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CHEMICAL AND STRUCTURAL CHANGES IN BIOTITE DURING PREPARATION OF *AYURVEDIC* MEDICINE

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ABSTRACT

Biotite mica is used in the preparation of some kinds of *Ayurvedic* medicine. An investigation was undertaken to study the elemental exchange between biotite and other ingredients such as vinegar, cow-urine, paddy and jaggery which are used in the purification of biotite and the preparation of mica-*basma*. Further, the structural changes in biotite during the processes of preparation of medicine were also studied.

Present study revealed that the pH of the cow urine decreased with time, when the heated biotite mica was immersed in the solution. In contrast, the pH of the vinegar-mica mixture increased with contact time. It was also observed that cow urine can absorb some major elements (Mg, K, Na, and Fe) and trace elements (Mn and Zn) of mica better than vinegar. When mica was subjected to traditional cooking treatment repeatedly, a gradual breakdown of the biotite structure was noted. The results of the study indicate that paddy, jaggery and cow-urine act as agents to remove toxic elements of mica thereby providing some nutrients to the mixture. The accumulation of mineral nutrients into the mixture has taken place gradually with the destruction of biotite structure. The final mica mixture can be considered as a product of elements derived from biotite and other ingredients.

Keywords: biotite mica, Ayurvedic medicine, heat treatment, mica-*bhasma*, cow-urine

INTRODUCTION

The use of fossils, minerals, and rocks for healing dates back to thousands of years. Different people all over the world have applied these geologic materials in various forms to soothe and cure. Today many countries manufacture tonnes of pharmaceuticals from minerals and fossils. Various traditional and non-traditional medical disciplines take advantage of these earth materials. Research on their healing effects are rather scarce. Over eighty minerals and mineraloids were documented for medicinal purposes in the past and present. The clay minerals lead the list, followed by quartz, amber, hematite, pearl and malachite (Limpitlaw, 2004).

Medicinal rocks and minerals have been used extensively in Ayurveda treatments. The word "Ayurveda" is composed of two Sanskrit terms, "ayu" meaning "life" and "veda" meaning "knowledge" and taken together, it means the "Science of Life". It is a natural healing system used throughout India for more than 5000 years, and which later spread throughout Sri Lanka. Ayurveda was known to have been first developed and established by the great sages who developed India's original systems of meditation and yoga, i.e. it has originated as part of the Vedic science.

Ayurveda is part (upaveda) of the Atharva Veda of four Vedas. Several other traditional systems had been developed in various countries as China, Middle East, Greece and Egypt. But some of them have not persisted due to ignorance of fundamental principles (Dash and Vaidya, 1978, Samarasekara, 2003).

For the *Ayurvedic* medicines, natural materials are being used as raw materials. Materials used in *Ayurveda* belong to three different categories. The name *Udbhida* is used for whole plant or its parts, whereas *Jangama* is used for parts of animals (e.g.: horns, shells). Metals and minerals used in *Avurvedic* medicine are known as *Bhauma*. When preparing drugs, these are used separately, but most often the drugs are mixtures of all these materials. The science that deals with metals, gems and minerals is known as 'Science of Alchemy' (*Rasa shastra*). The resulting medications are known as *"rasas"* which mainly comprise of metallic ashes called *bhasma*. For the preparation of medicines, several common rock forming minerals, gems, metals and other inorganic compounds are used. During the processing, all ingredients are subjected to repetitive processes of purification *(shodana)* and incineration *(marana)* to bring them to the state of ash *(bhasma)* before the ingredients are added to the respective formulations. Minerals and metals may consist of impurities and toxic substances that are harmful to the human body. Therefore, all raw materials used in Rasa medicine need to be purified to eliminate such impurities and toxic substances, before they are used for medicinal purposes.

Mica, a phyllo-silicate, is one of the mostly used minerals in *Ayurvedic* treatments. Biotite (black mica) is the most therapeutically effective mica among the mica group minerals (Baily, 1984). Purification of mica is carried out before it is used as a medicine. Purification may remove the harmful substances or impurities that are present in minerals. Biotite preparation processes reduce the substances into fine particles so that they can be properly digested and absorbed easily into the system and mixed with the *dhatus*. Incineration *(Marana)* processes are also used to increase the natural property of the minerals and new properties are sometimes introduced into the mineral compound.

There have been some studies carried out to study the chemical effects of medicinal rocks used in *Ayurvedic* treatment in Sri Lanka (Hegoda *et al*., 2007, Hegoda, 2006, Samarakoon, 1995 and Silva, 1995). The present study focuses to identify the contribution of the biotite mica as (a) a material that may supply nutrients to the *bhasma* for the *Ayurvedic* medicine and (b) a medium which can absorb nutrients from other materials used for the preparation of the mica-*bhasma*. Further, it was intended to examine the effect of heat treatments on the ingredients of traditional medicines.

METHODS OF STUDY

Biotite was treated with both cow-urine and vinegar which are commonly used in the preparation of mica-*bhasma*. Biotite samples were heated seven times till they were red hot and then quenched with cow urine or vinegar at each heating step. The heat-treated mica samples were immersed in cow urine for a period of one month. During the traditional method, the treated mica was mixed well with paddy and cow urine and separated by squeezing through a cloth. Similar procedure was followed for a sample and it was further mixed with jaggery and made into a muddy paste by adding cow urine as prepared in the *ayurvedic* system of medicine. Small pellets were prepared from the mixture and heat treated in an oven (*puta*). When cooled, pellets were ground and made into a muddy paste by adding cow-urine and made into pellets again. This procedure was repeated 30 times.

For chemical and mineralogical studies, the mixture was separated by filtering. The chemical composition (Fe, Mn, K, Mg, Ca, Zn) of the filtrate was analysed using Perkin Elmer-2800 Atomic Absorption Spectrometer (AAS) at the Department of Geology, University of Peradeniya, Peradeniya. The pH of each sample was measured prior to treatments, just after mixing the ingredients and before filtering. X-Ray diffraction analysis was performed on treated mica samples at the Department of Chemistry, University of Peradeniya.

Sample Descriptions

Fresh biotite samples were taken for the analysis and treated with different methods as shown in the Table 1.

RESULTS AND DISCUSSION

The pH of all treated cow-urine samples decreased with increasing the number of times of treatments whereas the pH of all treated vinegar samples had increased. The pH of cow urine had noticeably changed compared to that of vinegar treated in the same manner (see Figures 1a and 1b). However, the samples C7 and C8 (mixtures of heated biotite in the cow-urine for a period of one month) show

abnormal variations. The pH variation of each solution may be due to (a) substitution of partly dissociated H^+ in water into the position of K^+ in the biotite structure, (b) releasing of Al_3^+ from the biotite or (c) releasing of OH^- or F^- of mica into vinegar.

Chemical Changes of Cow-Urine and Vinegar after Treatment

Chemical analyses for the filtrates were carried out using the atomic absorption spectrometry and the summary of the results are shown in Table 2. Concentrations of treated biotite were plotted against the treated conditions (Figure 2). Although the concentrations were measured for eight elements, changes in the concentration of the filtrate of iron, calcium, magnesium and potassium were considered to be most significant during the treatments.

Biotite is a sheet silicate and has very strong silicate-oxygen bonds. Aluminium usually replaces Si^{4+} in the tetrahedral position in most biotite structures.

Table 1: Description of the treated biotite samples used in analysis

Sample Name	Treatment Method of Biotite	Further Treatments
B_1	Biotite was heated seven times till it was red hot and quenched in **cow urine** at each heating step and then placed in cow urine for one month and treated with a mixture of paddy and cow urine	C_0 – Cow Urine C_1 – One time heated biotite with cow urine C_2 - Two times heated biotite with cow urine C_3 - Three times heated biotite with cow urine C_4 - Four times heated biotite with cow urine C_5 - Five times heated biotite with cow urine C_6 - Six times heated biotite with cow urine C_7 - Seven times heated biotite with cow urine C_8 – Non-heated biotite with cow urine C_9 -Biotite after *Dhanyabra process** with cow urine
B_2	Biotite was placed in cow urine for a period one month and then treated with a mixture of **paddy** and **cow urine**	C_{10} -Biotite after *Dhanyabra process** with cow urine
B_3	Biotite was heated seven times till it was red hot and then quenched in **vinegar** at each heating step and then placed in vinegar for a period of one month and treated with a mixture of paddy and vinegar	V_0 – Vinegar V_1 – One time heated biotite with Vinegar V_2 – Two times heated biotite with Vinegar V_3 – Three times heated biotite with Vinegar V_4 – Four times heated biotite with Vinegar V_5 – Five times heated biotite with Vinegar V_6 – Six times heated biotite with Vinegar V_7 – Seven times heated biotite with Vinegar V_8 – Non-heated biotite with Vinegar V_9 - Biotite after *Dhanyabra process** with vinegar
B_4	Biotite was placed in **vinegar** for a period of one month and then treated with a mixture of paddy and vinegar.	V_{10} -Biotite after *Dhanyabra process** with Vinegar

***** Incineration with paddy

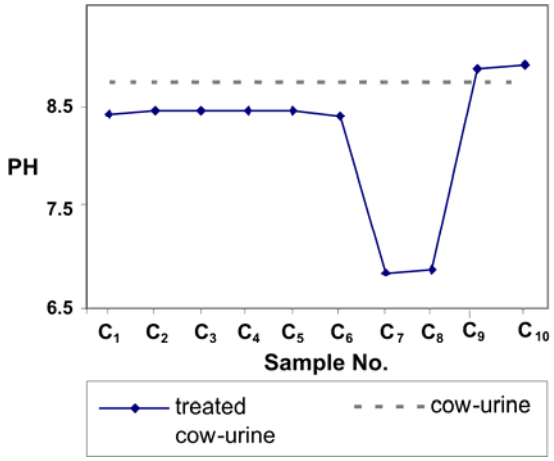

Figure 1(a): pH variation of cow-urine after treatment (refer Table 1 for sample nos.)

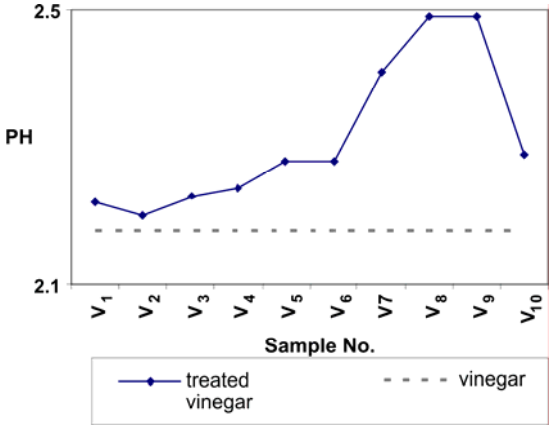

Figure 1(b): pH variation of filtrate of the mixture of vinegar and biotite (refer Table 1 for sample nos.)

Table 2: Elemental concentrations of treated and untreated mixtures

Elements (ppm) / Sample No.	Fe	Na	K	Mg	Ca	Zn	Mn
C_0	0.4	19.25	40900	128	11.3	0.8	0.1
C_1	0.64	18.35	34600	449	12.9	0.7	0.2
C_2	1.4	25.35	90000	451	44.3	1.6	0.2
C_3	1.28	23.05	60000	401	47.3	5	0.2
C_4	1.2	28.05	13600	445	46.4	0.6	0.2
C_5	0.59	27.95	20000	382	46.	0.5	0.2
C_6	0.56	24.55	66400	449	42.9	0.8	0.2
C_7	25.6	30.75	100000	752	19.4	3	0.8
C_8	0.77	19.60	401000	556	44.8	0.8	0.5
C_9	2.04	19.30	420000	463	19.8	2.3	0.3
C_{10}	1.04	28.60	413000	450	15.8	2	0.2
V_0	1.01	4017	423	129	8.09	0.8	0.3
V_1	4.24	6975	129	389	10.08	1	0.4
V_2	5.8	2620	236	165	8	0.9	0.5
V_3	4.24	5225	470	161	5.47	1.6	0.5
V_4	4.55	3540	400	157	6.32	1.7	0.5
V_5	4.68	5065	580	159	5.81	1	0.5
V_6	5.43	2955	107	157	5.48	2.7	0.5
V_7	28.3	5615	141	257	5.47	2	0.2
V_8	24.4	7490	120	491	89.3	0.8	0.4
V_9	4.4	5625	217	825	47.7	2.2	1
V_{10}	4.82	4650	281	931	27.9	2.7	0.9

Breaking of Al-Si-O tetrahedra in low temperature conditions is rather difficult (Baily, 1984). Interlayer cations of biotite are usually a form of very weak van der Waal bonds and potassium and sodium ions are the usual occupants in such positions. The octahedral position of biotite is usually occupied by divalent cations like Mg^{2+}, Fe^{2+}, Zn^{2+} and Mn^{2+} etc. Therefore, potassium, magnesium, iron, sodium, manganese and zinc are common ions which can easily be liberated to the filtrate.

Results show that higher content of iron has been released into the filtrate from the heat treated samples compared to that of non-heated samples (Figure 2, C8 &V8). Measured calcium values in the filtrates are not considerable except those treated with paddy only. It indicates that the calcium accumulation in the filtrate has been taken from the used paddy (see Figure 2). On the other hand, calcium is not a popular occupant in the octahedral position of biotite. However, accumulation of calcium in some filtrates is observed and it may be due to impurities in biotite which occur as mineral inclusions.

As shown in Figure 2, variation of the potassium content in the filtrates is inconsistent. Higher concentration of potassium in the cow-urine treated with biotite and paddy, and lower concentration of potassium in the cow-urine treated only with biotite, shows that higher potassium accumulation in the cow-urine has been taken from the paddy. The potassium accumulation into the vinegar filtrate is not considerable.

Analytical results indicate that treated cow-urine absorbed more magnesium than the treated vinegar (Figure 2). Higher levels of available magnesium in the vinegar, mica and paddy mixture could have come from paddy and not mica.

The releasing of higher amounts of magnesium, sodium, manganese and zinc from heated biotite than from non-heated biotite suggests that heat treatments enhance the chemical reactions of mixtures. During a 1 month period, the concentration of available elements in the mixtures gradually increased with contact time. The accumulation of potassium in cow-urine is higher than that in the vinegar. Analytical results show that heated biotite treated with a mixture of cow-urine and paddy has released higher amounts of magnesium, potassium and zinc than all other biotite samples.

Structural Changes of Biotite

XRD patterns of graphs show that diffraction angles of the biotite treated with cow urine have shifted, and the intensity of peaks has decreased relative to the untreated biotite (B) (Figure 3). XRD patterns of biotite treated with the vinegar mixture do not show a marked variation (Figure 4). In contrast, XRD patterns show significant decrease of peak intensity along with slight changes of diffraction angles in the biotite treated in *puta* (Figure 5).

Mica is an inert mineral which does not change its basic structure thermally and chemically even under extreme conditions. However, the interlayer cations in mica can be exchanged with other available ions, without changing the structure. Structure of the treated biotite has changed from that of the non-treated biotite. According to intensity of peak values and diffraction angle, variation of structure of the heated biotite is more intense than those of the not-heated biotite. XRD pattern of heated biotite treated with a mixture of cow-urine and paddy shows a significant change in the biotite structure.

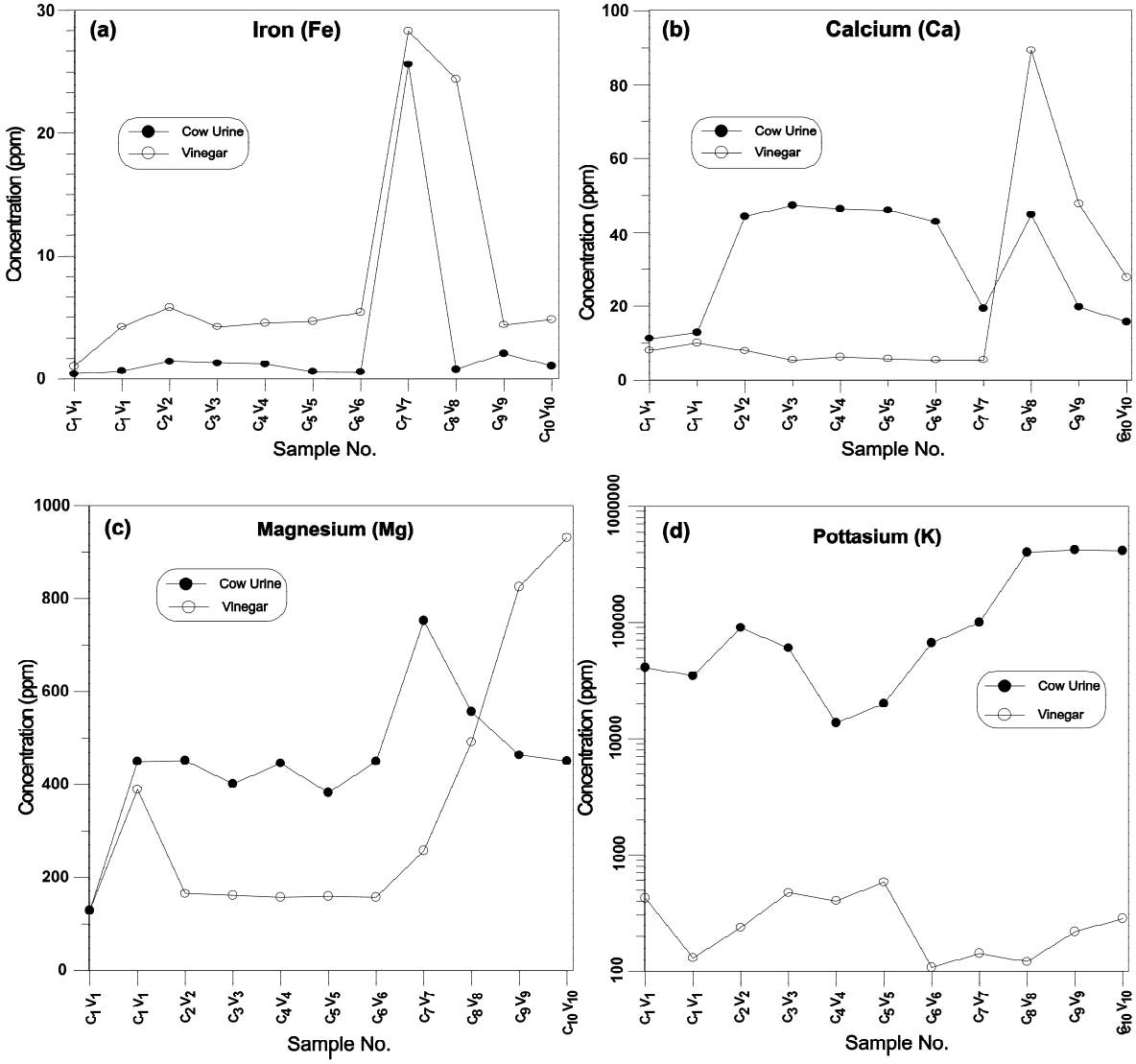

Figure 2: Concentrations of some selected elements in treated cow-urine and vinegar: (a) iron, (b) calcium, (c) magnesium, (d) potassium

The structure of the treated biotite gradually changed with increasing number of treatment. In general, the structure of the biotite sample cooked in oven for 30 times shows a significant to contrast from the structure of un-treated biotite. Hoiwever in a biotite sample cooked in puta (oven) for 30 times, the structure of the biotite stands unchanged. In literature, it says biotite cooked in puta (oven) for 1000 times. It may cause to total distortion of biotite structure. During cooking process, cow-urine and jaggery are mixed with biotite. Organic compounds adding to the biotite structure from jaggery, cow-urine and others may be destroyed due to the cooking of the biotite in the oven. Further, all ions in the cow-urine are added the biotite structure during the latter stages of cooking in oven 1000 times. Based on the results of the present study, it is revealed that final product of biotite mica is a result of elements both in the biotite structure and in the absorbed ions from other materials used for the preparation of the mica-*bhasma*.

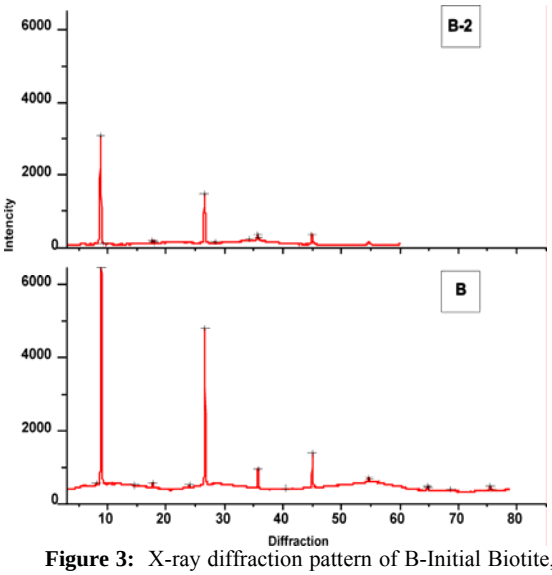

Figure 3: X-ray diffraction pattern of B-Initial Biotite, B-2 (Biotite was heated seven times and left immersed in cow-urine for a month)

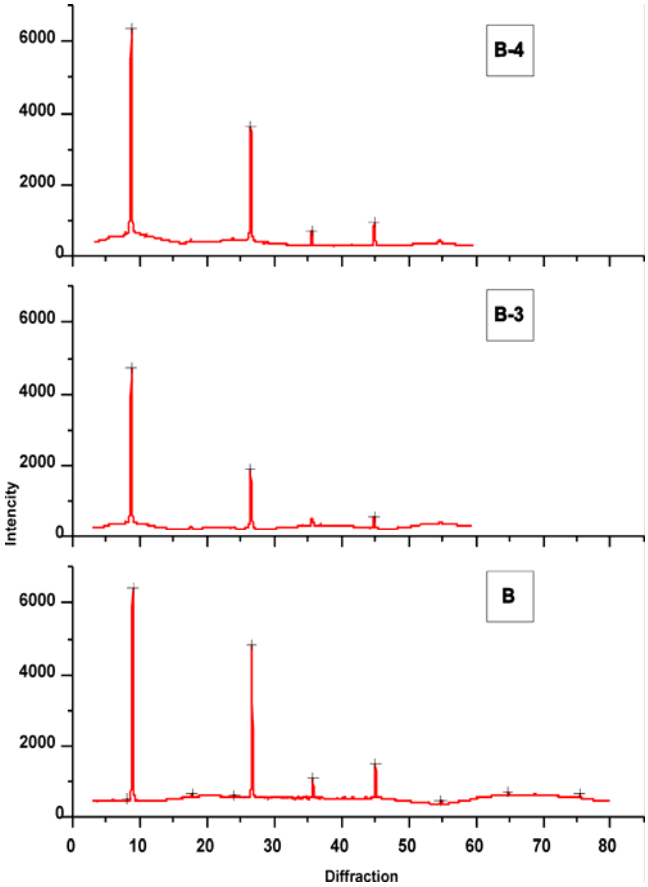

Figure 4: X-ray diffraction patterns of B - (untreated biotite), B-3 (heated seven times till it was red hot and quenched by vinegar at each heating step and after placing in vinegar for one month period and treated with mixture of paddy and vinegar), B-4 (placed in vinegar for one month period and treated with mixture of paddy and vinegar).

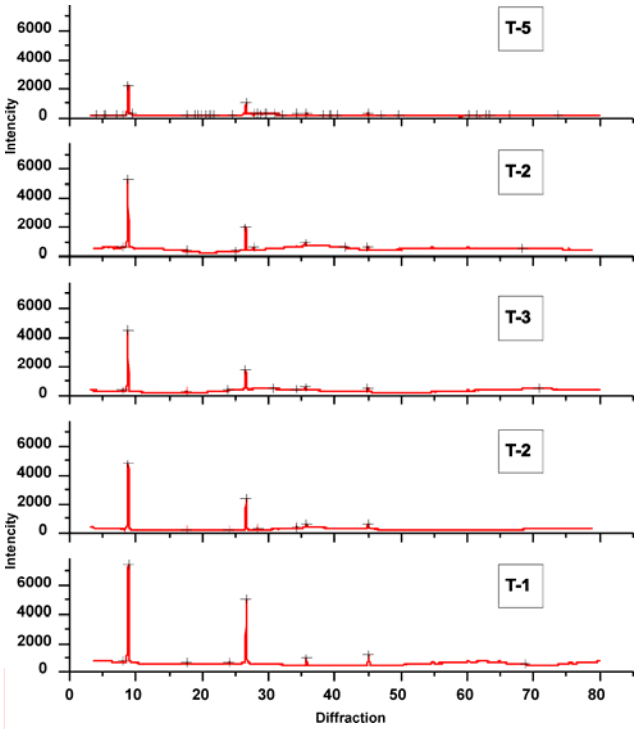

Figure 5: X-ray diffraction pattern of T-1 (untreated biotite), T-2 (16 times cooked in *puta*), T-3 (20 times coked in *puta*), T-4 (25 times cooked in *puta*), T-5 (30 times worked in *puta*). Note: cow urine, paddy and jagerry were used as other ingredients.

CONCLUSIONS

Present study reveals that major elements such as magnesium, potassium and iron as well as trace elements such as manganese and zinc in the biotite structure are released into the filtrates during the *Ayurvedic* treatments.

Paddy, jaggery and cow-urine act as purifying agent of mica mixture. On the other hand, they provide nutrients to the *Ayurvedic* product. Organic materials accumulate from the paddy, jaggery and cow-urine are destroyed during the heating processes. Therefore, the final product is enriched with nutrients accumulated from minerals and other ingredient used. Based on the results of the present study, it can be suggested that the final product of mica *basma* has been formed from both biotite structure and other ingredients used.

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