ISSN: 2574-1241
DOI: 10.26717/BJSTR.2018.08.001710
Saeeda Yousaf. Biomed J Sci & Tech Res

Review Article
Open Access

Efficiency of Different Indigenous Raw Materials for Removal of Arsenic from Aqueous Solution

Saeeda Yousaf* and Muhammad Ilyas
Department of Environmental Sciences, University of Peshawar, Pakistan
Received: August 30, 2018; **Published:** September 07, 2018
***Corresponding author:** Saeeda Yousaf, Department of Environmental Sciences, University of Peshawar, Pakistan

Abstract

This study was carried out to investigate arsenic removing efficiency of different indigenous materials. The materials used were including bauxite, plaster of paris, alum, activated alumina, limestone, and plastic clay. Different ratios of these raw materials were grinded, mixed thoroughly in a motor and enough water was added to the mixture to form thick slurry. The mixture is preferably calcined and is reafter converted into granular form. Removal of as from the aqueous system is readily accomplished by contacting the aqueous system with the media until the arsenic is substantially removed from the aqueous solution and their efficiency towards removal of arsenic from water was studied through atomic absorption spectrophotometer. It was found that when these materials were used individually the results showed that bauxite removed 97.5% arsenic from standard solution, alum individually removed 37%, plaster of paris 72.8%, Activated Alumina (AA) removed 40% arsenic from standard - solution and plastic clay did not show any affiliation for adsorption of arsenic. It had only 2% removal efficiency when used alone. Lime is quite effective for arsenic removal. It has 99 % efficiency for arsenic removal. Different media were developed after mixing the above said indigenous materials in different ratios. These different media showed different removal efficiencies. After conducting several experiments, it was concluded that the media developed after mixing of limestone, bauxite and plastic clay (1:1:1) showed maximum (100%) removal of arsenic from drinking water. It is an excellent media in terms of adsorbing and removal of arsenic.

Keywords: Arsenic; Removal; Raw Materials; Bauxite; Plastic Clay; Limestone

Introduction

Globally, several research studies have been reported that arsenic (As) is a carcinogen and its adverse effects are primarily due to consumption of as (100 µg/L) contaminated drinking water [1]. In Bangladesh and India, millions of people are suffering from cancer and keratosis because of chronic as poisoning from their drinking water [2]. Water contaminated with as has emerged as a serious public health concern in Pakistan. In Punjab province, >20% of the population is exposed to arsenic contamination, the as concentration being over 10 µgL-1 in drinking water while nearly 3% of the population is exposed to as over 50 µgL-1. In Sindh province, the situation is even worse; 36% of population is exposed to as contaminated water with over 10 µgL-1 and 16% of population is exposed to as 50 µgL-1. Both shallow and deep-water sources have also contaminated with As. In district Mardan, Khyber Pakhtunkhawa (KP), and the presence of as is reported to be over 50 µgL-1 [3].

The above-mentioned facts indicate that there is an urgent need for an inexpensive as treatment method. As occurs in ground and surface waters in both organic and inorganic forms, though the organic form is uncommon [4]. Arsenite (As III) is present mainly as $H3AsO_3$ (Arsenic (III) trioxide) and predominates under reducing conditions. Arsenate (As V) is common under oxidizing conditions and mainly exists as H_2AsO-4 (Dihydrogen Arsenate) and $HAsO_2-4$ (Monohydrogen Arsenate) at pH ranging from 2 to 9 [4]. The detrimental health effects of as prompted the World Health Organization (WHO) and the United States Environmental Protection Agency (US- EPA) to reduce the drinking water as standard from 0.05 to 0.01 mg/L [5]. Effectively removing as from waters with concentrations >0.01 mg/L is costly, commonly requiring expensive man-made arsenic sorbents. Consequently, there is a real need in developing countries for low-cost materials and methods to remove as from drinking water. One of the promising methods appears to be the sorption of inorganic as compounds from solution using natural sorbents.

Adsorption is a mass transfer process where a substance is transferred from the liquid phase to the surface of a solid and becomes bound by chemical or physical forces. In water treatment in the developed countries, the adsorbent (solid) is typically activated carbon, either Granular (GAC) or Powdered (PAC) and it is used for taste and odor removal. In high-tech applications, adsorption is also used for the removal of Synthetic Organic Compounds (SOCs), Volatile Organic Compounds (VOCs), and

Naturally Occurring Organic Matter (NOM). As is adsorbed onto the surface of granular materials, clays and processed cellulosic materials including: activated carbon; metal-treated activated carbon; oxides (e.g. hydrated ferric oxide, titanium oxide, silicium oxide); clay minerals (e.g. kaolinite, bentonite, Bijoypur clay); bauxite, hematite, feldspar; synthetic anion exchange resins; chitin and chitosan; bone char; iron oxide-coated or MnO_2-coated sand; cellulose materials (sawdust, newspaper pulp) [6].

Characteristically, each media has different associated performances and costs. Some are now available in small packet or tablet form for As removal from drinking water. The efficiency of each media depends on the use of oxidizing agent(s) as aids to the sorption of arsenic. The sorption capacities of various adsorbents were summarized. Some low-cost adsorbents are superior including treated slags, carbons developed from agricultural waste (char carbons and coconut husk carbons), bio sorbents (immobilized biomass, orange juice residue), goethite and some commercial adsorbents, which include resins, gels, silica, treated silica etc. The tested for arsenic removal come out to be superior. Immobilized biomass adsorbents offered outstanding performances. Desorption of as followed by regeneration of sorbents has been reported. Strong acids and bases seem to be the best desorbing agents to produce as concentrates [7].

The materials used for different media development are bauxite, plaster of paris, alum, activated alumina, commercial alumina, limestone, and plastic clay. Bauxite is a rock composed mainly of aluminum oxide and aluminum hydroxide minerals. Alum (potassium aluminum sulfate) is a white mineral. A particularly stable oxide of aluminum, aluminum oxide (Al2O3) can be obtained from alum. Activated alumina is a media produced by the controlled calcinations of alumina trihydrate. It has been used for the removal of undesirable contaminants such as fluoride, hydrogen chloride, hydrogen sulfide, carbonyl sulfide, carbon dioxide, arsenic, alcohols and ethers. Activated alumina is primarily aluminum oxide (AL2O3). Plastic clay is an extremely rare minerals found in very few places around the world. It is also sometimes referred to as ball clay. Plastic clays are sedimentary in origin. Plastic clay usually contains three dominant minerals; kaolinite, mica, and quartz.

Limestone

Limestone is defined as a rock of sedimentary origin composed principally of calcium carbonate or the double carbonate of calcium and magnesium, or a combination of these two minerals. Chemical composition of all indigenous clay materials used is given in Table 1. The basic purpose of this study is to develop an economically viable media for removal of arsenic by using indigenous materials. The materials used in the present studies are abundantly available within the country and the process developed is economically viable and commercially feasible.

Table 1: Chemical Composition of Clay's materials.

S. No	Clay/ Material	Chemical Name
1	Bauxite	Al_2O_3 (with less Si, Fe, Ti)
2	Plastic Clay	Kaoline: $Al_2O_3.2SiO_2.2H_2O$
		Mica: XY2-3 Z4O10(OH, F)2:
		X= K, Na, Ba, Ca, CS (H_3O), NH4
		Y= Al, Mg, Fe+2, Li, Cr, Mn, V, Zn
		Z= Si, Al, Fe+3, Be, Ti
		Quartz: SiO_2
3	Plaster of Paris	(Gypsum cement): $CaSO_4$. 1/2 H_2O
4	Lime stone	$Ca(Mg)CO_3$
5	Alum	$Al_2(SO_4)3.12(H_2O)$
6	Activated Alumina	Al_2O_3
7	Commercial Alumina	Al_2O_3

Table 2: Individual Material Efficiency for removal of arsenic.

Alum	Activated Alumina	Bauxite	Lime	Plastic Clay	Plaster of Paris
37.30%	39.80%	97.50%	99%	2.40%	72.8

Results and Discussion

Different media were prepared for removal of as from an aqueous system. The mixture is preferably calcined and is reafter converted into granular form. Removal of as from the aqueous system is readily accomplished by contacting the aqueous system with the media until the arsenic is substantially removed from the aqueous solution. Pseudo arsenic solution of 500 µgL-1 was prepared for this study. A total of six adsorbing materials (alum, activated alumina, bauxite, plastic clay and plaster of paris) were initially evaluated individually (Table 2). The different temperature levels for calcinations and the characteristics of the media were used (Table 3). The pH of the aqueous as solution after passing through the media was also determined to know about the pH changes due to the materials used for as removal. The present study findings indicate that samples passed through the media containing bauxite, plastic clay, activated alumina and plaster of paris have lower pH in the range of 1-6. Whereas the aqueous as solution passed through the media containing lime have higher pH in the alkaline range of 8-11.8.

Table 3: Characteristics of different media, used for the removal of arsenic.

S. No	Media	Ratio	Temp (C^0)	Characteristics of Media
Media 1	Bauxite		900	Slightly pinkish, very hard, soluble
Media 2	Bauxite + Plaster of Paris	1:01	900	Lightly pink, soluble, very hard
Media 3	Bauxite+ Plastic Clay+ Plaster of Paris	1:01:01	900	Porous, hard, slightly pink, not much soluble
Media 4	Bauxite+ Plastic Clay+ C. Alumina	1:01:01	900	Porous, hard, soluble, light pink color
Media 5	Bauxite+ Lime+ Plastic clay	2:01:01	900	Irregular, not easily soluble, pink, hard
Media 6	Bauxite+ Lime+ PC	1:01:01	900	Pink, hard, porous, not much soluble in water, slow heating

Media 7	Bauxite +Plaster of Paris+ Lime	1:01:01	900	Light pink, irregular, small in size, soluble in water, slow heating
Media 8	Bauxite + Plaster of Paris + Alum + Activated Alumina	1:1:1:1	900	Hard, slightly pink, not easily soluble, porous, slow heating
Media 9	Bauxite+ Alum+ Commercial Alumina+ Plaster of Paris	1:1:1:1	1000	White, porous, brittle, soluble
Media 10	Bauxite+ Alum+ Plaster of Paris+ Plastic Clay	1:1:1:1	900	Pink, slightly hard, porous, not much soluble
Media 11	Plaster of Paris		900	Off white, very hard, water soluble but not much, irregular,
Media 12	Activated Alumina + PP	1:01	900	White color, hard, insoluble
Media 13	Alum		900	White color, very fragile, soluble in water
Media 14	Alum+ Plaster of Paris	1:01	900	Very light pink, less hard, soluble,
Media 15	Plastic Clay		900	Very hard, insoluble, pink
Media 16	Plaster of Paris+ Plastic Clay+ Lime+ Bauxite	1:1:1:1	900	Light pink, hard, irregular, slow heating,
Media 17	Plaster of Paris+ plastic clay + Bauxite+ lime	2:2:1:1	900	Pink, hard, not much soluble, irregular
Media 18	Plastic Clay +PP	1:01	900	Hard, pink, irregular shape
Media 19	Lime		900	Not very hard, v. much soluble, irregular
Media 20	Lime+ Plaster of Paris	1:01	900	Irregular, off-white, soluble in water, crack in structure

Note: After 900C^0, starting with slow heating.

The lower pH of as solution is probably due to the presence of plaster of paris and plastic clay having sulphate contents in their luster (ref) which behave like acid whereas the cause of higher pH in the alkaline range may be due to presence of lime having basic characteristics. When bauxite alone was used 97.5% removal is achieved (Figure 1). Nil removal is achieved, when bauxite was mixed with plaster of Paris in 1:1 ratio and lumps were made after heating to 900C0 in 2 hours (Figure 2). Physically the media was pinkish in color, very hard and less soluble in water. Arsenic solution with concentration of 500 μgL-1was passed through the column containing the media and was thoroughly mixed. Elute was taken after an hour and it was analyzed through Hydride Formation System of AAS for arsenic content and the results show 0% arsenic removal. It was indicated that bauxite and plaster of paris was not effectively contributing in the removal of arsenic. Plastic clay was added with plaster of paris and bauxite in the ratio of 1:1:1. This media had 52.4% removal efficiency. In the next trial commercial alumina and plastic clay was mixed with bauxite in the ratio of 1:1:1.

Figure 1: Bauxite alone.

Figure 2: Bauxite + PP (1:1).

Figure 3: B+ Lime + PC (2:1:1).

This media had 48% removal efficiency. But when bauxite was mixed with lime and plastic clay in the ratio of 2:1:1 (Figure 3), it removed 89% arsenic from 500 μgL-1 standard solutions. The efficiency increased up to 100% when the same materials were mixed in the ratio of 1:1:1 (Figure 4). 72% of arsenic removal was obtained by a media containing bauxite, lime and plaster of paris in the ratio of 1:1:1 (Figure 5). Removal efficiency was decreased to

75.6% when bauxite, plaster of paris, alum and activated alumina were used in media (Figure 6). The media containing bauxite, plaster of paris, alum and commercial alumina were less effective and only 54% arsenic was removed with it. The efficiency of bauxite, alum, plaster of paris and plastic clay (1:1:1:1) was about 77%. Plaster of paris removes 72.8% (Figure 7). Plaster of paris when added with AA reduced as up to 70% from standard solution (Figure 8). Alum removed 37% arsenic from standard-solution but when mixed with plaster of paris (1:1 ratio) its removal efficiency increases up to 91%. Plastic clay did not show any affiliation for adsorption of arsenic (Figure 9).

Figure 4: B+PC + Lime (1:1:1).

Figure 5: B+ Lime + PP (1:1:1).

Figure 6: B + Alum+ Commercial Alumina+ PP.

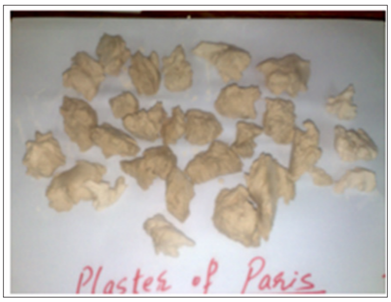

Figure 7: Plaster of paris.

Figure 8: Activated Alumina + Plaster of Paris (1:1).

Figure 9: Plastic clay.

It had only 2% removal efficiency when used alone. The combination of lime, bauxite, and plaster of paris and plastic clay 1:1:1:1 decreased arsenic by 88.7% (Figure 10). The efficiency decreased to 50.9% with the media containing plastic clay, plaster of paris, lime and bauxite (2:2:1:1). 75% removal was achieved with plastic clay and plaster of paris. Lime is quite effective for arsenic removal. It has 99 % efficiency for arsenic removal. Lime with PP in 1:1 ratio can remove arsenic up to 95%. The efficiency of the media lime, bauxite and plastic clay (1:1:1) remained maximum (100%). The results showed that the minimum as removal was observed in media without lime. The mixture of bauxite, alumina, plastic clay and plaster of paris has low efficiencies for the removal of arsenic from the aqueous solution. The arsenic removal efficiency varied among different media in the materials and raised from 0-50%, whereas, the media having lime have maximum efficiency in the removal of arsenic. The arsenic removal in these media varied from 50-100% using materials in different ratios(Table 4).

Figure 10: B+PP+PC +Lime (1:1:1:1).

Table 4: Removal of arsenic (%) using different adsorbing media.

Media	Vol of pseudo solution (ml)	Wt. of media (g)	Rotation/ min (l/hr)	pH of pseudo sol.	pH of elude	Standard solution (μgL^{-1})	As remaining (μgL^{-1})	Removal (μgL^{-1})	Removal (%)
Media 1. Bauxite	350	236	16.8	4	6.7	12.3	7.8	6.5	97.5
Media 2. Bauxite and Plaster of Paris	260	135	16.8	5.8	1.8	404.5	526.2	0	0
Media 3. Bauxite, Plastic Clay and Plaster of Paris	250	160.62	16.8	3.8	5.25	632.6	301.07	331.53	52.4
Media4. Bauxite, Plastic clay and C. Alumina	250	140	25.2	9.3	3	382	198.15	183.85	48
Media 5. Bauxite, Lime, and Plastic clay (2:1:1)	250	120	16.8	11.6	11.7	404.5	42.2	362.3	89
Media 6. Bauxite, Lime and PC (1:1:1)	470l	233	16.8	3.82	11.52	751	6.1	744.9	100
Media 7. Lime, Bauxite and PP	250	250	16.8	3.67	10.25	335.74	93.39	242.35	72
Media 8. PP, Bauxite, Alum and AA	540	311	8.4	3.55	4.9	360.57	87.85	272.72	75.6
Media 9. Bauxite, Alum, Commercial Alumina and									
Plaster of Paris	250	136	25.2	6.5	5	764	225.07	538.93	54
Media 10. Alum, Plaster of Paris, Bauxite and Plastic Clay	250	135	16.8	5.5	6.2	441.3	101.2	340.1	77
Media 11. Plaster of Paris	500	255	16.8	3.98	8.83	225.4	61.2	164.2	72.8
Media 12. Activated Alumina and PP	360	346	16.8	4.31	8.46	500	149.4	350.6	70
Media 13. Alum	190	2.8	16.8	4	6.3	12.3	7.7	4.6	37.3
Media 14. Alum and Plaster of Paris	260	125	16.8	6.5	1.86	404.5	39.4	401.9	91
Media 15. Plastic clay	300	229	16.8	4	5.91	12.3	12	0.3	2.4

Media 16. Plaster of Paris, Plastic Clay, Lime and Bauxite	680	313	16.8	3.54	12.15	226.3	25.42	200.88	88.7
Media 17. Bauxite, Lime, Plaster of Paris and plastic clay	800	482	16.8	3.66	10.4	527.8	259	268.8	50.9
Media 18. Plastic Clay and PP	270	135	16.8	5.6	8.97	441.3	109.7	331.6	75
Media 19. Lime	490	213	16.8	3.98	9.5	225.4	1.9	223.5	99
Media 20. Lime+ Plaster of Paris	250	130	16.8	11	1.6	404.5	20	384.5	95

Note: AA: Activated Alumina; PP: Plaster of Paris; PC: Plastic Clay.

Experimental

Preparation of Adsorbing Media

The different indigenous materials were used for the preparation of adsorbing media (alum, activated alumina, bauxite, plaster of paris, plastic clay and limestone). Different ratios of these materials were grinded and mixed thoroughly in a motor and enough water was added to the mixture to form thick slurry. Lumps of irregular shapes were prepared from the mixed materials, dried, ignited at 9000C in a muffle furnace for 2-h and then cooled in a desicator. The lumps were then feeded in a glass lined vertical column fitted with a stopper at the lower end. The pseudo solution of as with a known concentration was poured in the column filled with a media at a flow rate of 16.4 L/h. The solution was retained for different interval of times and then collected in a beaker through the stopper at the end of the column.

Instrumentation

Atomic absorption spectrometer (Z-8000-Hitachi Japan) in the Hydride Formation System (HFS) mode was used for the analysis of as in water samples. All samples were analyzed on HFS-3 (Hydride Formation System), an accessory (AAS, Vario 6 Analytik Jena AG) to avoid carbonates formation. The hydride technique makes use of fact that hydrogen liberated in the reaction of the weakly acidic sample solutions with sodium boro-hydride which combines with metal ions to form gaseous hydrides. These are carried to the hot quartz cell by the carrier gas and decomposed by collision processes in a series of steps, until free as atoms were obtained. From the selected materials, 14 different media were prepared with different combination and ratios and it was noted that the media prepared from limestone, bauxite and plaster of paris gave excellent results in terms of adsorbing and a better removal of as from solutions containing As.

Conclusion

Based on the results obtained, it is concluded that as could be removed from an aqueous solution using indigenous materials. The materials involved in these studies are inexpensive and abundantly available within the country. The method is simple and economically viable and could be employed for the removal / reduction of as in potable water. These bench scale studies could also be scaled up to pilot plant scale to find out the techno-economic feasibility of this method. It is low cost/ technology and may also be employed in the backward areas of the country for the reduction of as from drinking water.

Acknowledgement

The study was financially supported by the Higher Education Commission Pakistan.

References

1. Pontius FW, Brown KG, Chen CJ (1994) Health implications of arsenic in drinking water. J Amer Water Works Assoc 86: 52-63.
2. Pinon Miramontes M, Bautista-Margulis RG, Perez-Hernandez A (2003) Removal of arsenic and fluoride from drinking water with cake alum and a polymeric anionic flocculent. Fluoride 36: 122-128.
3. Ahmad T, Kahlown MA, Tah ir A, Rashid H (2004) Arsenic an emerging issue: Experiences from Pakistan. 30th WEDC International Conference, Vientiane, Lao PDR.
4. Smedley PL, Kinniburgh DG (2001) A review of the source, behaviour and distribution of arsenic in natural waters. Appl Geochem 17: 517-568.
5. WHO (1998) Guidelines for drinking water quality (2nd edn.), Addendum Geneva, Switzerland.
6. Ahmed MF (1999) Water supply options in arsenic affected rural areas. Presentation at International Arsenic Conference, Dhaka Community Hospital, Dhaka, Bangladesh.
7. Partey F, Norman D, Ndur S, Nartey R (2008) Arsenic sorption onto laterite iron concretions: Temperature effect. J Colloid Interface Sci 321:493-500.

ISSN: 2574-1241

DOI: 10.26717/BJSTR.2018.08.001710

Saeeda Yousaf. Biomed J Sci & Tech Res