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Review

Overview of Sang-e-Surma (Antimony Sulphide or Lead Sulphide): A Mineral Origin Unani Drug

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Abstract

The ultra-fine powder of Sang-e-Surma (Surma Stone), also known as Kohl, is used for various eye ailments either alone or in combination with other herbal or mineral ingredients. The earliest use of Surma is reported in Egypt about 3100 BC. Data from a number of studies conducted on Sang-e-Surma using modern analytical techniques have cleared the uncertainty that Surma stone is mainly composed of lead sulphide. Surma is reported to be used for impaired eye-sight, ophthalmia, cataract, itching, redness, irritation, watering of eyes, shedding of eyelashes and in the initial stage of cataract. The use of lead based Surma is discouraged owing to reports of lead toxicity though the sun glare protection and antimicrobial effects of Surma have been established. Reports on lead toxicity by the application of Surma in eyes are conflicting. Appropriately planned studies are warranted to elaborate the toxic effects of lead based Surma/kohl in terms of detoxification of Sang-e-Surma, preclinical toxicity and clinical trial.

Keywords: Unani medicine; Sang-e-Surma; Ithmed; Kohl; Lead toxicity

Introduction and historical perspectives

The ultra-fine powder of Sang-e-Surma (Surma stone), used for various eye ailments either alone or in combination with other herbal or mineral ingredients, is known as Surma or kohl. The use of Surma for protection and treatment of various eye diseases has been given importance since antiquity. The earliest use of Surma is reported in Predynastic Egypt about 3100 BC. Kohl (Surma) played an important cosmetic, medicinal and social role in the lives of the Egyptian royalty. It was the indicator of social rank and achievement. Essentiality of

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Kohl for Egyptians can be assessed by its application before mummification of dead bodies. The first written document regarding use of Kohl was found in Egyptian *Ebers Papyrus* and the material used in preparation of Kohl was malachite and galena. Both of these materials were discovered in the graves in several conditions. The history of use of kohl then continued through “Christian period” (from phase of Egyptian culture to the end of 3rd century AD) until the uprising of “Islam” AD 641. During this period, soot (carbon black) was also used as a black pigment. The use of Surma (Kohl) before sleeping is described as “Sunnah” in Islam as it was recommended by Prophet Muhammad (Peace be upon Him) for enhanced vision and hair growth [1-5].

Controversy

There has always been a controversy regarding the chemical composition of Surma stone. There are two schools of thought, one says it is Antimony sulphide the other says it is Lead sulphide. Initially, the source of Kohl was believed to be a black stone namely “Ithmed”, composed of antimony trisulphide (stibnite ore /$Sb_2 S_3$). It is grey-black lustrous metalloid. This stone was used alone as Surma (Kohl) or used in combination with other ingredients such as camphor, menthol, almond seeds, wood essence, charcoal etc. Due to scarcity and high cost, the antimony trisulphide was gradually replaced by the lead sulphide (Galena ore) owing to its same grey black colour and shiny appearance like stibnite [6]. The others say that in Egyptian rule, galena was known by the name *mestem* or *stim*; the word stim is similar to the Greek word, stimmi or stibi, and to the Latin word *stibium*, which means antimony. Therefore, some authors have misinterpreted these words and reported the Surma (Kohl) as antimony, instead of lead sulphide. Finally, the controversy regarding the chemical composition of kohl was resolved when Professor V. X. Fischer analysed 30 samples of ancient Egyptian Kohls obtained from Fayum (Egypt) and concluded that lead sulphide was the chief constituent of Egyptian Kohls [5-8]. Further, the material found in Kohl boxes of Plain Cilicia, Turkey was examined chemically. It was concluded that the main ingredient of kohl was Galena (lead sulphide) and the antimony was not found in the said sample [9]. In Arabic Surma stone is known as Hajar al Kohl. Another Unani mineral drug namely Sang-e-Basri (zinc carbonate) is also known as *Hajar al Kohl* because of its stone like appearance and its use in eye ailments [10]. However, it is realized that both antimony sulphide and lead sulphide are still in use as Surma (Kohl). In Indian market galena (lead sulphide) is primarily sold as Surma stone. In present review chemical composition, toxicological and pharmacological studies of both stones have been compiled from the available sources.

Vernacular names

Arabic: Kohl, Hajar al Kohl, Ithmed (Ismad), *English*: Collyrium, *Hindi*: Anjan, Surme-ka-pathar, Kala Surma, *Persian*: Surma, Sang-e-Surma, *Sanskrit*: Srotanjana (antimony sulphide), Nilanjana (lead sulphide), Sauviranjana (stibnite), Anjan, Varnag; *Urdu*: Siyah Surma.

Description and Occurrence

Antimony sulphide (stibnite) is the main ore of antimony having silvery, grey glittery appearance that is very brittle and has a flaky texture. Whereas, the lead sulphide is the chief ore of lead having bluish, grey, or black mineral of metallic and glittery appearance (Figure 1). Lead sulphide is also known as galena. The occurrence of antimony trisulphide in the earth's crust ranges from 0.2 to 0.5 parts per million and is reported in Indian states of Andhra Pradesh, Bihar, Jammu & Kashmir, Karnataka and Uttar Pradesh. Worldwide antimony trisulphide is mainly found in China (82% of world production) followed by Tajikistan, Bolivia, Russia, South Africa, Turkey and Australia [11]. The *lead sulphide* (galena) deposits are found in Indian states of Rajasthan, Uttarakhand, Jharkhand, Jammu and Kashmir, Madhya Pradesh, Chhattisgarh, Karnataka, Orissa, Himachal Pradesh and West Bengal. It generally occurs in association with sphalerite (ZnS) [11,12]. Worldwide lead sulphide is found in different regions of Egypt (*viz. Aswan, mountains of Gebel-el-Zeit, Mountain Sinai (Kohe Tur) and Gebel Rasas (mountain of lead), near red sea*) and China, Australia, United States, Peru, Russia, Mexico and Iran [9,13]. According to Unani literature, it is found in Indian Himalayan region and Punjab and the best one found in Isfahan (Iran). It is lustrous, highly brittle, made of layers, free from extraneous matter and having crumps when broken. It is of various colours according to source of origin e.g., black, grey, reddish and white. White Surma is generally used as calx [14-21].

Figure 1. Image of Surma stone obtained from Saudi Arabia

Temperament

It is Cold and Dry with two degrees [10,14-28].

Action

Qabiz (Astringent), *Mujaffif* (Desiccant), *Muqawwi-e-Basar* (Vision improving), *Man-e-ufoonat* (Antiseptic), *Habis-ud-Dam* (Styptic), *Mubarrid* (Febrifuge) [10,14-28]

Therapeutic uses

Kohl is traditionally used to improve the personal appearance its powder is used as a cosmetic to the eye-lids and eyebrows especially by women.

Ulcers and wounds: It is useful for healing of ulcers. It removes excessive flesh from wounds and ulcers and heals them. When applied on a burnt part of the body with fresh fat, it does not ulcerate. Its application with wax and white lead heals the ulcerated part.

Organs of the head: It is useful to stop cerebral rhinorrhagia, especially when it emanates from the meninges.

Eyes: Its application on the eye is useful for impaired eyesight, ophthalmia, cataract, itching, redness, irritation, watering of eyes, shedding of eyelashes and in the initial stage of cataract. It removes the filth from eye ulcers and protects them. It improves in old age when applied after mixing with musk.

Other organs: When used as a pessary, it stops bleeding of the uterus. When sprinkled on fresh wounds of external male genital organs, it stops bleeding. When applied to the body along with suitable oil, it is reported to kill body lice [10,14-28].

Potent action

Dafe-Amraz-e-Chasm (prevents eye diseases) and *Habis-ud-Dam* (Styptic) [10,14-28].

Dosage

Kohl is not recommended for oral use due to toxic effects. The white variety of Surma is used orally in the form of Kushta (calx).

Adverse action

According to Unani scholars it may be harmful for lungs, joints and voice [10,5,16,20-28].

Correctives

To counter the toxicity of the Surma, Kateera (Tragacanth gum), Shakar (cane sugar) and Aab-e-Dhania (Coriander juice) are recommended [5,10,16,20-28].

Substitute

In case of non-availability of Surma, Sang-e-Basri/Burnt lead as a substitute may be used [5,10,16,20-28].

Detoxification/purification of Sang e Surma

To minimize the toxic effects of Sang-e-Surma, Unani Scholars have recommended for detoxification before its medicinal use. It can be detoxified by any one of following methods:

a) Cover the piece of Sang-e-Surma with the goat's fat and keep it on low fire till all the fat is completely burnt into fumes. Remove the piece of Sang-e-Surma from the fire with tongs and immerse in Arq-e-Gulab or ice water. Repeat the whole process three times.

b) Immerse the piece of Sang-e-Surma in Arq-e-Gulab or Arq-e-Badiyan and heat till the Arq is evaporated. Repeat the process seven times.

c) Immerse the Sang-e-Surma in Aab-e-Triphala and boil for 12 hours.

d) Immerse Sang-e-Surma in rain water (Aab-e-Baran) or distilled water for 21 days [29].

Important formulations

Some important classical Unani formulations in which Sang-e-Surma is one of the ingredients mentioned in National Formulary of Unani Medicine are Aksireen, Kohl-e-Jawahar [29], Kohl-e-Basaliqoon Kabir, Kohl-e-Chashkham, Kohl-e-Mameeran, Shiyaf-e-Aswad, Shiyaf-e-Gharb, Shiyaf-e-Aqleemiya Quroohi, Shiyaf-e-Nasoor [30], Barood-e-Aswad [31], Surma-e-Booraq, Surma-e-Nooran, Surma-e-Zahiri [32].

Elemental analysis of different Surma/kohl samples

Ali Al-Kaff *et. al.*, analysed various samples of kohl namely; stone kohl (black stone) and the in-powder forms: Ka-Noori, Kohl Noori (imported from India), brown kohl, and kohl Samah (made locally in Saudi Arabia) for their chemical composition by using electrothermal atomic absorption spectrophotometry. It was observed that Black stone contained 88% lead and 0% antimony, Ka-Noori 4.1% lead and 0.53% antimony, Kohal Noori 3% lead and 1.82% antimony, Brown powder had traces of lead and 1.13% antimony and Samah powder contained 20% lead and 9.97% antimony [33].

Osama *et. al*., collected 16 samples of Kohl Al-Ethmed from Saudi Arabia, Pakistan, India and Yemen to know their chemical composition through Atomic Absorption Spectrometer (AAS). It was found that most of the kohl available in the market contained harmful levels of lead ranging from 12.34-78.32%. Only one sample having reddish black colour matched the description quoted in the literature. Further this sample contains 0.01% (1 mg Pb/1 g of sample) of lead along with 27.79% iron, 0.03% Zinc, and 0.01% antimony. The author concluded that it may be Kohl Al-Ethmed that Prophet Mohammed (peace be upon Him) recommended to use [34].

Pervaiz Habib Ullah *et. al.* analysed the kohl stone sample brought from Madina (Saudi Arabia) through X-Ray Diffractometer to know its chemical composition. It was found that the main constituent was Lead (85.51%) followed by Sulphur (11.43%) and antimony (2.06%). While Carbon was detected in small quantity (0.689%) [35].

Pasha *et. al*., analysed two samples of Kohl stone one procured from Saudi Arabia and other from India to know the chemical composition through X-ray and concluded that both stones were galena (lead sulphide) [36].

Al-Ashban *et. al*., reported that 107 kohl samples were collected from different parts of Saudi Arabia, and analysed for the presence of lead, aluminium and antimony. Lead levels up to 53% were detected in some kohl preparations, and some samples were found to contain camphor and menthol. The blood analyses of regular kohl users revealed a high Pb concentration and relatively low haemoglobin levels [37].

In another study, 23 kohl samples were analyzed using X-ray powder diffraction (XRPD) and scanning electron microscopy (SEM) out of which 19 samples were obtained in Abu Dhabi and four in Dubai. None of the samples were produced in the United Arab Emirates. The major component of 11 samples was found to be galena (PbS). For the remaining 12 samples, the main component was found to be either amorphous carbon, zincite (ZnO), sassolite (H_3BO_3) or calcite/aragonite ($CaCO_3$) [38].

Daar *et. al*., conducted X-ray fluorescence analysis of Pb, Fe and Zn concentrations in 135 samples of bottled dry powder form kohl from nine randomly selected suppliers (15 samples of each brand) which revealed that Pb was found at elevated levels in products claimed as Pb-free [39].

Filella *et. al*., analysed 23 samples of Kohl cosmetics purchased in Europe by portable X-ray fluorescence (XRF) spectrometry and scanning electron microscopy-energy-dispersive X-ray spectroscopy. Pb was detected in 17 kohls at concentrations ranging from a few mg kg^{-1} to

> 400 g kg^{-1} and Cd was also present as a trace contaminant in 13 products. Authors concluded lack of quality control in manufacture and an illegal trade in Europe [40].

Quality standards of galena for medicinal use

The Government of India has laid down standards of various mineral drugs to check adulteration, quality standards for galena (lead sulphide) are as follows [41]:

Physical properties: It should be in the form of lump and heavy cubic crystals, grey in colour, evenly fractured, brittle, opaque with metallic lustre. Specific gravity 7-8 and hardness range should be 2 to 3.

Chemical properties: Galena fuses easily, emits sulphurous fumes when heated on charcoal through a blow pipe. On continued heating it yields a globule of metallic lead. Galena should contain not less than 50% Lead (Pb) when analysed by AAS. It should contain not less than 10% Sulphur by analysing with gravimetric method. Silver (Ag) should not be less than 500 ppm when analysed by AAS. Arsenic should not be more than 2 ppm and Cadmium not more than 22 ppm. Galena may contain Copper = 70 ppm, Gold = 0.10 ppm and Zinc = 20 ppm within ± 20% of the stated limits.

Distinction from Antimony sulphide: Cubic cleavage, greater specific gravity and darker colour of lead sulphide distinguish it from antimony sulphide (Sb_2S_3) [41].

Toxicological profile of antimony and lead in human

Antimony: Antimony is obtained from mines in the form of antimony trisulphide. This is also known as Sang-e-Surma. Antimony can enter the human body through oral, respiratory and dermal routes. Long term exposure to antimony in the air at levels of 9 milligram per cubic meter (mg/m3) may cause irritation of the eyes, skin, and lungs. It can also cause pneumoconiosis, cardiac problems, stomach pain, diarrhoea, vomiting, and stomach ulcers. The association of antimony toxicity with use of Surma is not established [42].

Lead: Lead mostly occurs as lead sulphide. Lead is a non-essential element to the human body and found in at least 45 human tissues including vital organs. Approximately 90% of the total body load of lead is found in the skeleton. Absorption of lead to the human body takes place through three routes i.e., oral, inhalation and cutaneous route. Absorption through oral route is poor (5-15%). However, absorption of chloride, oxide and acetate of lead is more than carbonate and sulphate compounds of the lead. Thus, we may presume that lead sulphide is least absorbed through the oral route. The individual having some nutritional deficiencies like calcium and Vitamin D deficiency have higher absorption rate through oral route. Lead present as particulate matter during air pollution is likely to be absorbed through the respiratory tract. Large particles of the lead are cleared by ciliated epithelium of the upper respiratory tract. Whereas, fine particles may reach the lower respiratory tract. Ninety percent of the absorbed lead is cleared by the macrophages. Absorption of lead through skin is insignificant. Eighty to ninety of absorbed lead is excreted through faeces, urine, sweat, milk hair and nails [43].

Toxicity of lead: The World Health Organization (WHO) permissible limit of lead for medicinal products is 10 ppm [41]. WHO has suggested the upper limit for blood lead for adults is 10 µg/dL and for children it is 5 µg/dL. In adults symptoms of lead poisoning may be seen if blood lead level is greater than 40 µg/dL, whereas children with level greater than 45 µg/dL requires treatment of poisoning. [44]

The central and peripheral nervous system, cardiovascular, gastrointestinal, renal, endocrine, immune and haematological systems are affected due to lead toxicity. In children, lead can cause intellectual disability because the child's brain is at a developing stage. Elevated blood lead levels in children may show behaviour problems, low IQ, poor grades at school, difficulties with hearing, short- and long-term learning difficulties and growth delays. Exposure to lead during pregnancy may cause spontaneous abortion, reduced fetal growth, premature birth, blood pressure elevation, and cognitive deficiencies in the child [45].

Can Surma application to eyes cause lead poisoning?

The main ingredient of Surma (collyrium) available in markets is primarily galena (lead sulphide). The regular use of Surma may cause lead poisoning. In this regard, it is pertinent to mention that the scientists have different opinions. One group of researchers is of the view that application of lead-based kohl is safe and it does not cause lead poisoning. The other group says that application of lead-based kohl may cause lead poisoning.

Is the application of lead-based kohl safe?

In 1968, Warley *et. al*., published a report relating to lead poisoning from eye cosmetic (Surma) and concluded that lead sulphide was responsible for lead poisoning. Later, Srivastava and Varadi countered that report by concluding that lead sulphide (Kohl) was not responsible for poisoning; however, the product was colourful lipstick made up of lead carbonate [46,47]. In another study published by Ali *et. al*., it was reported that blood lead concentration of the Asian children who applied Surma in their eyes was higher than those who were not applied Surma. The authors concluded that the use of Surma may be associated with high blood lead concentration. However, another Scientist rejected the hypothesis by questioning about the socioeconomic status of the test and control group, the number of children in the test group were more than twice that in the control group [9]. Another study was conducted by Khalid *et al.* on sixty-two human volunteers (23 children and 39 adults). Their ages ranged between 12 to 55 years. The blood lead level recorded at day zero was treated as a control. After application of Surma once a day (0.5 to 1.0 mg each time with an applicator) the blood lead levels were measured at day 30, 60 and at day 90. The results showed no significant difference between the control and test [48]. When Kohl (Surma) is applied on the eye, the lead is not absorbed through the corneal route. However, the possibility of some of the material being washed into the naso-lachrymal duct. The main route for ingestion of lead in children is oral, via the fingers. Only 0.2% of this applied lead is transferred to the mouth via fingers. The sole application of Surma cannot be the source of lead intoxication. The absorption of lead in the body is dependent on a variety of factors including diet, state of health and solubility of ingested lead compounds. Susceptibility to lead toxicity may be influenced by various physiological and environmental factors (e.g., age, season of the year, body temperature, and dehydration, ultraviolet light), nutritional factors (including calcium, phosphorous, iron, vitamin D, protein, ascorbic acid, nicotinic acid intake), alcohol, as well as some other heavy metals. In the United States, the major risk factors for lead toxicity in children are deficiencies of essential metals, calcium, iron, and zinc, and housing and socio-economic status [9]. The bioavailability of lead sulphide is least in comparison to other lead compounds such as lead acetate and lead oxide as evident from a study conducted by Dieter *et. al.* in animal model [49]. Summarizing all these data and reports, it can be derived that, lead from kohl is not absorbed through trans-corneal route as well as by oral ingestion which is certainly minimal in adults (women and men). The higher blood lead level in individuals after the application of Kohl, may be linked with the socio-economic factors and nutritional deficiencies and not possibly from the use of Kohl (Surma). The author of one review on Surma and lead toxicity concluded that, lead toxicity due to application of Surma is more of theoretical nature than a practical health hazard [9].

Is the application of lead-based kohl hazardous?

Many researchers believe that application of lead based Surma may cause lead toxicity in humans particularly in children. In a review authored by Tiffany-Castiglioni *et. al*. regarding the role of Kohl/Surma in lead toxicity, the author summarized 26 studied published between 1968 to 2012 to support his conclusion that Pb-based kohl is associated with increased PbB (blood lead) in women and children who used kohl. It is worth mentioning some important studies associated with increased PbB level in kohl users. In a study conducted at Saudi Arabia in 1996, the author concluded that kohl was the major contributor to elevated PbB in children. In a case study of an 11-month-old child of the US published in 1999, the author mentioned that the initial PbB of the child was 43 µg/dL. The child had the history of Surma application for 5 months and no other obvious source of Pb exposure was identified. The child's PbB was 23 µg/dL after 8 weeks of Surma discontinuation. It was concluded that Surma was the major source of lead toxicity. The 22-year-old Moroccan woman presented with abdominal pain, behavioural evidence of encephalopathy, and other signs of Pb poisoning. Her initial PbB was 490 µg/dL, and after 6 months of chelation therapy reduced to 49 µg/dL. It was concluded that the source of Pb exposure was daily application of Moroccan kohl on her conjunctivae for many years. In a case-controlled study conducted in Saudi Arabia, PbB was significantly higher in kohl users in comparison to the non-users. The author of the review concluded that application of lead-based Kohl may be the one reason of the lead toxicity [50]. In a study conducted by Goswami (2013), the researcher collected 34 samples of Surma/kajal available in the market. The samples were analysed for the presence of lead content and found that the majority of the samples contained high levels of lead. Further, a trial was conducted on 93 children out of them 69 were Surma users and 24 were non users. The result showed that blood lead concentrations of 36 children among Surma users was 38 μg/100 mL or above and their hemoglobin levels were low. None of them had evidence of lead toxicity. Low levels of hemoglobin may be due to high lead concentration in blood. However, the author could not mention socioeconomic and nutritional status of the affected children [51].

Other reported activities of Surma

Surma application on eye protect from sun glare: In one study the author reported that lead sulphide thin films had higher absorption and lower transmittance in the UV light band. Thus, it may be concluded that application of lead sulphide (Galena) in the form of Surma in the eyes has a natural protective effect against glare of the sun. The photo resistant, humidity and temperature sensor and solar properties of galena (lead sulphide) have also been reported [9].

Antimicrobial activity of Kohl: The facial cosmetics available in the market of Nigeria namely "*Tiro*, *Otanjele* and *Buje*" contained up to 81% of lead. This facial cosmetic showed considerable antibacterial activity against *Staphylococcus aureus*, *S. epidermidis, Streptococcus pyogenes* and *Pseudomonas aeruginosa* [52]. The antimicrobial potentials of different Kohl samples collected from the market of Karachi, Pakistan were tested against some clinically important pathogens, responsible for causing ocular infections. Seventy-five percent (75%) of the total Kohl samples exhibited good antimicrobial activity against *Proteus mirabilis, Staphylococcus epidermidis, Candida* and *Mucor* Species [52].

Conclusion

Few Unani authors have mentioned that Ithmed/Kohl is antimony sulphide in their text. However, on the basis of elemental analysis and geographical distribution Surma stone, it may be concluded that Sang-e-Surma is lead sulphide. Due to high content of lead, long-term use of lead-based Surma is supposed to cause lead toxicity. However, the mechanism of lead absorption from application of lead based Surma in eyes is still unclear. It is still doubtful whether the sang-e-Surma was detoxified or not before its use. Sun glare protection and antimicrobial effects of Surma have been identified by researchers. Pharmacopoeial standards of Sang-e-Surma have also been developed. There are conflicting reports regarding toxicity of lead based Surma. It is suggested that planned studies/trials are required to elaborate the toxic effects of Surma in human like preclinical toxicity and efficacy studies on animal model. Further, well designed clinical studies on Kohl are warranted to ascertain the toxicity potential of lead via topical application of Kohl formulations and to exclude the role of confounding factors (environmental and nutritional factors).

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