



## Research article

## Characterization of biotite drugs used in traditional medicine

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## ABSTRACT

Temperature-induced mineral alterations are extensively used in traditional pharmaceutical industry. Studies on the traditional heating methods for enhancing pharmaceutical properties and on the toxicity of mineral-based medicines are limited. This study focuses on the effect of thermal alterations on mineralogical and chemical changes of biotite with respect to two traditional drugs (Abhrak Bhasma and Abhrak Chendhuram). Samples of the drugs and heat-treated and untreated biotite minerals were characterized using X-ray diffraction analysis, Fourier-transform infra-red spectroscopy, thermogravimetric analysis and differential scanning calorimetry. Total and water-soluble cation concentrations of drugs were measured using atomic absorption spectrometry. The study reveals that the degree of collapsing the biotite structure increased with the thermal oxidation process that produced nanoparticles of crystalline and amorphous iron oxides and secondary silicates. The thermal products of biotite had nano-crystallinity and high water solubility. The study suggests that modern pharmaceuticals can be developed from mineral-based traditional drugs.

## 1. Introduction

Temperature and chemical induced alterations in phyllosilicates have been extensively studied regarding pharmaceutical applications [1]. Most widely studied varieties are artificially synthesized mica minerals and clay varieties in modern pharmaceutical industry [2, 3, 4, 5]. Minerals with pharmaceutical importance have specific physical and chemical characteristics such as high ion exchange capacity, high specific surface area, swelling property and solubility [6, 7]. However, such pharmaceutically favourable properties are somewhat limited in macromicas such as biotite due to their fixed interlayer cations. Hence, in traditional pharmaceutical industry, such properties are said to be induced by chemical and thermal treatments.

Biotite is one of the commonest mineral ingredients used in traditional 'Rasashastra' and 'Siddha' preparations in some South Asian countries such as India and Sri Lanka [14]. Biotite mica drug preparation process in traditional medicine involves a unique method of heating, quenching and incineration together with herbal, mineral or animal

materials [8, 9]. Altered biotite drug 'Abhrak' is extensively used together with appropriate herbal material by traditional physicians due to its proven antimicrobial, anthelmintic and hypoglycemic activities [13, 14].

Biotite mica is enriched with essential nutrients (such as Fe, Mg, K, Si, Zn and Mo) which help in body functions. Biotite also contains toxic trace elements Ni, Cu, Cr, Se, Pb etc. [10, 11]. However, both essential and toxic cations are limitedly available when the mineral is in unaltered form. Therefore, the digestion of biotite mica based products under gastrointestinal conditions needs further research [12, 13, 14]. Also, the characterization of biotite mica treated under traditional methods is important to understand the mobility and availability of toxic and nontoxic elements.

This research mainly focuses on the mineralogical and chemical characterization of two commercially available mica drugs Abhrak Bhasma (AbBh) and Abhraka Chendhuram (AbCh). Further, the results demonstrate that the alteration of biotite mica with regard to excessive calcinations at high temperatures effects enhanced solubility of the final products.

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## 2. Materials and methods

### 2.1. Sample description

Two mica-based drug samples Abhrak Bhasma (incinerated once) and Abhraka Chendhuram (mica incinerated thirty times) were purchased from Dabur India LTD, and the Indian Medical Practitioners' Co-operative Pharmacy & Stores LTD, respectively.

### 2.2. Mineralogical analysis

Mineralogical composition of drugs and mineral samples was determined using Siemens D-5000 X-ray diffraction (XRD) instrument. The particle size was estimated using "X-powder 12" software and manually using the Scherrer equation.

The temperature dependent changes during drug preparation were measured using KBr pellets on FTIR (Fourier-Transform Infra-Red)-Shimadzu IR Prestige 21 instrument. For comparison, two sets of reference biotite samples (biotite 1, biotite 2) were used; these samples were obtained after heating each sample twice at 1000 °C in a muffle furnace for 4 h.

Thermal characteristics of drug samples were studied using thermogravimetric analysis (TGA) and loss of ignition method (LOI). Differential scanning calorimetry (DSC) was used to study the nature of thermal reactions of biotite samples during dehydration/oxidation processes. Both the TGA and DSC analyses were performed on Scinco STA N-650 machine.

### 2.3. Chemical analysis

Total cation concentrations of drug samples were measured after acid digestion [15], using Perkin Elmer-2800 Atomic Absorption Spectrophotometer (AAS) and Inductively Coupled Plasma Mass Spectrometer (ICPMS-Perkin Elmer Sciex ELAN-6000). Water soluble portion of each drug sample was obtained by filtering each aqueous suspension that had been kept in a horizontal shaker at room temperature for 5 h and then allowed to equilibrate for 19 h. Analysis of the solutions was carried out on AAS. The instrument was calibrated using analytical grade standard solutions of each element. The accuracy of the analytical procedure was evaluated by analysing reagent blanks routinely with samples and by triplication of sub-samples. Detection limits of different elements ranged 0.001–0.04 mg/L for AAS while detection limits of ICP-MS ranged 1–0.001 mg/L.

## 3. Results

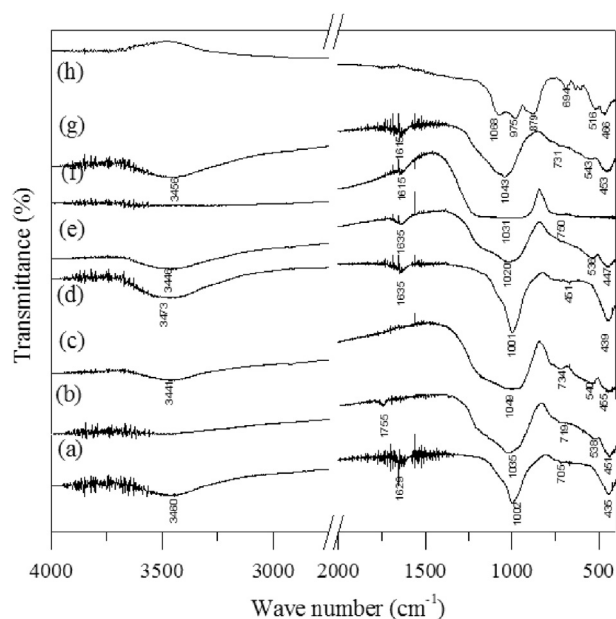
### 3.1. Mineralogical and structural variations

#### 3.1.1. Physical properties of heated mica samples

Thermally treated biotite mica showed a characteristic red colour with a golden lustre which increased with increasing number of heating steps. The golden lustre was higher towards the edges while there was a greyish lustre in the middle of biotite flakes after the first stage of thermal treatment. At the second thermal step, the flakes became almost reddish and brittle.

#### 3.1.2. Bond characteristics

The FTIR spectra of the two heat-treated biotite mica samples showed shifting as well as disappearance of some peaks compared to the unheated samples. The broader peak that appeared in the range 3480–3460  $\text{cm}^{-1}$  (Figures 1a and d) indicating the presence of structurally bound water or OH groups of unheated biotite mica, shifted towards lower frequency region with a reduction in peak intensity consistent with dehydration of biotite mica. Similarly, AbBh showed a low intensity broad peak at 3456  $\text{cm}^{-1}$  (Figure 1g). However, the corresponding peak was not present in the FTIR spectrum of AbCh (Figure 1h) indicating that water and



**Figure 1.** FTIR spectra of (a) unheated biotite 1 (b) biotite 1 heated at 1000 °C (c) biotite 1 heated twice at 1000 °C (d) unheated biotite 2 (e) biotite 2 heated at 1000 °C (f) biotite 2 heated twice at 1000 °C (g) Abhrak bhasma (AbBh) (h) Abhraka chendhuram (AbCh).

hydroxyls in the drug had been driven off during its preparation. All the heated samples showed the simultaneous disappearance of IR bands (Figure 1 b, c, e and f) in the range 1615–1755  $\text{cm}^{-1}$  corresponding to adsorbed moisture.

Appearance of new bands in heated samples at 516–543  $\text{cm}^{-1}$ , which can be assigned to the stretching vibrations of the Fe–O bond, suggests the presence of  $\text{Fe}_2\text{O}_3/\text{Fe}_3\text{O}_4$  particles [16]. FTIR analysis shows that Fe–O bonds are absent in the unheated biotite samples. The peaks at 435–439  $\text{cm}^{-1}$  represent the Fe–O–Si bonds in the structure. These two peaks are slightly more intense in the samples heated twice (Figure 1 a-h) indicating that the amount of iron oxides increased with the number of heating steps. Similar bands at 516, 543 and 453  $\text{cm}^{-1}$  observed from the two mica drug samples (AbBh and AbCh) correspond to Fe–O stretching and bending vibration modes [16, 17].

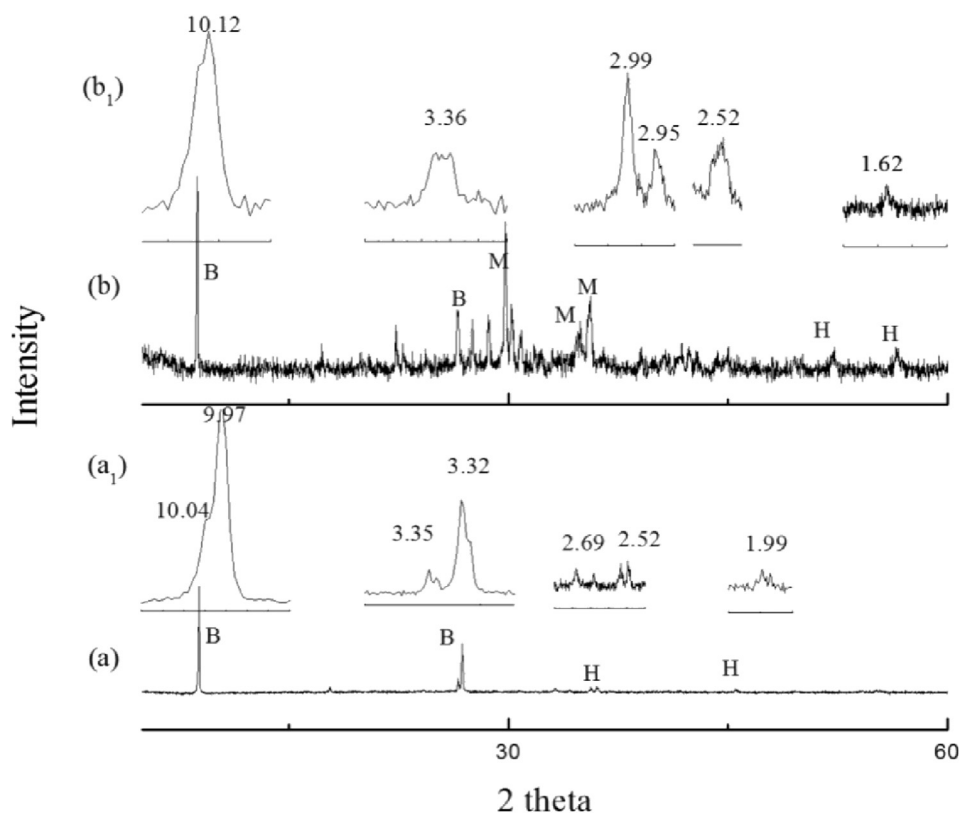
In addition, AbCh (Figure 1h) showed much stronger bands at 650 and 694  $\text{cm}^{-1}$  than AbBh indicating an additional iron oxide phase which could probably be a secondary mineral phase like magnetite arising from excessive heating [18].

Characteristic Si–O stretching bands centered at 1000  $\text{cm}^{-1}$  in unheated mica samples showed significant broadening and shifting towards either side and ranged 1250–900  $\text{cm}^{-1}$ , upon heating. At the same time AbCh showed split peaks in the same range signalling alterations or distortions associated with silica tetrahedron sites [19].

#### 3.1.3. Structural characteristics

The position and intensity counts of the XRD peaks of the treated biotites and drug samples showed some difference to those of untreated biotite mica (Figure 2 a - b<sub>1</sub>) and it may be a result of thermal oxidation and dehydration of samples. The interlayer thickness of the two mica drugs AbBh and AbCh were 9.97 and 10.12 Å, respectively. However, typical biotite micas generally have interlayer thicknesses of 10.04–10.1 Å. The three splitted peaks in both AbBh and AbCh (Figure 2 a<sub>1</sub> and b<sub>1</sub>) confirm the structural distortions and alterations associated in the silica tetrahedral region as noted in the FTIR spectra.

The XRD diagrams of AbCh showed that peaks could be attributed to iron oxides (see Figure 2a<sub>1</sub>, b<sub>1</sub>) in two forms, as hematite and magnetite [17]. However, the magnetite peaks (2.99 and 2.95 Å) were not prominent in the XRD pattern of AbBh; very low intensity peaks corresponding



**Figure 2.** XRD spectrum of (a) AbBh (a<sub>1</sub>) enlarged peaks of AbBh (b) AbCh (b<sub>1</sub>) enlarged peaks of AbCh (B- biotite, M-magnetite, H- hematite).

to hematized iron oxides were identified. These observations are consistent with the fact that the two drugs AbBh and AbCh had been prepared using different thermal treatments. The estimated size of the drug particles using Scherrer's formula ranged 75–500 nm for biotite drug particles and 15–100 nm for FeO–Fe<sub>2</sub>O<sub>3</sub> particles. The remaining peaks with low intensities corresponding to other phases indicate further alterations of initial biotites into secondary clay minerals [20] during the drug preparation.

### 3.1.4. Thermal characteristics

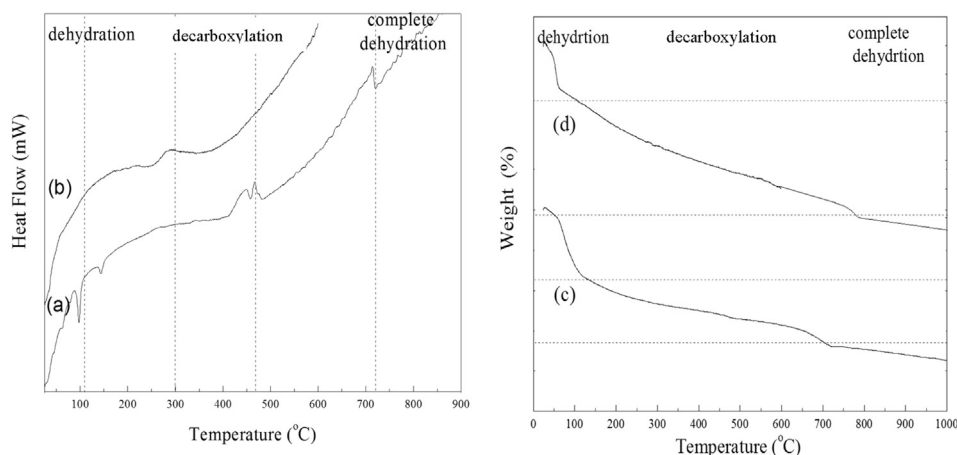
The DSC curve (Figure 3a) of AbBh consists of three shallow minute peaks; one endothermic peak at 105 °C and two endothermic peaks at 475 and 725 °C. These three peaks may represent heat absorption for the removal of adsorbed moisture, oxidative removal of organic matter and

dehydration of structural hydroxyls, respectively. AbCh showed only one endothermic peak around 250–350 °C in its DSC curve (Figure 3b).

The weight-loss curves (TGA) of both AbBh and AbCh comprise three main regions (Figure 3c, d); a low-temperature region (0–105 °C) with steep gradient of weight loss, intermediate-temperature region (105–600 °C) with low gradient of weight loss and high-temperature region (725–1000 °C) with low weight loss, corresponding to expulsion of adsorbed moisture, oxidative removal of organic matter and dehydration of structurally bound water molecules, respectively.

### 3.1.5. Chemical composition and water solubility of mica drugs

AbBh had higher concentrations of Fe, Mn, Ti, Zn and Al cationic constituents, which were presumably inherited from biotite mica [23], than AbCh (Table 1). However, the Ca concentration in AbCh was



**Figure 3.** DSC curves of (a) AbBh (b) AbCh and TGA curves of (c) AbBh (d) AbCh.

**Table 1.** Comparison of water soluble cations in AbBh and AbCh with respect to total cations and the raw biotite in different liquids.

		Cation concentration (ppm)										
		Na	K	Ca	Mg	Fe	Mn	Zn	Cu	Pb	Cd	Sr
Total ion (n = 4)	AbBh	1280	48000	1210	23000	176000	1330	331	65	9	5	26
	AbCh	11000	39000	120000	87000	73900	825	312	202	33	6	720
water solubility (n = 4)	AbBh	3505	4800	2100	2100	1900	bd	bd	bd	bd	bd	bd
	AbCh	1725	4000	4800	1700	700	bd	bd	bd	bd	bd	bd
<sup>a</sup> water solubility	raw bt	61	378	6	53	25	bd	bd	bd	bd	bd	bd
<sup>b</sup> citric acid solubility	raw bt	132	1650	nd	234	984	bd	bd	bd	bd	bd	bd
<sup>c</sup> acetic acid solubility	raw bt	50	3800	140	475	350	40	16	7	bd	bd	bd
<sup>d</sup> HCl (pH 1.27)	raw bt	1023	683	641	724	580	6.4	nd	nd	nd	nd	nd

bd - below detection limit; n - number of samples; nd - not determined; rawbt - raw biotite; a, c-authors' unpublished data; b- [24]; d- [41].

unusually high compared to those of both AbBh and untreated raw biotite mica [21, 22]. In addition, Mg, Na and P concentrations were significantly high in AbCh compared to those of untreated raw biotite and AbBh samples (Table 2). This indicates that the respective cations were introduced into the final products by other ingredients during the preparation of the drugs.

The cations Mn, Zn, Li, Rb, Sn and Cs were found in trace amounts in both AbBh and AbCh samples compared to those in raw biotite, implying the expulsion of those cations during the preparation of the drugs. However, relatively higher amounts of cations such as Pb, Ni, Cu, Cr and V were observed in both drugs.

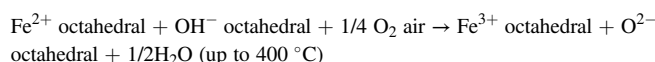
Water solubility of toxic elements such as Mn, Zn, Cu, Pb, Cd and Sr was below the detection limit for both drugs. Both drugs showed significantly higher solubility of the cations like Na, K, Ca, Mg and Fe in water. These values were considerably high compared to the solubility of raw biotite mica in water and some of the organic and mineral acids such as citric, acetic and HCl (see Table 1) [24]. This indicates that the amounts of bioavailable essential and non-toxic chemical constituents had enhanced during the drug-preparation process.

## 4. Discussion

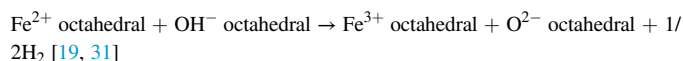
### 4.1. Effect of temperature on structural alterations

The enhancement of golden lustre and reddish colour of heated biotite mica with repeated heating reflects the increased extent of oxidation [25]. Further, the brittle nature indicates that the interlayer bonds had weakened upon thermal exposure. Brittleness is developed mainly by raising compressive stresses within the mineral structure, which increases the volume during oxidation. The stresses lead to the development of cracks and spallations in the micaceous plates and facilitate the penetration of organic liquids—herbal extracts used during the drug preparation, decoctions, neat extracts of fruits etc.—in the mixture into

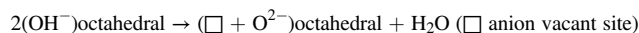
the inner layers of mica. Thermal oxidation of biotite results in chemical and structural alterations [26, 27] which in turn aids to obtain fine drug particles which can be absorbed more easily by the human body. Spectroscopic and thermal data obtained from the biotite mica samples 1 and 2 showed decomposition of the mineral at several stages. At the initial stage biotites were thermally modified by reacting with oxygen in the surrounding air.



At the same time, a redox reaction takes place between hydroxyls attached to the octahedral cations within the structure and the octahedral  $\text{Fe}^{2+}$  ions, causing expulsion of structural hydrogen and oxidation of further ferrous ions to ferric ions [28, 29, 30]. Hydrogen may diffuse towards the edges of biotite plates and gets oxidized to water molecules when in contact with the organic liquids during the drug preparation.



Remaining hydroxyls in octahedrons detach from the structure and are completely removed as water molecules at temperatures above 800 °C. Further, it causes the creation of anion vacancies within the structure and leads to exfoliation of original single sheets of mica into much thinner units [19, 28, 31].



The absence of hydroxyl peaks in AbCh (Figure 1g and h) and lower gradient of weight loss in TGA curves (Figure 3c and d) reflect that AbCh had been subjected to more exposure to heat than AbBh.

As the  $\text{Fe}^{2+}$  ions in octahedrons are largely converted to the relatively smaller  $\text{Fe}^{3+}$  ions upon repetitive heating, octahedron positions get shrunk with developing cation vacancies. To keep the charge balances in

**Table 2.** Representative bulk chemical compositions of AbBh, AbCh drugs and raw biotite.

	Mn	Zn	Li	Rb	Sn	Cs	Ba	Sr	Pb	V	Cr	Co	Ni	Cu	Sc	
Trace elements (ppm)																
raw bt	2870	582	606	1300	10.5	19.5	159	3	5.7	6	8	11.5	3.4	47.7	4	
<sup>e</sup> raw bt	3262	1300	1200	2200	70	180	150	5	13	135	90	23	35	5	14.8	
AbBh	1770	341	90.7	427	1.5	16.7	500	22	10.2	106	120	57.6	85.1	75.1	57	
AbCh	990	322	11.5	121	8.7	1.2	4110	815	38.5	110	145	70.4	323	211	27	
Major elements (wt. %)																
	Fe		Mg		Al		Ti		K		Ca		Na		P	
raw bt	25.2		0.90		6.14		0.408		4.34		0.05		0.065		0.003	
<sup>e</sup> raw bt	21.3		2.36		5.71		0.039		7.71		0.10		0.012		nd	
AbBh	18.7		3.02		6.63		0.575		4.72		0.11		0.152		0.011	
AbCh	7.82		9.26		3.6		0.214		3.73		11.9		1.08		0.599	

e- [42], not determined - nd.

the oxidized layers, some of the  $\text{Fe}^{3+}$  ions are ejected out from the biotite mica structure. Therefore, heating leads to buckle the existing octahedrons. The remaining octahedral ions such as  $\text{Al}^{3+}$  are expelled consequently facilitating the transformation to stable ditrigonal arrangement of silicates [32].

$3(\text{Fe}^{2+})_{\text{octahedral}} \rightarrow (\square + 2\text{Fe}^{3+})_{\text{octahedral}} + \text{Fe}\downarrow$  ( $\square$  cation vacancy in octahedral site)

Due to the charge imbalances created by the repetitive thermal oxidation in adjacent layers, structural components would collapse and produce secondary materials of biotite mica [26, 33, 34, 35].

Thus, ion oxides dissected from the biotite structure are found as fine amorphous residues and microcrystalline magnetite and hematite phases. Further, the distortion of silica tetrahedral layers indicated by the FTIR studies confirms the above arguments. Thus, the rearrangement of secondary silicates and reduction of particle size into nanoscale dimension are possible [29, 30, 36] upon heating during mica drug preparation. Since fine particles and amorphous materials can increase the availability of metallic constituents, extensive heating is conducive to both destruction of mineral structure of biotite and production of materials that can easily extract nutrients.

#### 4.2. Chemical composition and dissolution behavior of mica drugs

The variation of major and trace element contents of samples (Tables 1 and 2) can be accounted for by the compositional variability of raw mica and the preparation procedures adopted. The use of acidic herbal liquids and inorganic salt rich media during the drug preparation might have assisted the removal of oxidized toxic elements present in raw materials by forming soluble complexes [37]. The removal of interlayer  $\text{K}^+$  ions is facilitated by the exposure of the cations in the octahedral and tetrahedral sites to the outer aqueous medium. The removal of interlayer cations from the biotite structure provides a better path for eliminating the rest of the ions from the biotite structure [36]. Further, exfoliated edges in thermally altered mica sheets provide additional space for leaching out of ions. Micro-crystalline and amorphous-oxides as well as secondary silicates have higher surface area compared to raw biotite. The presence of comparatively lower amounts of  $\text{Fe}^{2+}$  ions and  $\text{K}^+$  ions observed in acid digested AbCh and AbBh drug samples reveals the leaching out of cations from their weakened structures. Less soluble silica components seem to remain in distorted and amorphous forms (Figure 2a<sub>1</sub> and b<sub>1</sub>). Also, the higher amounts of  $\text{Na}^+$ ,  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  ions and the presence of P (may be as  $\text{PO}_4^{3-}$  anions) in AbCh and AbBh could result from the mixing of other inorganic or organic ingredients.

The enhancement in solubility of cations that occupy the interlayer and octahedral sites of biotite reflects compositional modifications resulting from thermal treatments perhaps together with herbal treatments. Such treatments also bring down the level of toxic elements of drug samples below the detection limits. The production of nanoscale iron-rich crystalline and amorphous materials and the destabilization of mica structure are the ultimate outcome of the traditional method.

Under acidic conditions in the stomach the dissolution of the drug can increase [38, 39]. The biocompatibility of the drug and bioavailability of ions could be further improved for consumption when the drug is mixed with organic ingredients such as milk, butter, honey, ghee or herbal decoction [40]. It is noted that the levels of some toxic elements such as Cu and Zn, although they are not water soluble, are relatively high in the two traditional drugs compared to most of the modern oral pharmaceuticals. Further investigations are needed to unravel their bioaccessibility under physiological conditions.

#### 5. Conclusion

Drug preparation involving heating steps caused alterations in both the structure and chemistry of raw biotite. The physical structure of

biotite was distorted at the octahedral sites as a result of oxidation and ejection of ferric ions. Further, the changes in the interlayer distances reflect the collapsing of interlayer bonds among the biotite flakes. Such structural changes can occur when raw biotite is subjected to thermal processes and exposed to organic extracts (herbal extracts) during drug preparation. Nanoparticles of crystalline and/or amorphous iron oxides and secondary silicates could be formed by the materials ejected from the biotite during the above alterations. Increased solubility of altered products could be a result of the favourable solvation interactions between the enhanced surface area of the solutes and the liquids. The temperature induced disintegrations can thus be utilized to enhance the solubility of desired medicinal constituents. Chemical analyses confirm the expulsion of undesired toxic elements such as Mn, Zn, Li, Rb, Sn, and Cs during drug preparation.

#### Declarations

##### Author contribution statement

Apsara Wijenayake: Conceived and designed the experiments; Performed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

Amarasooriya Pitawala, Ratnayake Bandara, Charmalie Abayasekara: Conceived and designed the experiments; Analyzed and interpreted the data; Contributed reagents, materials, analysis tools or data; Wrote the paper.

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##### Competing interest statement

The authors declare no conflict of interest.

##### Additional information

No additional information is available for this paper.

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