

## PREPARATION AND CHARACTERIZATION OF *KOWSIGAR KUZHAMBHU*, A *SIDDHA* FORMULATION

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### ABSTRACT

*Kowsigar kuzhambhu* is a *Siddha* formulation comprising *Piper longum*, *Terminilia chebula*, *Brassica Nigra*, *Ferula Asafoetida*, *Cuminum Cyminum* Linn., *Aconitum Lycotonum*, *Croton tiglium*, *Carum carvi*, potassium chloride, sodium borate, mercury, arsenic sesquisulphide and arsenic trisulphate. The major steps involved in the preparation of *Kowsigar kuzhambhu* are (i) purification of raw materials and (ii) formulation of dosage form. The purification step involves treatment of raw materials with various purification agents. The average particle size of the formulation was found to be 112  $\mu$ m. The importance of purification steps has been ascertained from the elemental analysis of raw materials and intermediates obtained from purification steps.

**Keywords:** *Kowsigar kuzhambhu*, Elemental analysis, Particle size, Scanning electron micrograph

### INTRODUCTION

*Siddha* is one of the traditional systems of medicine practiced in Southern parts of India. This system of medicine that originated in the Dravidian regions has its documentation in Tamil, one of the oldest languages of human civilization. *Siddha* system, apart from suggesting medicines for treatment of diseases, deals with balanced way of living that includes exercises like *yoga*, *asana* etc. *Siddha* system identifies diseases in the human body as irregularities of *vatha*, *pita* and *kappa* that could be induced due to environment and the activities performed. A balanced and appropriate diet is considered to play an important role in maintaining regularity of *vatha*, *pita* and *kappa*.

The use of metals and minerals is higher in *Siddha* system compared to *Ayurveda*, another traditional Indian system of Medicine. The formulations of different types like powders, pills, syrup, oils, dilute essence and thick paste are used in *Siddha*. *Kowsigar kuzhambhu* is one of the *Siddha* formulation prepared from constituents like *Piper longum*, *Terminilia chebula*, *Brassica Nigra*, *Ferula Asafoetida*, *Aconitum Lycotonum*, *Croton tiglium*, *Carum Carvi* etc. which individually themselves possess therapeutic activity.

### MATERIALS AND METHODS

The preparation involves purification of raw materials, followed by their mixing with herbal ingredients to achieve a product of semi-solid consistency called *Kowsigar kuzhambhu*. A flow diagram depicting the various steps in the preparation of *Kowsigar kuzhambhu* is outlined in Figure 1.

#### Purification of raw materials

##### Purification of red orpiment

About 200 g of crude red orpiment was ground with 300 mL of ginger juice for about 3 hours followed by grinding with 300 mL each of lime juice and butter milk in succession for 3 hours each. Purified red orpiment was obtained upon filtration of the mixture.

##### Purification of Croton tiglium

Crude Croton tiglium was taken in a clay pot filled with urine, boiled for 3 hours and filtered. The filtrate was discarded while the solid matter was taken in another clay pot filled with a mixture of water and cow dung. The mixture was boiled for 3 hours after which the same was filtered. The filtrate was discarded while the seed coat present on Croton tiglium was removed. Seed-coat removed Croton tiglium was taken in a clay pot filled with lime juice and heated to evaporate water. This was fried with ghee in an iron pan for about

10 minutes, before being subjected to wet-grinding with a mixture of *Pergularia daemia* and coconut milk to obtain purified Croton tiglium (Figure 2A).

##### Purification of mercury

Crude mercury was ground with brick powder for 6 hours followed by grinding with turmeric powder for 3 hours. *Acalifa indica* was added to this mixture and ground to obtain a paste of mercury and *acalifa indica* (Figure 2B).

##### Purification of Naabi

The crude naabi was taken in a cow urine-filled clay pot, exposed to sunlight for 8 hours a day for 3 days. The purified naabi was obtained after filtering the mixture and discarding the filtrate.

##### Purification of rock salt

The crude rock salt was ground in a pestle and mortar, before mixing with the juice of sour rice. The mixture was heated to evaporate excess water to prepare a paste of rock salt.

##### Purification of borax

The crude borax was roasted in an iron pan to produce a white powder of purified borax

##### Purification of arsenic trisulphate

The crude arsenic trisulphate was immersed in limestone solution and boiled for 6 hours before being dried under sun. The solid product after drying is the purified arsenic trisulphate.

##### Preparation of *Kowsigar kuzhambhu*

The purified ingredients in appropriate quantities were taken, to which *Acalifa indica* was added and mixed to prepare a semi-solid, green colored paste of *kowsigar kuzhambhu* (Figure 2C).

### RESULTS & DISCUSSION

#### Purification of red orpiment

The red orpiment is  $As_4S_4$ . The elemental analysis of crude red orpiment, determined by X-ray fluorescence spectrometry, showed the presence of Arsenic and sulphur as the major components along with Mg, Ca. The ratio of As to S in crude red orpiment was found to be 2.517, while the ratio of As to S in  $As_4S_4$  by stoichiometry is 2.34. The higher As to S ratio in the crude red orpiment indicates the probable presence of arsenic in a form other than safe  $As_4S_4$  form. Due to the sequential treatments with ginger juice, lime juice and

butter milk, the As to S ratio in the purified red orpiment is 2.42 closer to the value of 2.34 estimated by stoichiometry (Table 1). Hence certain quantity of arsenic present in form other than  $As_4S_4$  may have been removed during the treatment.

The composition of essential elements like iron, potassium and calcium has increased as a result of purification. Ginger contains iron, calcium and phosphorus while butter milk contains potassium<sup>1</sup>.

The increase in composition of Fe, K and Ca may be attributed to the incorporation of these essential elements from the purifying agents. The scanning electron micrograph of purified red orpiment (Figure 3) shows the presence of particles in the size range of 5-20  $\mu m$ .

This could be attributed to the trituration of red orpiment with ginger juice, lime juice and butter milk in succession, which simulates the wet-grinding in modern terminology.

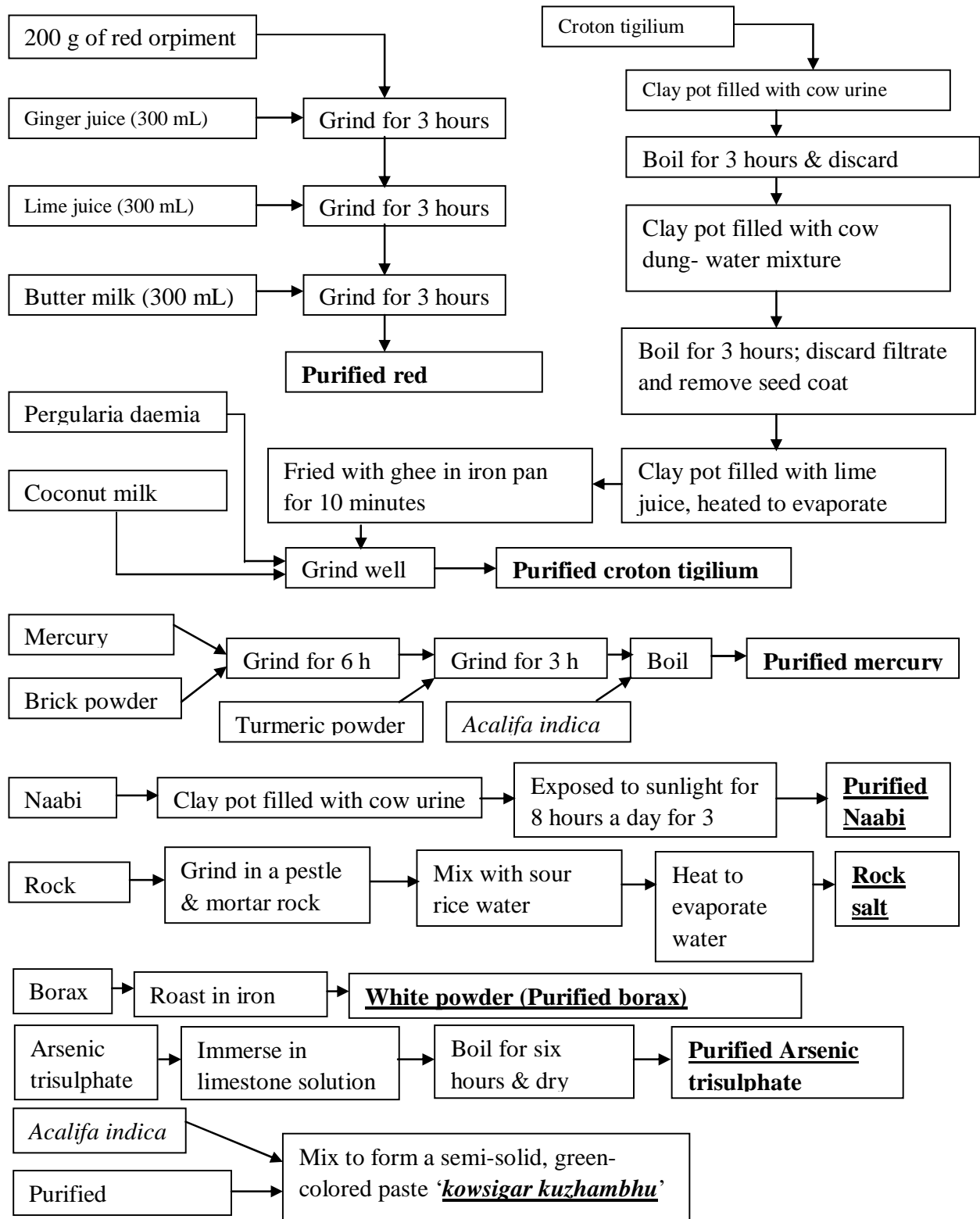


Fig. 1: Process flow diagram for the preparation of 'Kowsigar kuzhambhu'



Fig. 2: Photographs of [A] purified croton tiglium; [B] mixing mercury with herbal ingredients; [C] wet-grinding of ingredients with *Acalifa indica* to prepare *Kowsigar kuzhambhu*.

#### Purification of arsenic trisulphate

During the purification of arsenic trisulphate, limestone has been used. Lime stone is well known for its capability to remove toxic elements from water by precipitating them through the increase of pH<sup>2</sup>. However, the arsenic trisulphate used for the preparation contained arsenic, oxygen, sulphur only with the composition of heavy metals remaining below detectable limits (Table 2). Hence the changes in composition of arsenic, sulphur and oxygen due to purification are insignificant, except for the addition of sodium and calcium (from limestone).

#### Purification of rock salt

The purification of rock salt involves mixing the powdered rock salt with the juice of sour rice. One of the constituents of rice is phytic acid, a well-known chelating agent. The treatment of rock salt with sour rice juice may result in chelation of metals in rock salt with phytic acid. The reduction in composition of potassium, sulphur, calcium, magnesium (Table 3) in the purified rock salt may be attributed to their chelation with phytic acid.

#### Purification of borax

The purification of borax involves roasting in an iron pan, till dry white powder is obtained. Volatile components present in crude borax are expected to be removed during this treatment. This was confirmed from the increase in composition of sodium from 86.8 %

(crude borax) to 95.6 % (purified borax). Considerable reduction in percentage of sulphur from 2.2 % to 0.5 %, among other elements was also observed.

#### Composition of *Kowsigar khuzambhu*

The elemental composition of the final product (*Kowsigar kuzhambhu*) is shown in Table 4. The major elements are arsenic, chlorine, potassium, sulphur, calcium and sodium. However, these elements are present as compounds such as Arsenic disulphide etc. The rationale for addition of *acalifa indica* during the final stages of preparation of *kowsigar khuzambhu* may be of two fold (i) the organic constituents of *acalifa indica* may form complexes with the metal-based ingredients into a form suitable for easy absorption in the body (ii) the organic constituents added may have medicinal values that supplement the activities of the drug. The particle size distribution of *kowsigar kuzhambhu* is shown in Figure 4, from which it is evident that the product is composed of particles in the size range of 15-250  $\mu\text{m}$ . It may be recalled that most of the raw materials utilized for the preparation (before purification) were granular with sizes in the scale of several mm. The size reduction during the various purification and preparation steps may be attributed to the wet-grinding of granular materials in the presence of various extracts. This size reduction is accompanied with increase in surface area which facilitates better interaction between solid particles and liquid<sup>3,4,5,6,7</sup> during preparation and therapeutic stages.

Table 1: Elemental composition of crude red orpiment and purified red orpiment

Element	As	S	Mg	Si	Ca	K	Others
Crude red orpiment (%)	65.71±	26.11±	4.883±	0.190±	2.457±	-	< 0.66 %
Purified red orpiment (%)	62.76±	25.98±	4.673±	1.240±	2.883±	0.543±	< 1.35 %

Table 2: Elemental composition of crude arsenic trisulphate and purified arsenic trisulphate

Element	As	S	Al	Si	Al	K	Sb
Crude arsenic trisulphate (%)	46.40±0.5624	29.06±	2.787±	17.58±	2.463±	0.597±	1.667±
Purified arsenic trisulphate (%)	45.23±0.3024	28.83±	2.895±0.0707	17.64±	3.083±	0.7067±	0.340±

Table 3: Elemental composition of crude arsenic rock salt and purified rock salt

Element	K	S	Mg	Cl	Na	Ca	Fe
Crude rock salt (%)	7.737±	12.13±	4.847±	42.28±	25.76±	6.660±	0.060±
Purified rock salt (%)	2.437±	7.777±	3.557±	51.32±	29.75±	3.873±	-

Table 4: Major elements present in *kowsigar kuzhambhu*

Element	As	Cl	K	S	Ca	Na	Si	P
Composition (%)	20.35±	16.62±	12.54±	9.820±	6.283±	5.10±	2.33±	1.613±

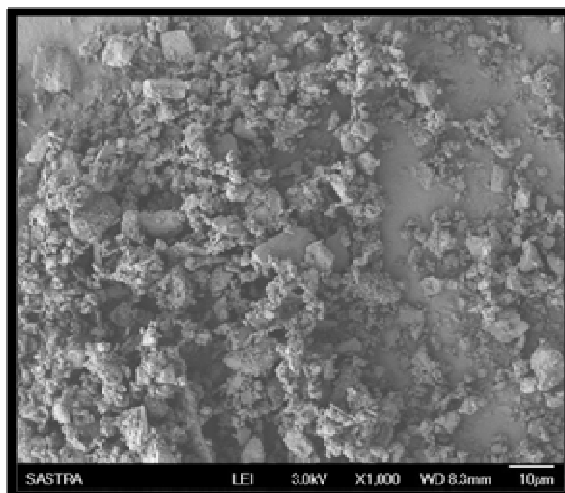


Fig. 3: Scanning electron micrograph of purified red orpiment showing the presence of particles in the size range of 5-20  $\mu\text{m}$ .

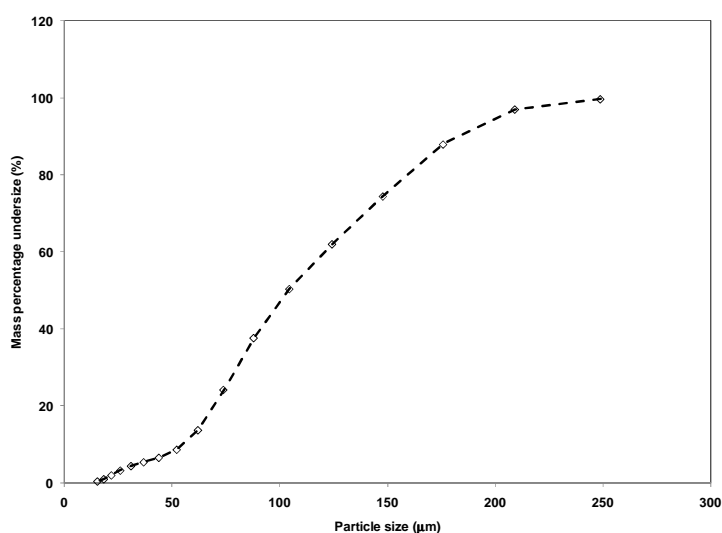


Fig. 4: Particle size distribution of *Kowsigar kuzhambhu*.

## CONCLUSIONS

*Kowsigar khuzhambhu* was prepared as per the procedures described in the classical literature. The role of herbal substances and treating liquids during the purification has been understood through the analyses of physico-chemical transformations accompanying these steps. Though the product contains arsenic, the same is present in the non-toxic form. The herbal ingredients present in the *Kowsigar khuzhambhu* facilitate the formulation of drug with appropriate particle size and consistency amenable for easy action in the body.

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