

Removal of Fluoride from Groundwater of District NUH (Haryana) using Dolomite as a Sorbent

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Abstract: A study aimed at removing fluoride from natural water through adsorption technique was carried out using dolomite [CaMg(CO₃)₂] as a sorbent. Batch experiments were conducted using dolomite in raw form and in calcined form to enhance the activity of calcium and magnesium for the adsorption of fluoride ions present in groundwater. Experiments were conducted using different dosages of dolomite for different concentrations of fluoride. It was observed that at 850°C, the activity of dolomite was high to form complex with fluoride, whereas adsorption through raw dolomite was insignificant. No significant change in TDS in the water samples was observed, while a rise in pH was observed when analyzing calcined dolomite. This could be a cost-effective method to remove fluoride from water for consumption.

Keywords: fluoride, fluorosis, dolomite, calcination, defluoridation, water

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I. Introduction

Water is an elixir for life. A large part of India depends on groundwater for domestic water supply. But fluoride contamination in groundwater is a serious challenge. The concentration of fluoride in groundwater highly depends on geological characteristics and the chemical composition of rocks. Some fluoride-bearing minerals are fluorite, fluoro apatite, biotite, apatite, muscovite, hornblende, cryolite, topaz, and many more. Groundwater contamination by fluoride is one of the major problems in semiarid and arid regions of the world (Adimalla, et al., 2019). Due to the high evaporation rate and less precipitation, there is a risk of fluoride enrichment in these affected areas (Adimalla, N., 2018; Ayoob and Gupta, 2006; Subha Rao, et al., 2015).

Groundwater mining is another major reason for an increase in the spread of fluoride contamination. When groundwater mining is more than the recharge, depletion is caused. This may further change groundwater flow direction, which in turn causes ingress of seawater or the intrusion of other saline ground/surface water or polluted water from surrounding areas, affecting the quality of groundwater itself (Sharma, L. M., 2014).

World Health Organization (WHO) has marked the upper limit of fluoride in drinking water not more than 1.5 mg/L but The Bureau of Indian Standards (BIS) has laid the limit to 1mg/L stating the fact that “Lesser is better” (Andezhath and Ghosh, 2000, WHO., 2004).

Fluorosis is a condition resulting from the ingestion of large amounts of fluoride. Excessive systemic exposure to fluorides can lead to disturbances of bone homeostasis (skeletal fluorosis) and enamel development (dental/enamel fluorosis). The severity of dental fluorosis is also dependent upon the fluoride dose and the timing and duration of fluoride exposure. Dental and skeletal fluorosis are known health hazards.

Fluorosis is worldwide health problem that occurs due to the excessive intake of fluoride in drinking water. It is an endemic disease that affects teeth (dental fluorosis) and bones (skeletal fluorosis). It also manifests like metabolic, structural, and functional damage in organs as the nervous system (Valdez-Jimenez, et al., 2011), endocrine glands (García-Montalvo, et al., 2009), reproductive system (Hao, et al., 2010), hypertension (Sun, et al., 2013), kidney and liver (Chattopadhyay, et al., 2011).

In India, a large number of people suffer from fluorosis due to intake of fluoride in drinking water. Per the National Health Program, high levels of fluoride were reported in 230 districts of twenty states of India. The population at risk per population in habitations with high fluoride is 11.7 million as of 1.4.2014. Rajasthan, Gujarat, and Andhra Pradesh are the worst-affected states. Punjab, Haryana, Madhya Pradesh, and Maharashtra are moderately affected states, while Tamil Nadu, West Bengal, Uttar Pradesh, Bihar, and Assam are mildly affected states (<https://hi.nhp.gov.in/disease/non-communicable-disease/fluorosis>).

Fluoride is one of the most widespread pollutants in groundwater. Global interest lies in cost-effective and sustainable technology for the removal of fluoride. Ayoob, et al., 2008, reported that coagulation and adsorption are the most widely used methods for the defluoridation in fluoride-endemic areas. The most commonly used method in India is through activated alumina. But this method is associated with many risks and challenges.

This paper focuses on the approach of adsorption mechanism of fluoride uptake using heated dolomite. In India, dolomite is found in sedimentary and metamorphic rocks in the form of dolomitic marble. It is used in construction aggregate, cement manufacturing, agricultural soil treatments, metallurgical flux, a source of magnesia in chemical industry, oil and gas reservoirs, and many more. On a scale of Mohs hardness, dolomite falls in the position of 4 on 10. In India, Rajasthan is the major source of dolomite. It is a very cheap material and easily available in abundance. Dolomite is a common rock-forming mineral. Its composition consists of calcium magnesium carbonate ($\text{CaMg}(\text{CO}_3)_2$). Calcination of dolomite at a high temperature helps in converting the salt to oxides of magnesium and calcium with the release of carbon dioxide as shown in the following chemical reaction (Staszczuk, et al., 1997).

II. Materials and Methodology

2.1 Materials

For this study, the waste of dolomite mines converted into powder was procured from the local market of Gurugram (Haryana). Considering the minimum cost of processing, the material was used as it was, without any further refinement process.

Samples of natural water containing fluoride ranging from 2 ppm to 12 ppm were used for analysis. These groundwater samples were collected from the villages of Nuh district of Haryana.

2.2 Reagents and instrument

For the determination of fluoride concentration, it was tested using selective ion electrode method (Orion, Star A214 by Thermo Scientific) by adding TISAB III (Orion ion plus by Thermo Scientific). The calibration was carried out until the slope of curve -54 to -60 mV was achieved; pH (by pH meter-80 by HM Digital) and total dissolved solid (TDS) (E-1 portable TDS meter by HM Digital) were measured for the same.

2.3 Experimental procedure

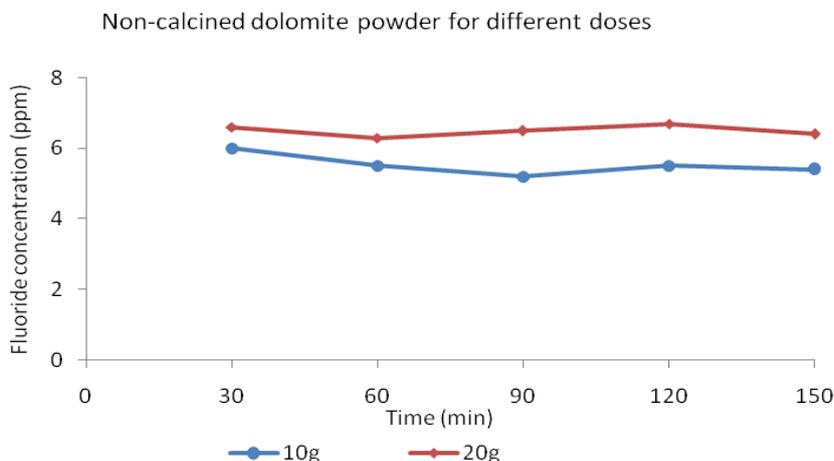
The dolomite powder procured was as it is available in the local markets for the construction industry. Heat treatment of dolomite (by muffle furnace) at different temperatures (650°C and 850°C) for different durations (two and three hours) was carried out to explore the potential of calcined dolomite as a sorbent. All experiments were conducted in triplicate. Variations among the results of these triplicates were not significant; the average value was adopted as the final result.

Batch experiments were conducted to investigate the effects of different doses of dolomite (1 mg, 3 mg, and 5 mg in 100 mL of sample water) for fluoride removal, pH, and TDS. These experiments were performed in triplicate. Natural groundwater with different concentrations of fluoride used was in the range from 2 ppm to 12 ppm. Under static conditions, the solution was tested mostly at every hour for fluoride concentration; pH, and TDS were also measured for the same.

III. Results and Discussion

3.1 Removal of fluoride using raw dolomite

Dolomite powder in its raw form was initially tested to remove fluoride from natural groundwater. Initial concentration of sample was 6.9 ppm. The graph below shows the removal activity of dolomite in its inactive form. There is no significant change in the fluoride concentration when tried with two different doses for 2.5 hours.

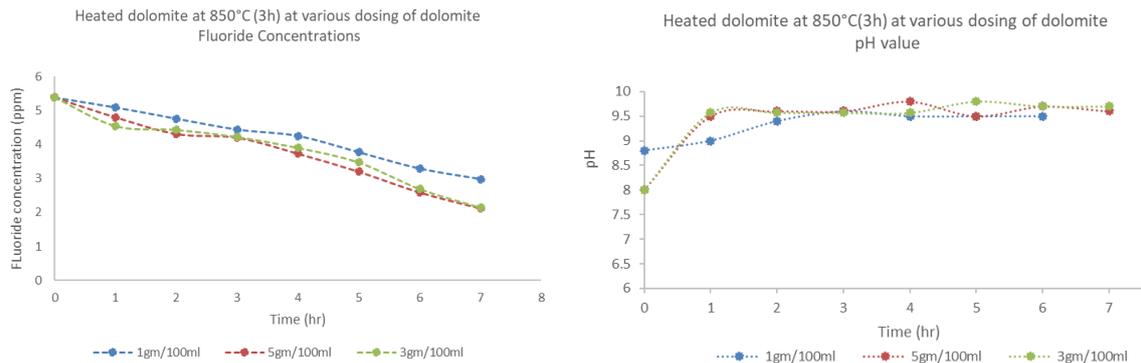


On the basis of the results obtained and also encouraged with some research study publications, the dolomite was heated at different temperatures in order to enhance their activity to form complex with fluoride.

3.2 Working with calcined dolomite

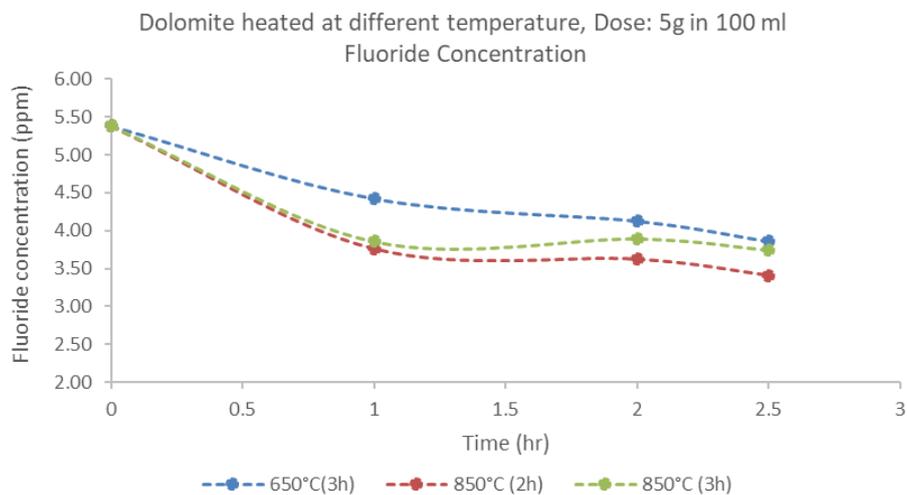
3.2.1 Different doses of dolomite

Dolomite powder (activated at 850°C for threehours) was used for trying out the different dosing (1gm, 3gm, and 5gm) for fluoride removal. The initial concentration was taken to be 5.38 ppm with pH as 8 and experiment was conducted for sevenhours. The figure below shows the overlapping of two doses, i.e. 3g and 5g, which also represents that 3g and 5g show ultimately the same efficiency in removing fluoride at the same time. For further experiments, 3g was taken as the optimal dose.



3.2.2 Heat treatment of dolomite at different temperatures

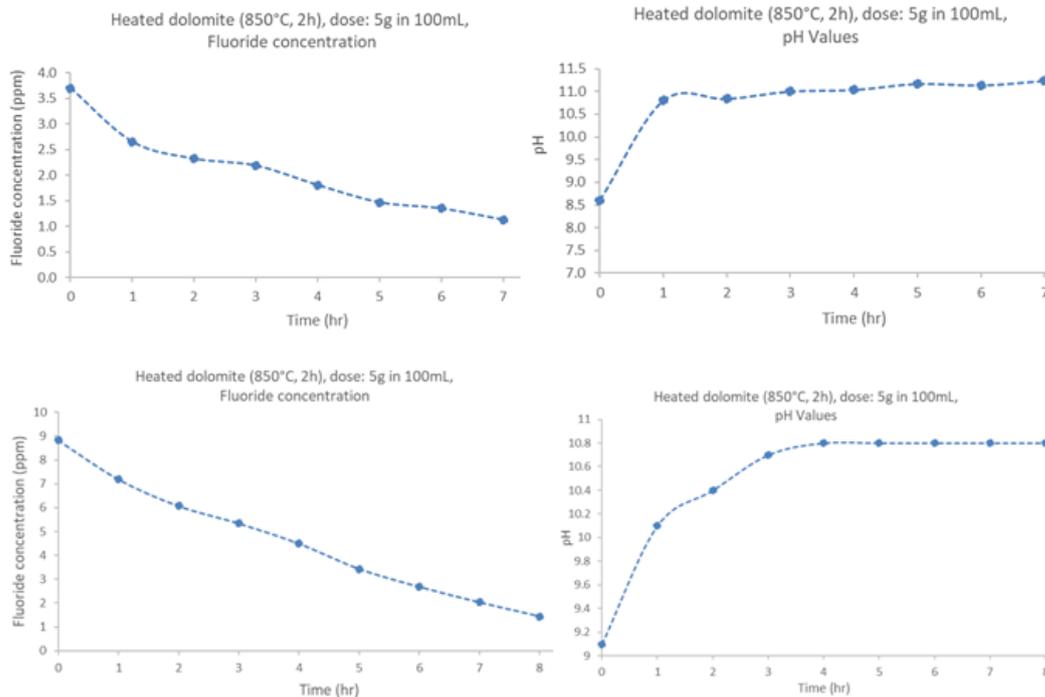
Dolomite was heated at temperatures, i.e. 650°C (threehours) and 850°C (twohours and threehours). The graph below shows the trend of fluoride removal when tested with dolomite activated at different temperatures. The dose of dolomite was taken to be 5g in 100mL for each sample. The initial concentration of fluoride was 5.38 ppm.



It was observed that dolomite calcined at 850°C shows higher fluoride removal. Various studies also show that, at 650°C, only magnesium ions get active; whereas at 850°C, calcium also gets active. By the above graph, we can conclude that fluoride removal is best when activated at 850°C (2h), as both calcium and magnesium are active and would make complexes with fluoride result in better removal.

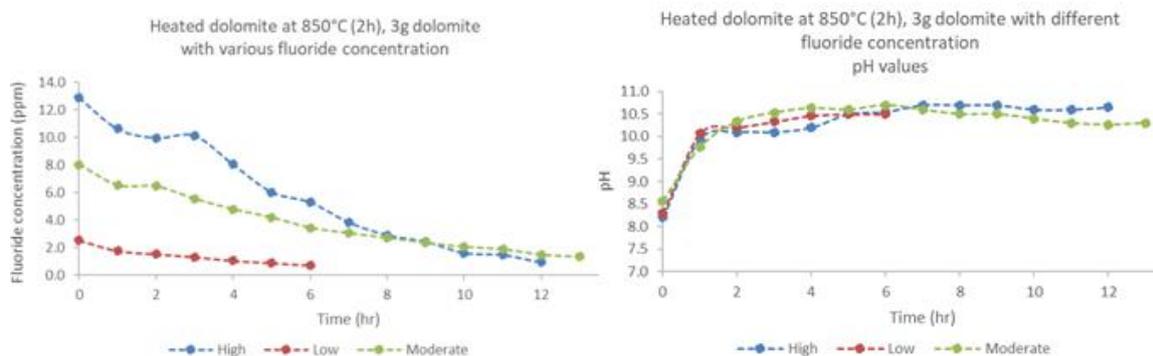
3.2.3 Removal of fluoride from low and high concentration

Dolomite powder activated at 850°C for twohours was taken in amount of 5g in a 100mL water sample. The initial concentration was taken as 3.7 ppm and the final concentration was 1.1 ppm in sevenhours. For a high concentration sample, the initial concentration of 8.83 ppm was taken, and the final concentration reached out to be 1.44 ppm. The below graphs depict the decrease in fluoride with low concentration and in high concentration.



3.2.4 Removal of fluoride for various concentrations

Activated dolomite powder at 850°C for two hours in the amount of 3g in 100mL of water sample was performed with the initial concentrations varying from low, moderate, to high. The graph below depicts the decreasing trend of fluoride removal. For low concentration, after seven hours the removal efficiency achieved was 72 percent. For moderate concentration, fluoride was removed up to 82 percent in thirteen hours. For higher concentration of fluoride, fluoride removal was 93 percent in twelve hours.



IV. Conclusion

Study Confirms that:

- adsorption was dependent on contact time, adsorbent dose, and concentration of fluoride.
- dolomite activated above 800°C showed better results for fluoride removal.
- a rise in pH is observed during the process.
- dolomite is a good sorbent material for fluoride removal. Dolomite is available in abundance and being low cost, it holds good potential for developing safe and cost-effective methods for the defluoridation of water for drinking.

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Brief Profile of Lalit Mohan Sharma



Lalit Mohan Sharma is director, adaptive technologies (water) at S. M. Sehgal foundation. he is a graduate civil engineer and holds a masters of technology (management & systems) from Indian institute of technology, Delhi. he also served as on panel of experts for the war for water; and member of program advisory committee (PAC), water technology initiatives under the technology mission of the department of science & technology (DST), government of India; he is member of “international network on household water treatment and safe storage”

Working on water and sanitation issues, he kept trying to develop innovations in the field of water harvesting structures design, alternative sources of water, low cost water treatment and sanitation systems. number of water resource augmenting and sanitation structures with his innovative design, are serving in different states of

India. he is also contributing to rural development through sensitization and capacity building sessions around water and sanitation issues.

He innovated a sustainable low cost water filter named as JalKalp, to address arsenic, iron, biological contamination and turbidity in water. he has shared this technology with organizations working in different parts of the world to adopt for the communities in need.

His another innovation ‘creating fresh water source within saline aquifer’ was selected by united nations for showcasing at solutions summit – 2015 and he was invited to make a presentation on this innovation at un headquarters, New York. both of these innovations have also been showcased on the web site of ministry of drinking water and sanitation, government of India, at innovation page.

Under his leadership S. M. Sehgal foundation’s water management program has been recognized widely. the organization has won Unesco-water digest ‘Best Water NGO’ award for ‘revival of rural water resources in 2009 and 2010; ‘Best Water NGO’ for ‘Rain Water Harvesting’ 2008 and ‘ground water augmentation award-2010’ from ministry of water resources, government of India in 2011, FICCI water award 2013: first prize under NGO category for excellence in water management and conservation, ‘water conservation award-2013’ from the institution of engineers (India) and FICCI water award – 2017, best NGO for providing rural drinking water solutions by Unesco water digest for 2019 etc. he has written and presented several papers on issues related to integrated and sustainable development of water resources.