Synthesis and Characterization of Lauha Bhasma

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Received: 21.9.2024, Revised: 24.10.2024 Accepted: 24.10.2024

Abstract

The preparation of ayurvedic bhasma is particularly challenging, as mass production in modern facilities often lacks the standardized equipment needed to ensure consistent quality, leading to concerns about toxic elements like mercury and arsenic in the final products. Addressing these concerns necessitates the standardization of the synthesis process through the implementation of stringent quality control measures at every stage of preparation. This study is dedicated to the synthesis and comprehensive characterization of lauha (iron) bhasma, employing both traditional methodologies and advanced analytical techniques. The synthesis process began with **samanya shodhana** (general purification), followed by **vishesh shodhana** (special purification) and **marana** (incineration) The resultant product then underwent **amrutikaran,** involving heating with aloe vera extract at an elevated temperature of 750°C.The synthesized lauha bhasma was meticulously characterized using a combination of classical Ayurvedic techniques and modern analytical methods, including Fourier Transform Infrared Spectroscopy (FTIR), Atomic Absorption Spectroscopy (AAS), X-ray Diffraction (XRD), Brunauer-Emmett-Teller (BET) surface analysis, Scanning Electron Microscopy (SEM), Particle Size Distribution (PSD), and Energy Dispersive X-ray (EDX) analysis. The final product, post-Amrutikaran, successfully passed all traditional Ayurvedic evaluations, affirming the proper formation of bhasma. XRD analysis revealed the formation of rhombohedral α-Fe_2O_3 with only trace amounts of free iron, indicating high purity. SEM and PSD analyses demonstrated the creation of nanosized particles ranging from 50 to 200 nm, complemented by a specific surface area of 12.55 m^2/g as determined by BET studies. Crucially, EDX analysis confirmed that the amrutikaran process effectively eliminated mercury from the bhasma, ensuring its safety and compliance with stringent quality standards.

Key words: lauha bhasma, Characterization, XRD, SEM, FTIR

The work was presented at J.ISAS meet at Pune on 30 July 2024

1. Introduction

The synthesis of bhasma, a revered Ayurvedic medicine, is an intricate process that masterfully combines time-honoured alchemical techniques with the use of herbal and mineral ingredients. This meticulous preparation is designed to yield a highly refined medicinal ash, celebrated for its potent therapeutic properties and superior bioavailability. A comprehensive review of the literature highlights an abundance of studies focused on the analysis of bhasma[1-5], however, there is a notable scarcity of research dedicated to its synthesis and characterization[6-9]. In our current work, we have successfully synthesized lauha (iron) bhasma using traditional methodologies[10,11], and have conducted a thorough characterization using both classical tests and modern analytical techniques. lauha bhasma holds a prominent place in ayurvedic medicine, being widely utilized for its efficacy in treating conditions such as anaemia, jaundice, pain, asthma, piles, skin disorders, and ailments of the spleen and liver.

Tiwari etal.[12] carried out characterization of lauha bhasma synthesized from tikshna lauha (clax of Fe turnings). They found particle size of the order of 100 nm and formation of FeO and Fe_2O_3 in the bhasma wherein $Fe^{2+}$and Fe^{3+} were in 40% : 60% as per Synchrotron based XPS studies. Bhargava etal.[13] presented studies on lauha bhasma prepared from tikshna ana kanta lauha. Their studies revealed formation of alpha and gamma Fe_2O_3along with oxides of some trace elements. Singh etal.[14] also reported physicochemical analysis of lauha bhasma synthesized from tikshna lauha which showed the particle size in the range of 100-500 nm. Krishnamurthy and colleagues[4] investigated lauha bhasmas produced by various manufacturers, focusing on their composition and structural properties. Similarly, Krishnamachary and his team[5] conducted studies on commercially available lauha bhasma, examining its morphology, structural, and chemical characteristics through contemporary analytical methods.

2.Experimental

2.1 Synthesis of lauha bhasma

Lauha bhasma was synthesized from iron sheet procured from local market. The various steps involved in synthesis are shown schematically in Fig.1.

Purification (samanya shodhana) of iron sheet

In this process, the iron sheet were heated till red hot condition and then quenched in different media viz. sesame oil, butter milk, cow's urine, kanji, decoction of horse gram. Changes in iron metal and media were observed during the process of heating and quenching of metal in various media. These observations are summarized in Table 1.

Special purification (vishesh shodhana) of iron sheet

In this process, the purified iron after the process of purification was heated again till red hot and then quenched in decoction of triphala kwatha.

The change in the weight of iron sheet during the purification (samanya and Vishesh shodhana) is graphically presented in Fig. 2.

Process of incineration (marana) for iron sheet

The incineration of Iron sheet was carried out in three stages using different media. For each stage, three different media used were aloe-vera, cow's urine and decoction of triphala.

The purified iron metal was first mixed with cinnabar ($1/12^{th}$ weight of iron metal). Cinnabar was purified according to ancient text by using juice of ginger. To this mixture extract of aloe-vera was added. The whole mixture was mixed thoroughly and was grinded in agate mortar-pestle. The mixture was grinded for 1h to ensure proper mixing. Small pellets of the mixture were prepared and dried in the shade. These pellets were weighed and then were placed in silica crucible for incineration in electric furnace at 750 ^{o}C for 3 h. After cooling, the pellets were grinded to fine powder in agate mortar-pestle. The procedure was repeated seven times. The iron powder thus obtained is treated with cow urine and then with triphala decoction in similar way. The final product obtained is of lauha bhasma.

The photograph of synthesized lauha bhasma is shown in the Photo 1.

3.Results and Discussion

3.1 Characterization of lauha bhasma according to the classical ancient text

The prepared lauha bhasma was evaluated based on classical analytical parameters, which reflect the physical as well as chemical characteristics of the lauha bhasma. The details of physical and chemical parameters are discussed in our previous communication[15].

The synthesized lauha bhasma was characterized by ancient classical analytical methods for its quality. The different tests and the corresponding observations are listed in the Table 2.

3.2Anaysis and characterization of lauha bhasma by modern analytical techniques

3.2.1Analysis of lauha bhasma by AAS

The elemental analysis of lauha bhasma for iron content was done by AAS technique during different processes of purification and incineration and is presented in Figs.3, the obtained results are between +/- 4%.

An examination of Fig.3 shows that during sesame oil process there is gradual increase in the weight of iron sheets during each stage. This may be due to coating of thin layers of sesame oil on the iron surface. The elemental concentration of iron at the end of the process was 93% and shows maximum concentration at 4^{th} stage and minimum concentration at the 6^{th} stage.

During butter milk process, there was increase in the weight of Iron sheets. The iron sheets were slightly brittle. The elemental concentration of iron at the end of the process was 88% and shows maximum concentration at 3^{rd} stage and minimum concentration at the 4^{th} stage.

The weight of Iron sheets was found to increase during cow urine process. The sheet starts to break in this process. The elemental concentration of iron at the end of the process was 96% and shows maximum concentration at 7^{th} stage and minimum concentration in the 2^{nd} stage.

In the fourth step, kanji was used in which the red-hot heated iron sheets were quenched for seven times. In this process, there is gradual change in the weight of Iron sheets

during each stage. This may be due to mustard oil (present in kanji) layer formed on iron sheet.

The elemental concentration of iron at the end of the process was 93% and shows maximum concentration at 4^{th} stage and minimum concentration at the 1^{st} stage.

In the last step of purification, decoction of horse gram was used in which the red-hot heated iron sheet was quenched seven times. In this process there was gradual decrease in weight of iron sheet. The concentration of iron at the end of the process was 94% and shows maximum in the 4^{st} stage and minimum on the 5^{th} stage. At the end of sammanya shodhana process the iron sheets became brittle and could be easily broken into pieces and the color of the sheet turned black.

In special purification process, decoction of triphala kwatha was used in which the red hot heated iron sheet was quenched seven times. In this process there was gradual decrease in weight of iron sheet. The concentration of iron at the end of the process was 94% showing maximum in the 4^{th} stage and minimum on the 3^{rd} stage. At the end of vishesh shodhana process, the iron sheets became more brittle and could be easily broken into smaller pieces and the color of the sheet turned black.

The purified iron metal was first mixed with cinnabar and then incineration was carried out with three different media: aloe vera, cow urine and decoction of triphala. Small pellets of mixture in each media were incinerated in an electric muffle furnace at 750 ^{0}C for 3 h. the process is carried out 7 times to get proper formation bhasma. The change in concentration of iron in each stage is shown in Fig 4. During the process it was observed that the metallic luster of iron particles was totally lost and was converted into fine powder. The concentration of iron was reduced drastically this may be due to conversion of inorganic iron into bhasma. The Final concentration of iron in lauha bhasma was 50%

3.2.2 Characterization of lauha bhasma by FTIR

The FT-IR spectra of the bhasma was recorded in the region 4000-400 cm^{-1} on Bruker Tensor -37 and is shown in the Fig.5. It shows sharp bands at 445 and 530 cm^{-1} which are assigned to Fe-O stretching and bending vibration mode[16]. The additional peak in the region of 1100 to 1000 cm^{-1} for the lauha bhasma in the figure could be due to the characteristic

frequencies of O-O bond arising from the absorbed oxygenates[17] . The presence of broad O-H stretching around 3400-3500 cm^{-1} suggests some moisture or hydroxylated compounds, and weak signals in the 1600-1700 cm^{-1} and 1400-1500 cm^{-1} regions might indicate traces of organic compounds or impurities.

3.2.3 Characterization of lauha bhasma by XRD

The X-ray diffraction study of lauha bhasma was done on D8 Advance Bruker AXS X-ray diffractometer. The scanning angle was from 20-80^o and the rate of scanning was 1^o/min. The XRD pattern of synthesized bhasma is shown in the Fig. 6.

The XRD pattern of prepared lauha bhasma matches well with the reported standard rhombohedral α-Fe_2O_3 (hematite) (JCPDS card no. 86-0550). The observed diffraction peaks, d spacing values and corresponding hkl planes are listed in Table 3. The high intensity of XRD lines in the XRD pattern suggests that the bhasma is present as a crystalline material; also, the broadening of the peaks suggests the nano size of the sample. The absence of a prominent peak at 45^o suggests that the amount of free iron is insignificant in the bhasma. The peak at (104) is characteristic of hematite (α-Fe_2O_3), which is a common phase in iron bhasma preparations. This confirms presence of iron oxide (Fe_2O_3) in the prepared lauha bhasma[18]. Peaks at (116), (214), and (300) further support the presence of hematite. The crystallite size of iron oxide in the lauha bhasma was calculated from XRD pattern following the Scherrer equation. The average crystallite size was found to be 50 nm.

The sharpness and intensity of the peaks indicate good crystallinity of the material. The presence of multiple peaks with low noise suggests a well-defined crystalline structure.

3.2.4 Characterization of lauha bhasma by TG and DTA

The TG/DTA analysis of lauha bhasma was carried out on DTG 60-H. The heating rate was kept as 10 ^{o}C/min over the range of 25-900 ^{o}C. The analysis was carried out under nitrogen flow of 50 ml/min. Aluminum cup was used as a sample holder.

The TGA curve (Fig.7) shows a slight decrease in weight as the temperature increases from 100°C to 900°C.The overall weight loss is minimal (6.6%), indicating that the sample is relatively stable over the temperature range studied. The weight loss may be due to removal of loosely bonded water in the sample. Two endothermic peaks were observed on the DTA curve: The endothermic peak at 120 °C corresponds to the loss of physically bound water

molecule. No exothermic peaks were observed in the region of 200 to 400^{o}C, hence can be stated that the bhasma formed is α-Fe_2O_3. As in this region phase transition from γ-Fe_2O_3 to α-Fe_2O_3 occurs, resulting in an exothermic peak.

3.2.5 Characterization of lauha bhasma by SEM and Particle Size Distribution

The SEM and EDS analysis of synthesized and commercial bhasmas was carried out on JEOL JSM-6360A Analytical electron microscope. The high resolution images of lauha bhasma at different magnification are displayed in the Fig. 8.

An examination of Fig. 8 reveals that The surface appears rough and irregular, with a large number of small, granular particles distributed throughout, the average particle size of the bhasma varies from micron to nano. This variation of particle size can be attributed to the different number of calcination cycles. It should be noted that most of the particles are in the range of 50-200 nm. However, few micron size particles are also observed. Particle size distribution analysis further supports these results.

Fig. 9 represents the particle size distribution analysis of lauha bhasma sample, which reveals most of the distribution of particles is in the range of 50-200 nm and residual part reflects at approximately 0.5μm range. Moreover, it can be also seen from Fig. 13 that few nano size particles are found to be clustered on the bigger (micron size) particles. All the particles are irregularly shaped. In general this process of bhasma formation is similar to the top-down approach for synthesis of nano- materials, in which successive cycles of calcination in the 'marana' process for the formation of bhasma resulting in nano size particles.

3.2.6 Characterization of lauha bhasma by Energy Dispersive X-Ray spectroscopy (EDX)

The elemental content of the final form of synthesized lauha bhasma was determined by using energy dispersive x-ray spectroscopic technique in order to check the presence of mercury in it. EDX spectrum showed presence of Hg to a significant level (1.41 %) in Fig.10a. Hence in order to remove mercury completely, the bhasma was subjected to amrutikaran. The EDX spectrum of bhasma after amrutikarna showed absence of Hg peak indicating complete removal of mercury from it. Thus, quality of bhasma was improved after amrutikaran process (Fig. 10b).

3.2.7 Characterization of lauha bhasma by BET

BET analysis was carried out on BET Thermo-scientific. BET pattern of synthesized lauha bhasma is shown in the Fig. 17. The Specific Surface area of the particles of the synthesized lauha bhasma was measured by N_2 desorption. From Fig. 11, the specific surface area of the synthesized bhasma was found to be 12.55m^2/g. This was in accordance with the fact that bigger particles were of lauha obtained due to the typical preparation method of the drug. This was also evidenced by the SEM results.

Conclusions:

Classical tests revealed proper formation of lauha bhasma as per the ancient text. XRD analysis revealed that synthsized lauha bhasma is in α-Fe_2O_3 phase form. SEM and PSD studies indicated the formation of nano-size particles in synthesized iron bhasma. Amrutikaran process was effective in complete removal of mercury.

Acknowledgement:

Authors are thankful to BCUD, SPPU, Pune for the research grant for this work

Figures:

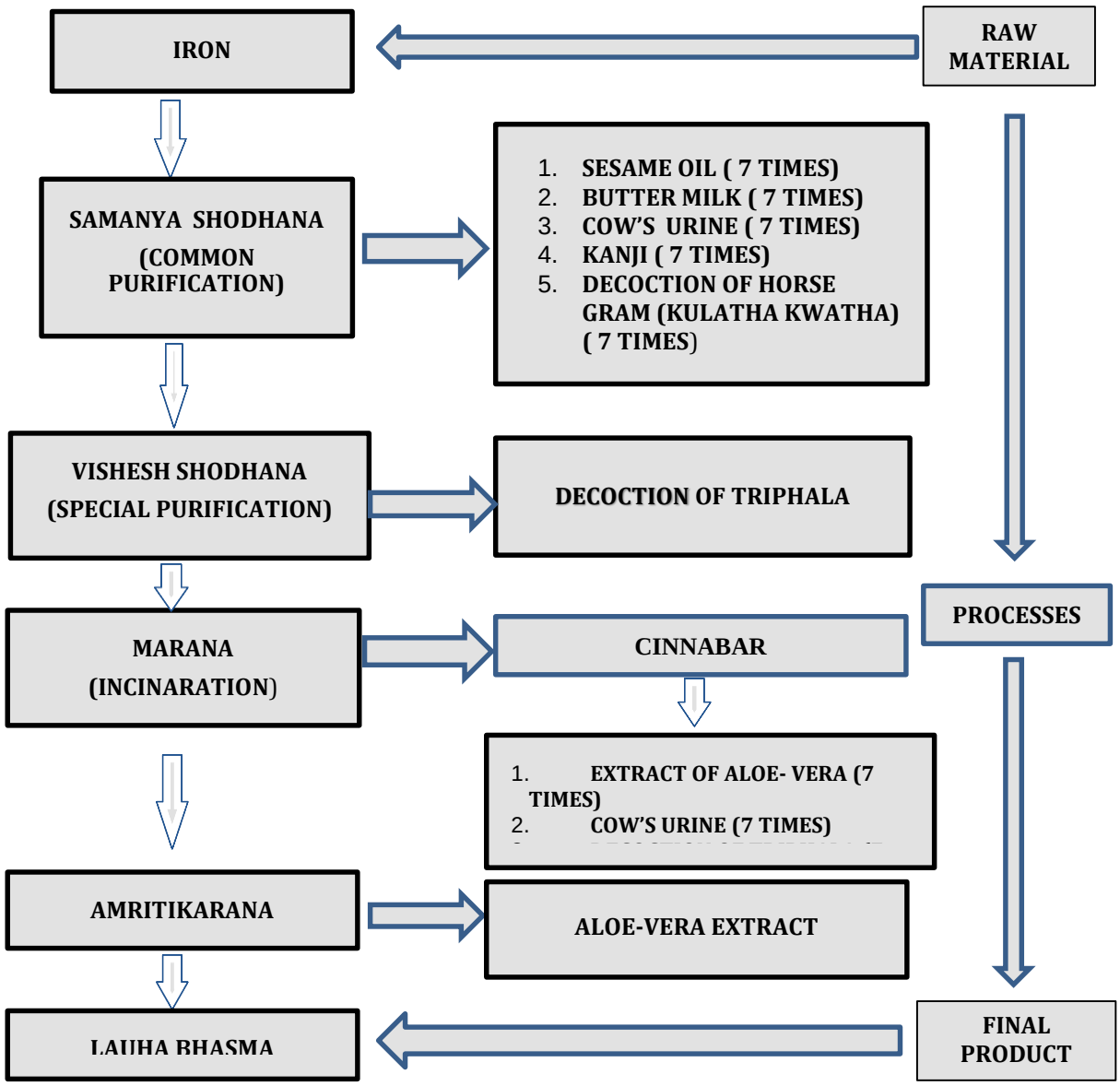


Fig. 1: Schematic diagram of synthesis of Lauha bhasma

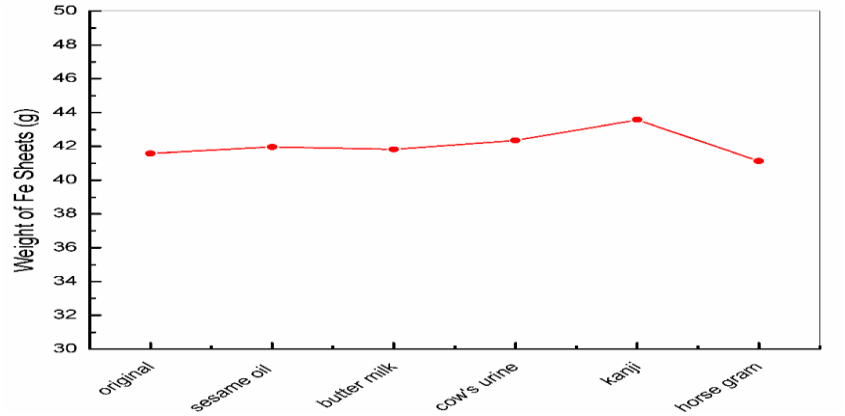


Fig.2 Variation of Weight in Iron during the Purification (samanya Shodhana and vishesh Shodhana)

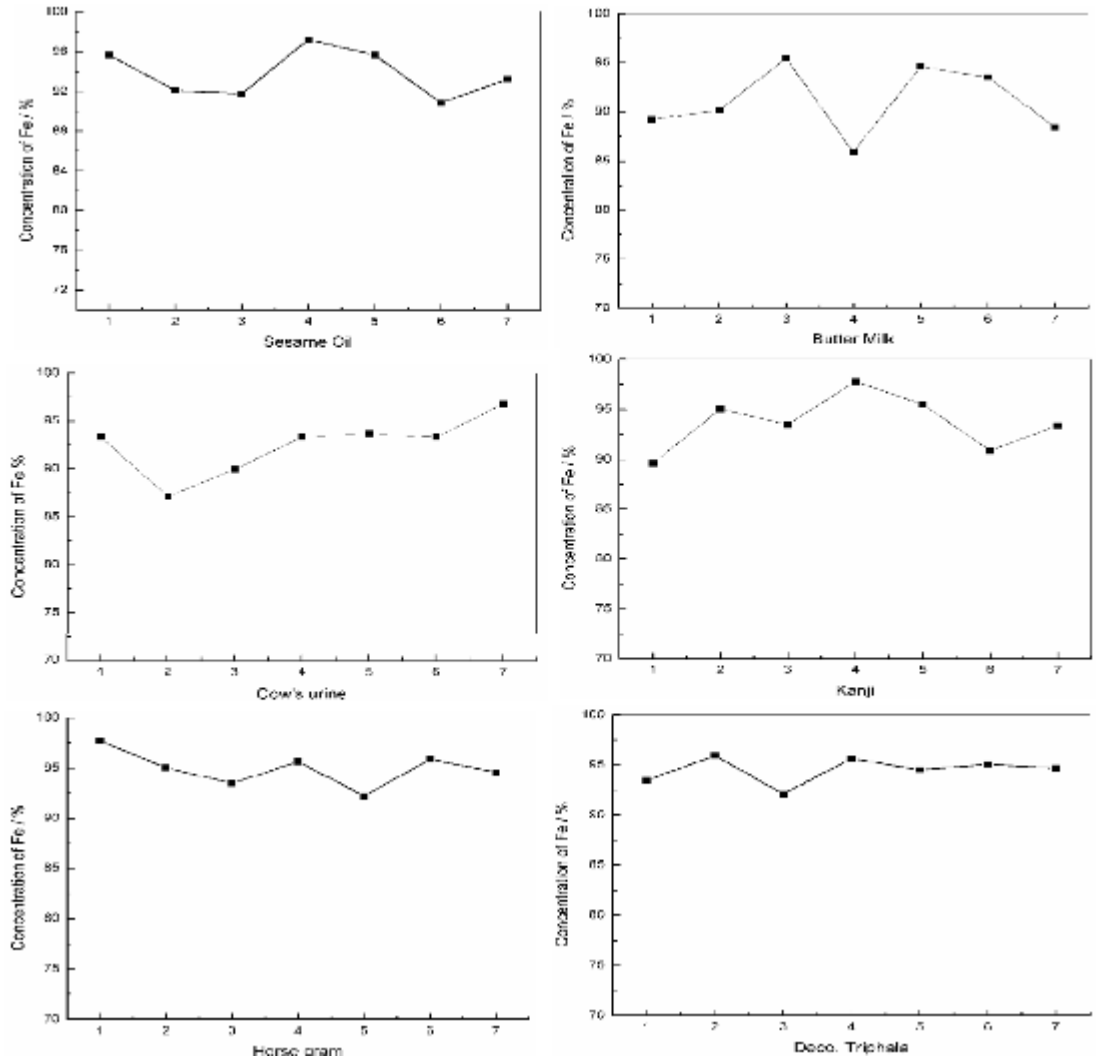


Fig.3 Variation of iron concentration during various processes

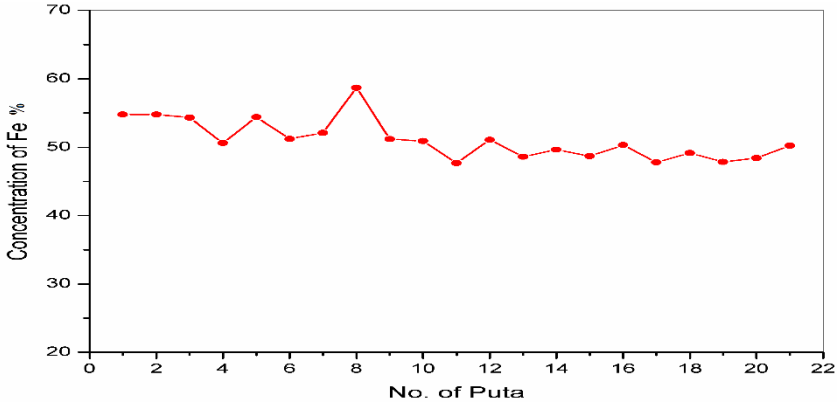


Fig. 4 Variation of iron concentration during incineration

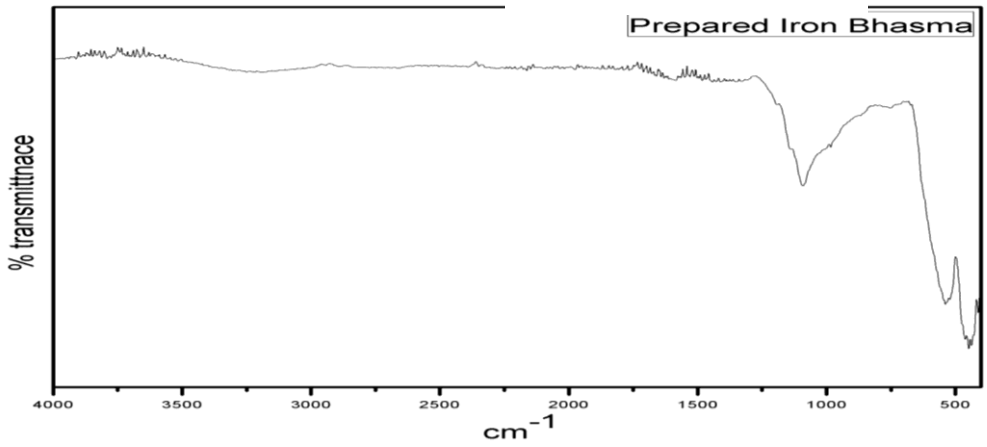


Fig.5 FTIR spectrum of synthesized Lauha bhasma

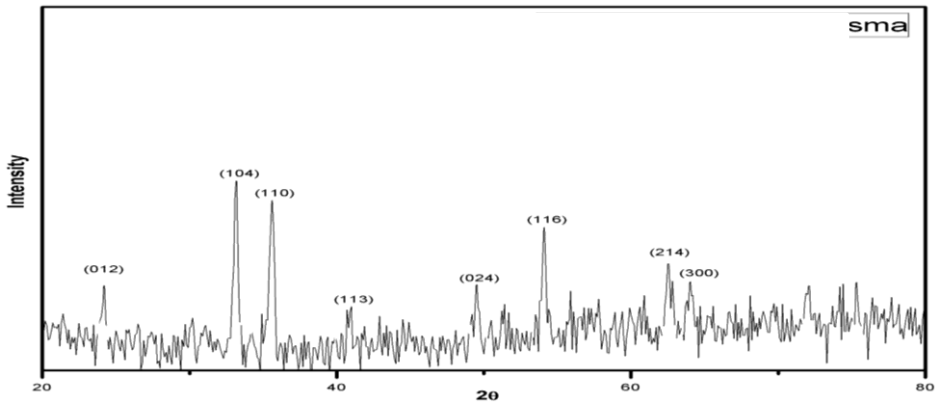


Fig. 6 XRD pattern of Synthesized Lauha bhasma

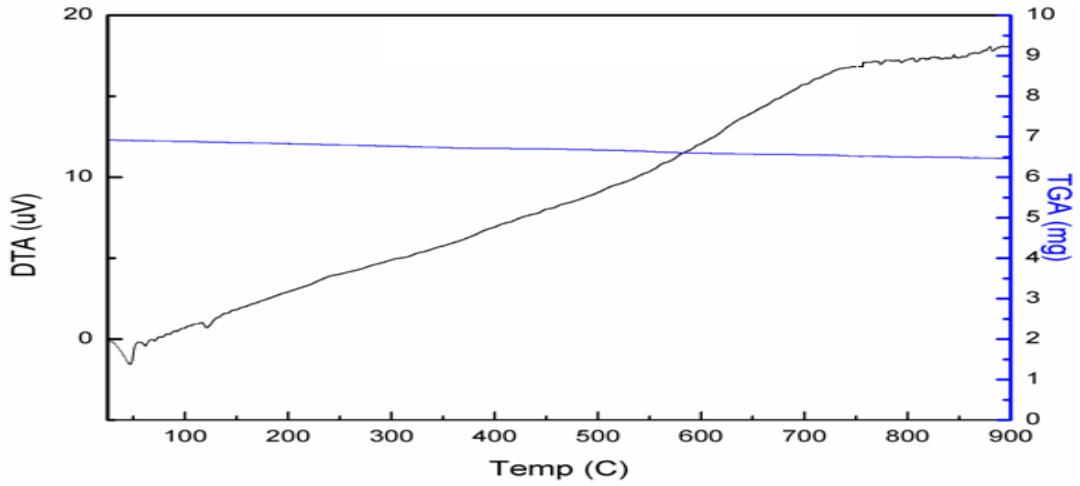


Fig. 7 TG/DTA pattern of Synthesized Lauha bhasma

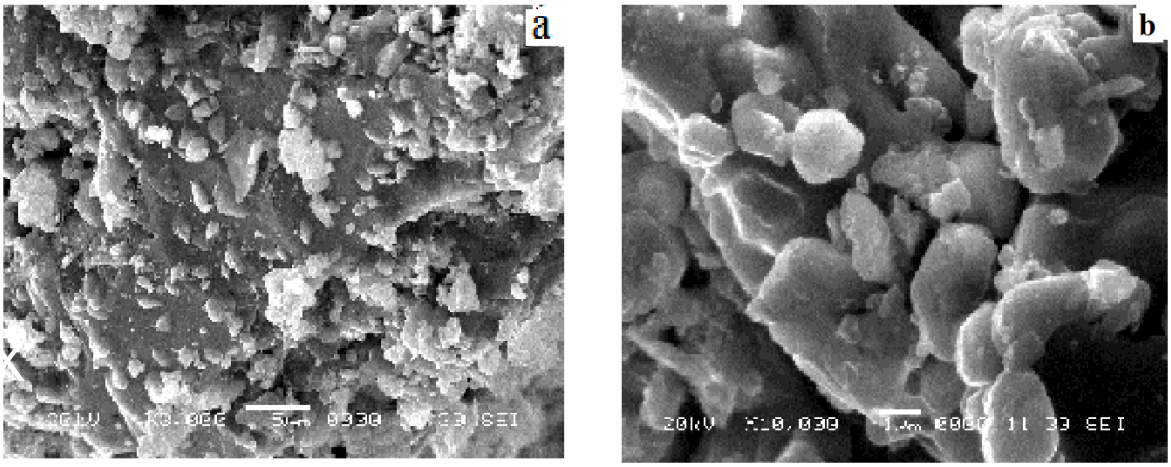


Fig.8 SEM of Synthesized Lauha bhasma a-20µm, and b- 1µm resolution

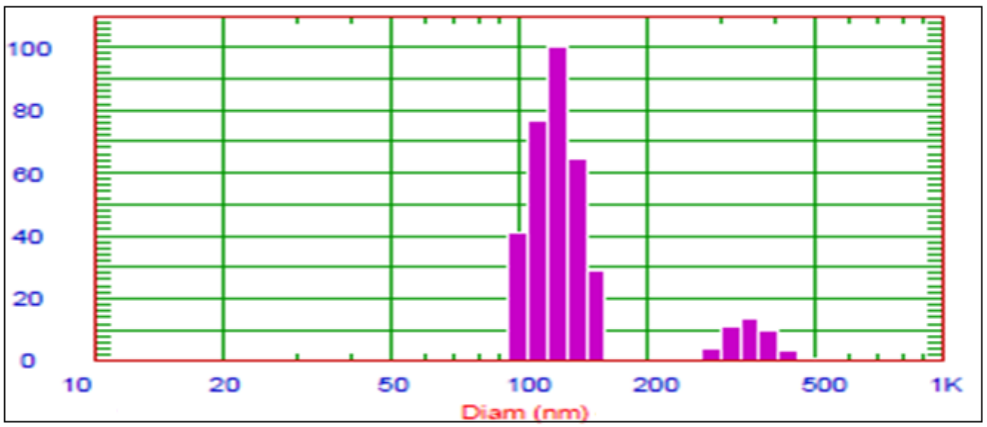


Fig. 9 Particle size distribution of Lauha bhasma

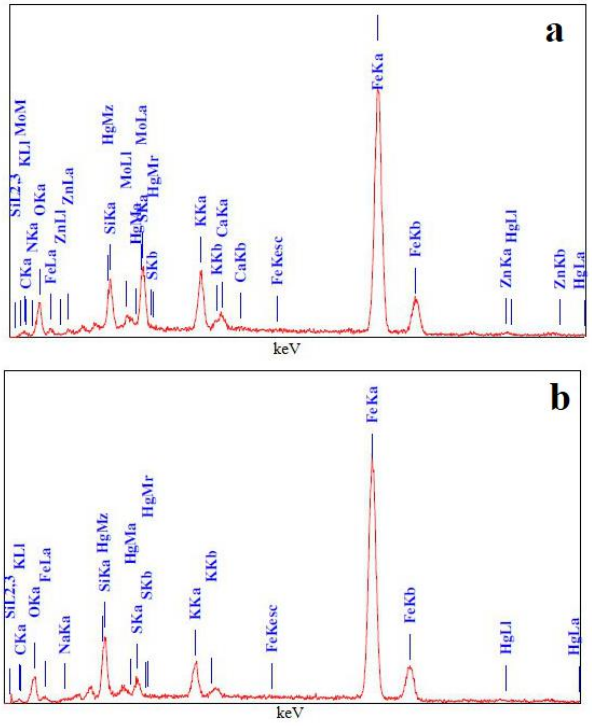


Fig.10 a) EDX spectra of synthesized Lauha Bhasma. b) EDX spectra of synthesized Lauha bhasma after amrutikaran

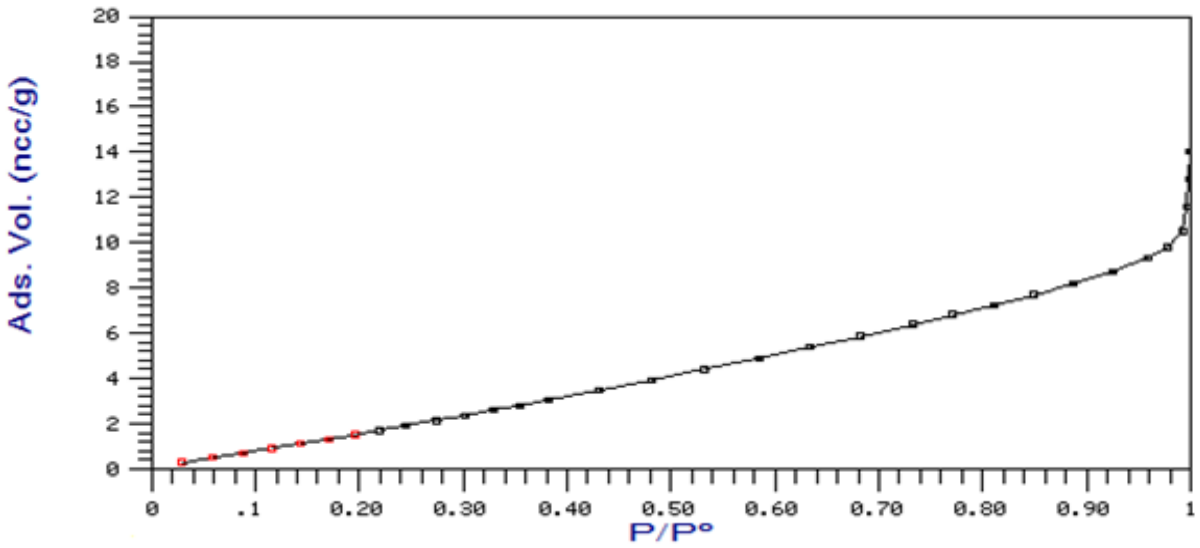
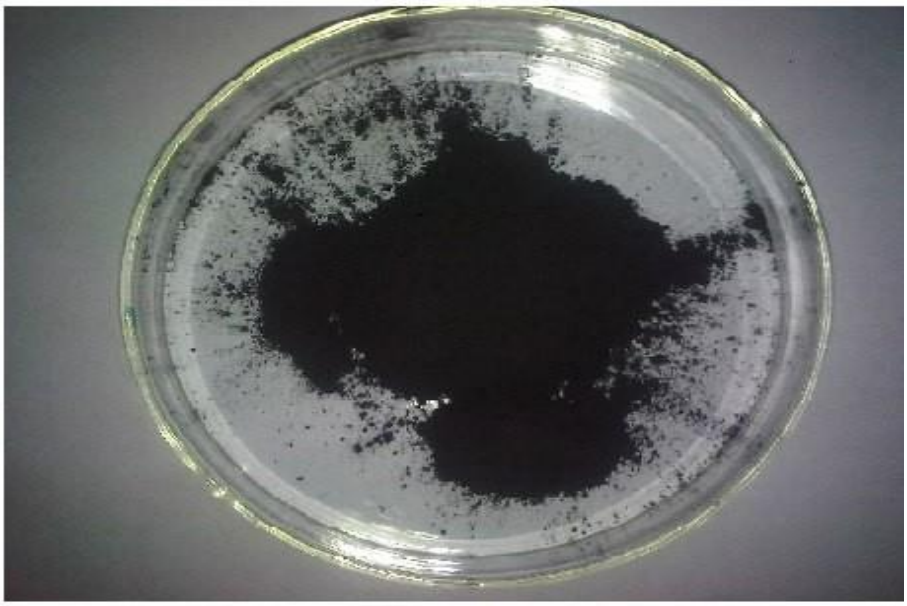
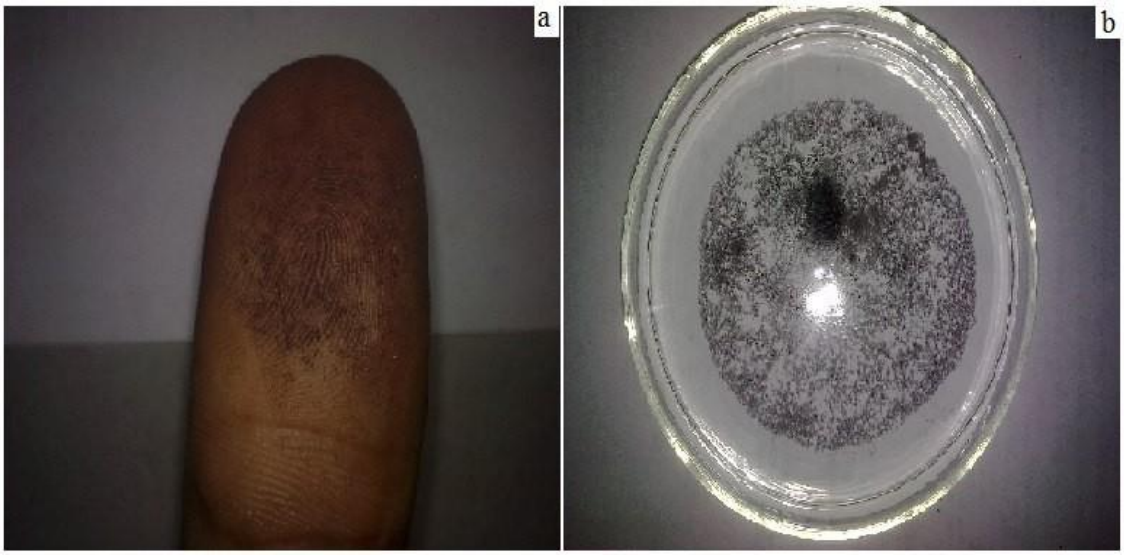


Fig.11 BET pattern of Synthesized Lauha bhasma

Photos:



Photograph 1 Synthesized Lauha bhasma



Photograph 2 a) Rekhapurnata Test, b) Varitara Test

Tables:

Table 1 Change in Metal and in media observed during purification process

Sr. No	Media	Change in Iron	Change in Media
1	Sesame oil	Brittleness is not altered.	Color turns darker. Becomes thicker.
2	Butter milk	Brittleness is slightly altered i.e. metal starts becoming soft.	Consistency is disturbed. No change in color.
3	Cow's urine	Starts breaking into pieces.	Typical smell emerges.
4	Kanji	Further reduction in size.	Typical smell emerges. No change in color.
5	Decoction of horse gram	Further reduction in size.	Slight discoloration.

Table 2 Observation and results for characterization of lauha bhasma according to classical ancient Text.

Sr. No	Test	Observation	Results
1	***Varitara***	Lauha bhasma floated on stagnant water.	**+ve**
2	***`Unam` Test***	The rice grain remained as it is on the layer of floated bhasma.	**+ve**
3	***Rekhapurnata***	The bhasma filled the minute furrows of the fingerTips	**+ve**
4	***Nirchandrata***	No lustered was observed in sunlight.	**+ve**
5	***Slakshnata***	The lauha bhasma was soft and smooth to touch.	**+ve**
6	***Niruttha***	Weight of the silver remained almost the same. No increase was observed.	**+ve**

Table 3 XRD data of synthesized iron bhasma

2Ɵ	d spacing/nm	(hkl) plane
24.2	3.672	(012)
33.2	2.694	(104)
35.6	2.518	(110)
41	2.1984	(113)
49.5	1.838	(024)
54.1	1.692	(116)
62.5	1.484	(214)
64	1.452	(300)

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