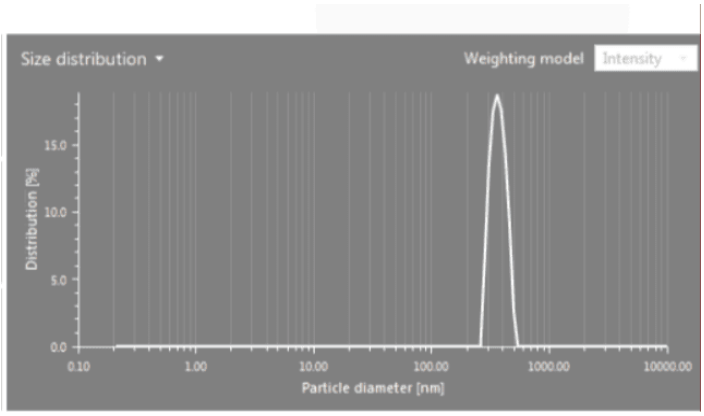
Particle size analysis of *Kajjali* samples was conducted at NIIST-National Institute for Interdisciplinary Science and Technology (CSIR), Thiruvananthapuram.

Graph no. 6: PSA data of *Kajjali* after 20 hours of grinding



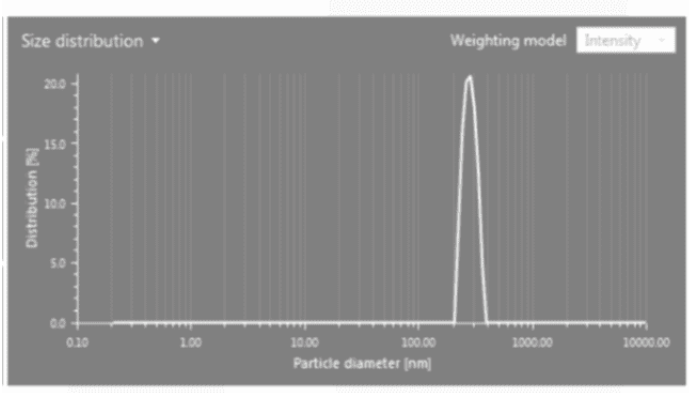
The hydrodynamic diameter of the sample was found to be 635.6467338 nm.

Graph no. 7: PSA data of *Kajjali* after 40 hours of grinding



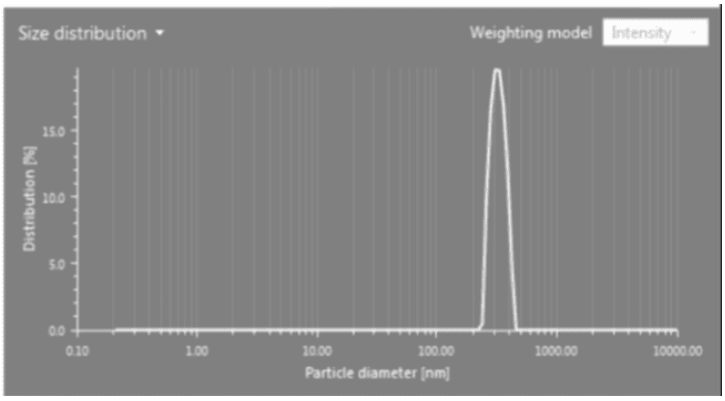
The hydrodynamic diameter of the sample was found to be 553.9672137 nm.

Graph no.8: PSA data of *Kajjali* after 60 hours of grinding



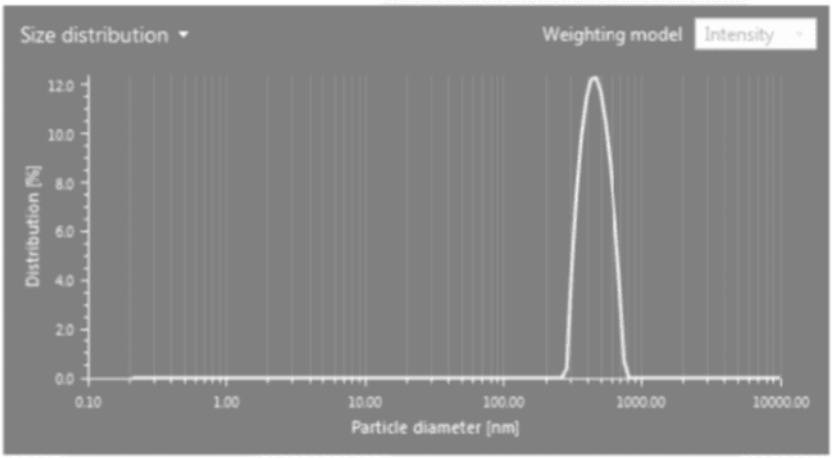
The hydrodynamic diameter of the sample was found to be 518.4536046 nm.

Graph no.9: PSA data of *Kajjali* after 80 hours of grinding



The hydrodynamic diameter of the sample was found to be 497.4138609 nm

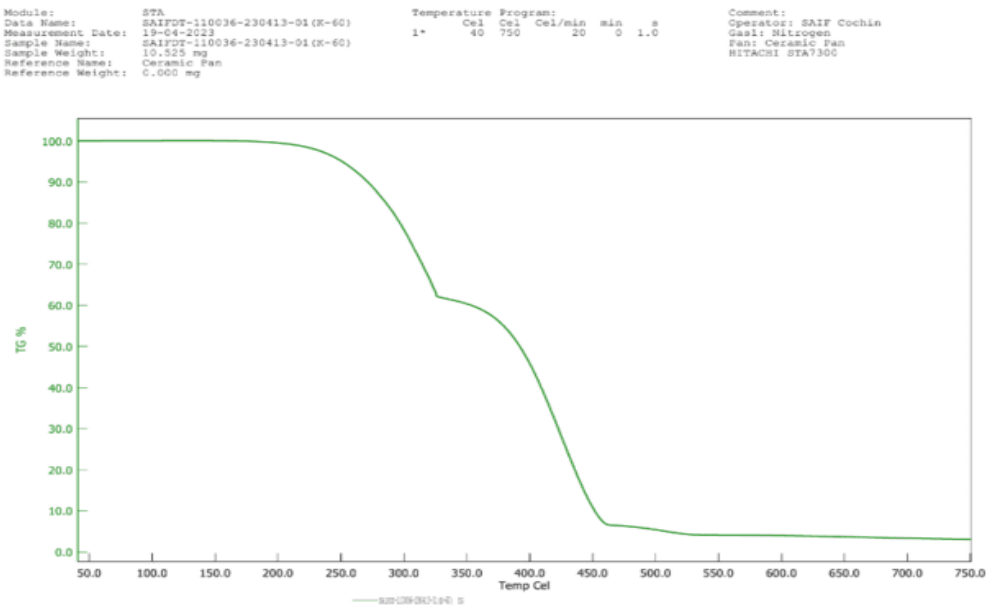
Graph no. 10: PSA data of *Kajjali* after 100 hours of grinding



The hydrodynamic diameter of the sample was found to be 480.453046nm.

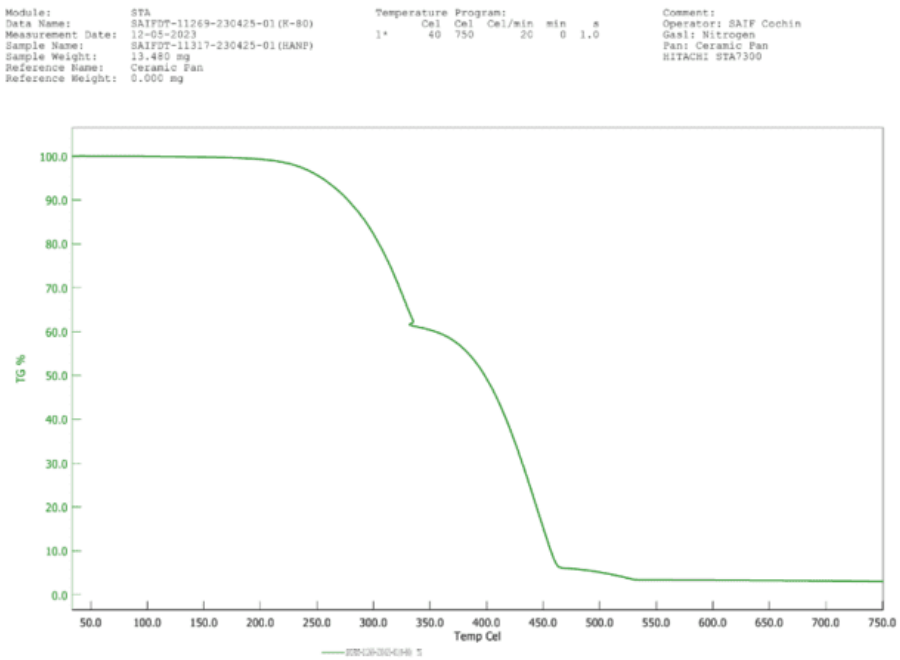
Thermogravimetric analysis

Graph no.11: TGA data of *Kajjali* after 60 hours of grinding



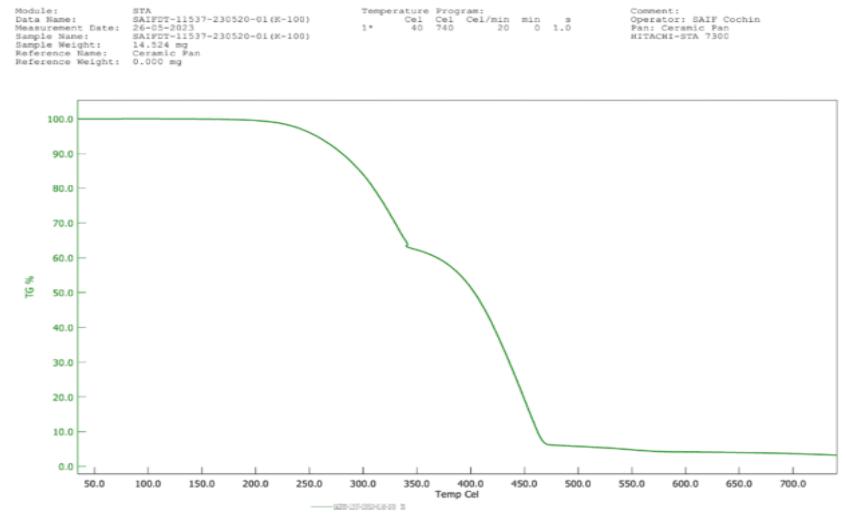
Upto 165.950149°C, the sample was thermally stable and was showing no weight loss. Afterward, there was a gradual weight loss of up to 38% of weight when the temperature reaches 325.213287°C. Then up to 390.58422°C the sample gradually loses weight and reaches 50% of mass. From 391 degrees onwards rapid weight loss was observed. When the temperature reaches 534°C weight of the sample reaches 4.19938 % and was observed to be stable afterwards.

Graph no.12: TGA data of *Kajjali* after 80 hours of grinding



Up to 168.4593°C, the sample was thermally stable and showed no weight loss. Afterward, there was a gradual weight loss and reached 62.7395 % of weight when the temperature reaches 334.752594°C. Then up to 394.123352°C the sample gradually loses weight and reaches 51% of mass. Then onwards rapid weight loss was observed. When the temperature reaches 536 °C weight of the sample reaches 4.19938 % and was observed to be stable afterwards.

Graph no 13: TGA data of *Kajjali* after 100 hours of grinding



The sample didn't show any weight loss up to 200.418°C and was thermally stable. Then there was a gradual weight loss up to 291.5361°C and weight becomes 86.7862% of initial weight. Then a sudden weight loss can be seen till 342.00668°C and weight reaches 62.96255% of the initial weight of the sample. Afterward, a very gradual weight loss is observed as the weight percentage reaches 50.914% at 401.3648°C. A sudden weight loss is observed afterward and the sample became thermally stable at 542.0923°C and at 4% of the weight.

DISCUSSION

Kajjali preparation is constituted of various steps. The first step was *parada shodhana* which was done according to a method from *Rasendra Sara Sangraha*, where *parada* was given *mardana* in *lasuna swarasa*, *nagavalli swarasa*, and *triphala kwatha* respectively for 12 hours each, and washed with warm *kanchika*. *Shodhana* with this method removes all dosas of *parada* and makes it suitable for *sarva karmas*. 450g of *parada* was taken for *shodhana*. On *mardana* with *lasuna swarasa*, the mixture turned blackish with jelly-like consistency on standing. As *lasuna* contains allicin, alliin which are sulphur containing compounds, it binds well with mercury. On using warm *kanchika* for washing, regaining mercury was possible. *Mardana* with *nagavalli swarasa* was easy as the mixture was watery, but washing was tedious as mercury settled in the base of mortar in a paste-like consistency. Mardana with *triphala kashaya* led to the highest loss as the binding was strong and regaining mercury was very time-consuming and tricky process. Total 27.92g of loss was there after *shodhana*.

Dalana method mentioned in *Rasa Tarangini* was taken for *shodhana* of *gandhaka*. It involves melting *gandhaka* with ghrita and pouring it into milk through a cloth 3 times. Even though chemically pure sulphur was purchased, *shodhana* is necessary as it imparts specific desirable properties to the drug rather than the mere removal of impurities. While reviewing different methods for *gandhaka shodhana*, the most repeated media used are *goghrita* and *goksheera*. Also, it is the antidote for *gandhaka* which makes it the perfect medium for *shodhana*. 900g sulphur was taken for *shodhana*. Even though the reference mentions taking an equal quantity of *ghrita*, it was observed that it is not necessary and ghee tends to trap inside the solidified *gandhaka* in small pockets which makes it very difficult to wash and may cause burns. Hence only a sufficient amount of *ghrita* was taken for melting. While melting,

a low flame was always maintained as on higher temperatures *gandhaka* may burn and become reddish in colour. After three *dalanas*, *gandhaka* appeared whiter and the *teekshna gandha* of *gandhaka* was also reduced. Proper drying was ensured before storing to prevent any fungal growth. 92g sulphur was lost during the *shodhana* of *gandhaka*.

Parada naturally has qualities like *chanchalya* and *dravatva* which persists after *shodhana*. To solve these problems of mercury, Acharyas have mentioned *Parada bandhas*. One among them is the *Kajjali bandha*. For the present study, 350g each of *shudha parada* and *shudha gandhaka* were triturated together for 100 hours. *Krishnatvam* was attained by 4 hours, *vareetaratvam* by 20 hours, *rekhapoornata* by 40 hours, and *nishchandratvam* was attained by 70 hours. *Nischandratwa* is found to be most valuable to know the binding of particles in *Kajjali*. The procedure of grinding has to be continued until the mixture of *parada* and *gandhaka* becomes lustreless which indicates the absence of free particles of mercury.

Analysis of *Kajjali* samples was done at an interval of 20 hours.

On XRD analysis, 20 hour and 40-hour *Kajjali* samples were identified as 100% metacinnabar with a cubic lattice system. Samples at 60, 80 and 100 hours were identified as a mixture of metacinnabar with a cubic lattice system and sulphur with an orthorhombic lattice system. 350g each of sulphur and mercury was used to prepare *Kajjali*. To form a molecule of metacinnabar, 1 atom of mercury combines with 1 atom of sulphur. So, in equal weights of mercury and sulphur, the number of atoms of sulphur is 6.2 times that of mercury atoms. That means almost 84%of sulphur will remain as free sulphur itself. *Kajjali* samples at 20 and 40 hours did not detect any sulphur, maybe because XRD analysis only takes less than 10mg of sample for analysis and it may have missed any free sulphur that was present. The crystallite sizes came in the range of 15nm to 84 nm.

While doing particle size analysis, a good dispersion was not obtained in water. Hence other dispersion media were tried and the analysis was done in ethanol. Particle size reduced as the duration of grinding increased from 635.64nm to 480.453 nm.

The thermal stability of the *Kajjali* sample at 60 hours was analysed and it was observed that there were 2 major phase transitions. One around 165°C and 2nd one is after 325°C. The same pattern can be observed in the *Kajjali* sample at 80 hours. 2 phase transitions are seen, 1st at 168°C and 2ndat 334°C. *Kajjali* at 100 hours showed phase transitions at 200°C and 342°C. In all 3 samples, the first weight loss may indicate the conversion of free sulphur present in *Kajjali* to sulphur oxides. The 2ndweight- loss may indicate the sublimation of *Kajjali* and its conversion to another form. These phase transitions are analogous to the conversion of *Kajjali* in *kupipakwa rasayanas* like *Rasa sindura*, where it sublimes, rises, and deposits in the neck of a corked *kupi*. Finally, the samples become thermally stable after 534°C, 536°C, and 542°C respectively when 5-4% of weight remains. By the time this temperature is attained, only 5-4% of mass remains in the same state and the rest changes its physical state. While analysing the data it can be seen that with increasing the duration of grinding there is an increase in thermal stability. These results show that the longer the *Kajjali* is ground, stronger the bonds between mercury and sulphur resulting in higher thermal stability.

CONCLUSION

Samaguna balijaarita Kajjali is a base for many mercurial formulations. Also, it has a wide range of therapeutic applications when used along with appropriate *anupanas*. While preparing *Kajjali, samyak lakshanas* mentioned in classics like *nishchandratvam, slakshnatvam, krishnatvam, kajjalabha* etc were observed every 20 hourly along with modern analytical methods like XRD, PSA and TGA. All the samples from 20 hours itself were identified as either metacinnabar or a mixture of meta cinnabar and extra sulphur in XRD and no further change in chemical composition is noted. Particle sizes decreased with increases in the duration of grinding and reached 480 nm after 100 hours of grinding. Thermal stability of the *Kajjali* sample improved with increased duration of grinding which indicates that the mercury-sulphur bonds in *Kajjali* become stronger with grinding. using a *Kajjali* with better thermal stability will yield a product that is more stable and safer to consume.

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