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"STUDY REMOVAL OF FLUORIDE FROM WATER BY USING LOW COST KOTA STONE SLURRY ADSORBENT"

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ABSTRACT

Fluoride related health hazards are a major environmental problem in many regions of the world. In India, 17 states have been identified as epidemic for fluorosis and Rajasthan is one of them. Due to fluoride pollutions and health problems that it causes, the World Health Organization (WHO) has specified the tolerance limit of fluoride content of drinking water as 1.5 mg/L .It has been observed that with the intake of fluoride above the permissible limit causes skeletal abnormalities and dental fluorosis. Fluoride produces cancer or birth defect. Long term ingestion of excessive fluoride has a chronic effect on kidney .Previous investigations show that the fluoride content of water can be reduced to the desired limit by precipitation and adsorption. Adsorption involves the contact of the fluoride containing water with an appropriate adsorbent. Objective of this study was to investigate or check efficiency of low cost adsorbent (Kota stone slurry) for the removal of excess fluoride from aqueous solution. Kota stone slurry is also major environmental problem. So we can use Kota stone slurry for removal of fluoride that is positive objective. The influence of various operational parameters viz. effect of adsorbent dose, pH, initial concentration and contact time were studied by a series of batch adsorption experiments from these studies, it may be concluded that Kota stone slurry is an efficient and economical adsorbent for fluoride removal from aqueous solutions.

Keyword: Adsorbent,Adsorption, batch adsorption,deflouridation fluoride,Kota stone slurry

I. INTRODUCTION:

Fluoride exists in the form of fluorides in various minerals such as Sellaite (MgF_2), Fluorspar (CaF_2),Cryolite (Na_3AlF_6) and Fluorapatite ($Ca_5(PO_4)_3F$). Thus fluorides are also found in rocks, soil, plants, animals, humans and fresh as well as ocean water [3,4]. Therefore fluoride occurs naturally in public water systems as a result of runoff from weathering of fluoride-containing rocks and soils and leaching from soil into ground water [3–6]. In addition to water, fluoride is present naturally in almost all foods and beverages but levels vary widely.

However, fluoride has both beneficial and harmful effects on human health depending on its level. Among the beneficial effects of fluoride in the human body, strengthening of bones and prevention from tooth decay are significant [6]. Compared to its beneficial effect fluoride is more detrimental. Thus fluoride is a toxic chemical and it is a risk factor for thyroid hormone production in children when the exposure to fluoride occurs during intrauterine growth period

[7]. A report during 2008 in Scientific American on ‘second thoughts about fluoride was a warning to all concerned as it revealed the risk of fluoride causing disorders affecting the teeth, bone, brain and thyroid gland [7,8]. It has also been reported that thyroxine and triiodothyronine in serum decreased with increasing urinary fluoride in cattle. Cattle affected with fluorosis developed hypothyroidism and anemia [7]. In addition, it has been confirmed that there is significant positive relationship between fluoride intake by water and the prevalence of dental fluorosis [3,9–12]. For the general population the intake of fluoride is mainly from drinking water and to a much lesser extent from foodstuffs i.e. drinking water is the major source of daily intake of fluoride [3,13,14].Due to fluoride pollutions and health problems that it causes, the World Health Organization(WHO) has specified the tolerance limit of fluoride content of drinking water as 1.5 mg/L [15]. Various defluoridation technologies available to remove fluoride from water, such as Precipitation Coprecipitation, Nalgonda technique, Adsorption, Electrocoagulation, Electrosorption, Reverse Osmosis, Nanofiltration, Electrodialysis and Donnan dialysis [16], but the adsorption process is generally accepted as the cheapest and most effective method for removal fluoride from water. The commonly used defluoride adsorbents include alumina, bone char, zeolite, gypsum material, kaolinite, cement clay, montmorillonite and rare earth metal [17-21]. However, the adsorbents have been restricted due to their unfeasibility, inoperability, expensive price, unsafely or not widely adaptability. fluoride by adsorption onto low-cost materials like limestone,kaolinite, bentonite, charfines, lignits and nirmali (strychnos potatorium) seeds extract, bone char, serpentine and sulfonated saw-dust carbon are of limited scope[22-30].Therefore, a low cost adsorbent with high efficiency, practically, economical and safety was developed for defluoridation in our study. In this regard, the present work elaborates the use of new type of adsorptive lime stone slurry waste, having more adsorptive efficiency for fluoride removal comparable to other costly adsorbents.

Solid waste lime stone slurry containing high amount of calcium and magnesium content, is produced in huge quantity from Kota stone industries. For this purpose, abundant availability of both solid wastes inspired us to develop low cost, effective adsorbent material for removal of fluoride from drinking water, which is mainly ground water in large portion of rural areas in Rajasthan. Batch adsorption studies are carried out systematically in terms of process parameters such as initial concentration of fluoride ion. Adsorption data are applied on Langmuir and Freundlich adsorption isotherms. Various kinetic models are tested to find out the suitable one to represent fluoride removal kinetics over stone slurry. This study presents the findings of an investigation on the use of waste kota stone slurry for the defluoridation of water by conducting batch studies and continuous down-flow column studies.

1.1 FLUORID GENERATION IN DRINKING WATER

The main occurrence of fluorine in rocks is in the form of fluoride bearing minerals.Usually the surface water does not contain high fluoride whereas groundwater may be contaminated with high fluoride content because the usual source of fluoride is fluoride rich rocks. When water percolates through rocks it leaches out the fluoride from these rocks. Fluorspar-CaF_2 (Sedimentary rocks like lime stones, sandstones), Cryolite – Na_3AlF_6 (Igneous rocks like Granite) and Fluoro-apatite – $Ca_3(PO_4)Ca(FCl)_2$ are the main rocks which are rich in fluoride.Fluorides come next to sulphur dioxide (SO_2) in the hierarchy of atmospheric gaseous pollutants. Fluorides are released in the air as gaseous hydrogen fluoride (HF) and volatile fluorides like cryolites and silicon fluoride (SiF4). These pollutants result from aluminum factories, brick kiln, pottery industries, Ferro-enamel works, stacks of factories processing-ore and oil.Fluoride contamination occurs through a natural process in which fluoride bearing rocks crumble and breakdown but the process can be speeded up if the chemistry of the aquifer is disturbed[31-35].

II. GENERATION OF KOTA STONE SLURRY WASTE

In general there are two types of waste named as quarry /cutting /sawing from in-situ stone site and polishing waste from construction sites. During the processing of stone, the raw stone block is cut as demanded either into tiles or slabs of various thickness (usually 2 or 4 cm), using diamond blades. Water is showered on blades while stone blocks are cut into sheets of varying thickness, to cool the blades and absorb the dust produced during the cutting operation. The amount of wastewater from this operation is very large. It is not recycled as the water is so highly alkaline that, if re-used, it can dim the slabs to be polished. In large factories, where the blocks are cut into slabs, the cooling water is stored in pits until the suspended particles settle (sedimentation tanks), then the slurry is collected in trucks and disposed off on the ground and left to dry. This water carries large amounts of stone powder. The polishing operation is fully automated with the use of powdered abrasives that keep on scrubbing the surface of the stone until it becomes smooth and shiny. Water showers are essential to prevent overheating of the blades.

III. ENVIRONMENTAL HAZARD-

3.1Fluoride hazardous –

The high concentration of fluoride in drinking water leads to destruction of enamel of teeth and causes a number of conditions referred to collectively as fluorosis. This disease is slow and progressively crippling malady. At low concentration (<1.0ppm), fluoride prevents tooth decay, but it has been medically proved that, high fluoride intake by individuals from water, food, air and medicines results in fluorosis [36].The excessive intake of fluoride may cause dental [34] and skeletal disorders [35]. Fluoride ion is attracted by positively charged calcium ion in teeth and bones due to its strong electro negativity which results in dental, skeletal and no skeletal forms of fluorosis i.e. high fluoride ingestion, in children as well as adults. Fluorosis in mild version can be evidenced by mottling of teeth and in high version by embrittlement of bones and neurological damage [36], in some of the cases it may even interfere with carbohydrates, proteins, vitamins and mineral metabolism and to DNA creation as well if intake excessively [37]. Studies have shown that major of the Kidney diseases have a great inclination of toxicity of fluoride. On high doses and short term exposure fluoride can exterminate the kidney function. Several research groups have also shows that fluoride can interfere with the function of pineal gland as well as of brain. Pineal gland is one of the major fluoride accrued site in body with concentration more than that of teeth and bones. Workers exposed to high fluoride concentration areas are diagnosed with bladder cancer [38]. Various diseases such as osteoporosis, arthritis, brittle bones, cancer, infertility, brain damage, Alzheimer syndrome, and thyroid disorder can attack human body on excessive intake of fluoride [39].

Harmful Effects—

- fluorosis,
- skeleton fluorosis,
- cardiovascular effect,
- gastro intestinal disorder,
- endocrine effects,
- neurological effects,
- reproductive effects,
- developmental effects,
- enzyme inhabitation,
- genetic damage,
- effect on the pineal gland

3.2 Kota stone slurry hazardous-Stone slurry is a semi-liquid substance consisting of particles originated from the sawing and polishing processes and water used to cool and lubricate the sawing and polishing machines. The main problems caused by this waste are –

1. When dried, the fine particles (size less than 363 micron) become air borne and cause severe pollution.
2. Apart from occupational health problems, it also affects machinery and instruments installed in industrial areas. Slurry dumped areas cannot support any vegetation and remain degraded.
3. When dumped on land, it adversely affects the productivity of land due to decreased porosity, water absorption, water percolation etc.
4. Excavation and disposal of such large quantity of waste in Kota stone industry cost about 25% - 35% of the total cost of production.
5. Besides, the disposal of such large quantity of solid waste causes serious environmental problems of degradation of land, loss of green pasture and loss of regional aesthetic values.
6. The heap of slurry remain scattered all round the industrial estate is an eye sore and spoil aesthetics of the entire region. Subsequently tourism and industrial potential of the state is adversely affected.
7. Waste from quarry and fabrication operations can be unsafe and environmentally detrimental.
8. During the rainy season, the slurry is carried away to rivers, drains, roads and water bodies affecting quality of water, reducing storage capacities and damaging aquatic life. The waste water from polishing operation

during rainfall is in a considerable large quantity and highly alkaline in nature. Due to unplanned and mismanagement in disposal of mining waste, the mining owner bought the rich fertile land of area near mining site and started disposing waste on the fertile land, leading it to turn barren. It has been noted (Leikin *et. al.*, 2009) that People working on mining sites and residents close to mining sites have reported many respiratory diseases like Silicosis, asthma, bronchitis.

IV. PROPERTIES OF STONE WASTE

Table 1: Physical and mechanical properties of powder and slurry stone waste

Lime stone slurry waste	Specific gravity	Bulk density	Water adsorption%	Maximum Particle size(mm)	Color
	2.58-2.68	1568-1745	2-4	0.2 dry	White / dirty white

Table-2Chemical composition of Kota stone slurry waste

Calcium Oxide (CaO)	38-42%
Silica (SiO_2)	22-24%
Aluminum Trioxide(Al_2O_3) + Ferrous Oxide (Fe_2O_3)	3-4% + 1.02-1.53%
Potassium Oxide K_2O+ Sodium Oxide Na_2O	0.35-0.62%
Magnesium Oxide (MgO)	1.0-1.5%
Loss on ignition (LOI) (%)	32-34%

The Kota stone slurry is found to contain Calcium, Potassium and Magnesium as macronutrients and iron, copper manganese zinc etc. as micronutrients. Some ultra- micronutrients are also found in the waste - Titanium,Vanadium,copper, Zinc,Manganese,rubidium and strontium [41]

V. ADSORPTION PROPERTIES OF KOTA STONE SLURRY WASTE-

Calcium has a good affinity for fluoride anion and it has been used for fluoride removal [40]. Crushed limestone (99% pure calcite) used as an adsorbent for fluoride removal by batch studies and surface-sensitive techniques from solutions with high fluoride concentration ranging from 3 to ~2100 mg/L. With different techniques, such as atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS) as well as ζ potential measurements, the authors were able to confirm that a combination of surface adsorption and precipitation reactions removed fluoride from aqueous systems. The removal capacity of fluoride was dependent on calcite surface area. Activated and ordinary quick lime as adsorbents used for fluoride removal from water [41]. When initial concentration was 50 mg/L, the removal of fluoride was 80.6% at optimum conditions from synthetic solution. Langmuir maximum sorption capacity of activated quick lime for fluoride was found 16.67 mg/g. The removal of fluoride was found due to chemisorptions and precipitation which was confirmed through scanning electron microscopy (SEM) micrographs and X-ray diffraction (XRD).

Aluminum hydroxide impregnated limestone as a adsorbent used for fluoride removal from water [42]. At pH 2, the adsorption in case of modified lime stone was decreased. The maximum sorption capacities of the limestone and aluminum hydroxide impregnated limestone were found 43.10 mg/g and 84.03 mg/g, respectively. The adsorption of fluoride depends upon the porous structure and high surface area of the modified granules. At pH range 3 to 4, maximum fluoride adsorption was found and further decreased as pH increased above 10 and the decreased in defluoridation was found due to the change in surface charge of the adsorbent. At pH 3, maximum defluoridation capacity was found 12.6 mg/g.

The removal of fluoride from aqueous solution with magnesia (MgO) and magnesia/chitosan (MgOC) composite were used batch equilibrium experiments [44]. It was observed that defluoridation capacity of MgOC composite (4440 mg/F^-/kg) was appreciably higher than MgO (2175 mg/F^-/kg). The adsorption capacity for fluoride was found 21.4 mg/g. Magnesia-amended silicon dioxide granules for fluoride removal were investigated [43].

Iron-based materials have been investigated for fluoride removal from water. Polypyrrole (PPy)/Fe3O4 magnetic nano composites is novel adsorbent in fluoride removal [33]. Fluoride removal by amorphous Fe/Al mixed hydroxides was evaluated [35]. At pH 7.5, mixed Fe/Al samples were prepared by the increase of Al content in Fe(OH)3 matrix increase the surface area. The fluoride adsorption followed first order kinetics and intra particle diffusion model. The sorption process followed both Langmuir and Freundlich isotherm models. The thermodynamic studies showed fluoride sorption to be spontaneous and exothermic in nature. Adsorption and desorption studies were also conducted to gain an insight into the adsorption mechanism on Fe/Al hydroxide surface.

Both granular ceramics FeSO4.7H2O and granular ceramic (Fe2O3) adsorbents was used for defluoridation of aqueous solution [38]. It was found granular ceramics FeSO4.7H2O is more effective than granular ceramic (Fe2O3) for fluoride removal. The adsorption experiments by batch and mini column scale to test the potential of granular ferric hydroxide for removal of various ions including fluoride was studied [39]. The fluoride adsorption onto granular ferric hydroxide was again found pH dependent. The fluoride capacity decreased with increasing value of pH. The fluoride has the highest adsorption capacity (1.8 m mol/g).

Activated alumina has been an effective adsorbent for defluoridation of water. Different studies have been focus on this. For the fluoride removal from water Acidic alumina [45], amorphous Al(OH)3, gibbsite or alumina (Al2O3) [46] have been used. It was found that this adsorbent react fluoride at pH range 3–8 with fluoride concentration 1.9 -19 mg/L. At pH 5.5-6.5, maximum fluoride uptake was observed 9 mol/kg. At lower pH, fluoride uptake decreased due to the preferential formation of AlFx soluble species but at higher pH, OH^- displaced F^- from the solid Al(OH)3 so the amount of fluoride adsorbed to complexes declined towards zero between pH 6–8. At lower rate, same reaction was followed with gibbsite. At pH 5–7, maximum fluoride removal was found 16.3 mg/g [47].

Table 3. Adsorption capacities (AC) and other parameters for the removal of fluoride by Calcium-based adsorbents magnesium oxide,aluminum oxide and ferrous oxide[33-44]-

s.no.	Adsorbent	AC (mg/g)	CR (mg/L)	pH
1	Quick Lime	16.67	10-50	-
2	Lime stone & Al(OH)3 impregnated lime stone	43.10 and 84.03	0-100	8
3	PPy/Fe3O4 nano composite	17.6 − 22.3	5-100	6.5
4	Fe/Al hydroxide	91.7	10-90	4.0
5	Magnesia-amended silicon dioxide	12.6	-	3.0
6	Magnesia,Magnesia/chitosan composite	2.2, 4.4	10-23	10

VI. CONCLUSIONS

From above study it is clear that Kota stone slurry have best chemical composition. Each chemical compounds show good adsorption of fluoride. So we can use Kota stone slurry as a cheapest adsorbent.

Study of fluoride adsorption on Kota stone slurry will be helpful in utilization of local waste and also to get rid of problem of excess fluoride in ground water in hadoti region.

This study will be future helpful to pollution control bord to develop fluoride removal model.

Use of Kota stone slurry is like “To kill two birds with one stone” and “Diamond cuts diamond”.

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