

Solar photocatalytic oxidation and removal of arsenic from ground water

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In a laboratory study, the effectiveness of combined tartrate and citrate as photocatalyst in solar photocatalytic oxidation and removal of arsenic from ground water was studied and a method, using a naturally occurring tartrate-citrate photocatalyst that can be used at the household level to treat small quantities of drinking water was developed. Kinetic tests performed with 250 µg/L As(III)-spiked ground water in presence of 5 mg/L Fe(II) and 50 µM photocatalyst, showed equal efficiency of tartrate and citrate in combination for oxidation and removal of arsenic from ground water in 6 h solar illumination. Presence of silicate moderately decreased the removal of arsenic, while phosphate had more drastic effect on arsenic removal. With the increase in initial As(III) concentration above 250 µg/L, 5 mg/L Fe(II) did not achieved the removal to below 50 µg/L, although the maximum removal efficiency was 80%. Extract of tamarind (fruit of the tree *Tamarindus indica*), a natural source of tartrate-citrate (equivalent to 50 µM tartrate and citrate) achieved the arsenic oxidation and removal from 250-260 µg/L arsenic-bearing water to below 50 µg/L. The method performed within a 1-L closed PET bottle showed similar removal and can be applied at the household level to treat small quantities of drinking water.

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In many countries ground water is the primary source of drinking water, but the presence of carcinogenic toxic substances in ground water such as arsenic poses a major threat to a large number of people. Arsenic possessing both metallic and non-metallic properties, is a shiny, gray and brittle element¹ and in the environment, it can occur in several oxidation states (-3, 0, +3, +5). In natural waters it is mostly found in inorganic form as oxyanions of trivalent arsenite [As(III)] or pentavalent arsenate [As(V)]. Trivalent arsenic is more toxic than the pentavalent form. Different anthropogenic activities such as fertilizer application, uses of arsenic pesticides, and industrial discharges such as wood preservations, decolorizers in glass manufacturing, fermentation of pigments, gold mining activities, arsenic sludge factory, coal preparation waste and fly ash from coal fired plants², and manufacturing of lead shots^{3,4} are the sources of arsenic in water. Inhalation of air, ingestion of food and water, and dermal adsorption are the three major routes through which exposure can occur. Gastrointestinal and respiratory

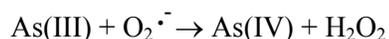
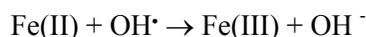
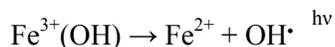
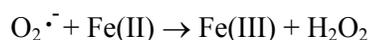
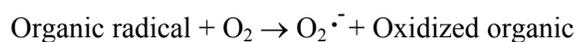
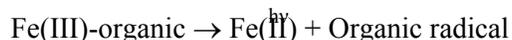
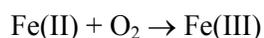
track, nervous system, and the skin⁵ are the major affected area of the body by arsenic poisoning. General muscular weakness, crawling and prickling sensations in toes, feet and legs, vomiting, inflammation of the mucus membranes, in the eye, loss of nails and hairs are the common symptoms that manifest chronic arsenic poisoning in human body^{5,6}. World Health Organization (WHO) recommended a guideline value for arsenic in drinking water of 0.05 mg/L⁷. In Argentina, Australia, Bangladesh, Chile, China, Hungary, India, Mexico, Peru, Thailand, and the United States of America, adverse health effects due to arsenic concentration greater than WHO guideline value (0.01 mg/L) have been reported. The Indian and Bangladesh drinking water standard for arsenic is also 0.05 mg/L^{8,9}.

Arsenic removal technologies generally fall into three major classes: chemical coagulation-precipitation, adsorption and membrane separation. Activated alumina¹⁰, manganese dioxide-coated activated alumina¹¹, and iron oxide and manganese dioxide-coated activated alumina¹² can be effectively used for oxidation/adsorption removal of As(V) and As(III). Iron oxide-coated sand also showed promise

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for removing As(III) and As(V) from ground water. Similarly, manganese dioxide-coated sand¹³ and iron impregnated sand¹⁴ also proved to be a good medium for removing arsenic to less than 0.05 mg As/L at domestic level. Simple filtration through charcoal yielded 70% removal of arsenic in the water¹⁵. Powdered activated carbon¹⁶ and copper-impregnated coconut husk carbon¹⁷ can adsorb As(III) from aqueous solution. Recently, some household treatment technologies such as — Alcan, BUET, DPHE/DANIDA, Sono 3-Kolsi, Stevens technology and Tetrahedron¹⁸ — were evaluated for the removal of arsenic from the ground water.

Effective oxidants used so far for oxidation of arsenic are free chlorine, hypochlorites, ozone, permanganate, Fenton's reagent, high pressure mercury lamps generating ultraviolet (UV) light in the 194-254 nm range (UV-C), and chlorine. In a recent study, solar photocatalytic oxidation of As(III) by ferric citrate-liberated photoradicals has proved to be an effective method of solar oxidation and removal of arsenic (SORAS) from ground water¹⁹. The method has been field tested in Bangladesh, using lemon juice as a natural source of citrate, to reduce arsenic concentration in ground water from 100-150 µg/L to below 0.05 mg/L²⁰. In an organic photocatalyst-mediated photooxidation and removal of arsenic, the following reactions are plausibly involved^{19,21-22}.



With a view to providing an effective method for solar photocatalytic oxidation and removal of arsenic from water, the present study was undertaken to examine the effectiveness of tartrate and citrate in

combination as a photocatalyst in solar photocatalytic oxidation and removal of arsenic from ground water, and develop a method (procedure), using a naturally-occurring tartrate-citrate (tamarind extract) photocatalyst, that can be used at the household level to treat small quantities of drinking water.

Materials and Methods

Arsenic bearing water

Water used in the study was IIT, Kanpur tap water (a direct ground water supply). An analysis of the water is given in Table 1. The water was spiked with As(III) using sodium arsenite (NaAsO₂).

Irradiation source

Sunlight was used as the irradiation source. Solar intensity during the experiment was measured with a radiometer (Model PSP, Serial No – L677F3; The Eppley Laboratory, INC., Newport, Rhode Island, USA).

Reagents

Sodium arsenite (NaAsO₂) (atomic weight 129.92 g, minimum assay 98.5%, R: 23/25-29, S: 1/2-20/21-28-45; Merck) was used to spike the ground water with As(III). Trisodium citrate 2-hydrate (C₆H₅N₃O₇·2H₂O) (molecular weight 294.1 g,

Table 1 — Analysis of I.I.T., Kanpur tap water

Parameter	Range/value
pH	7.5-8.21
Conductivity, µmho/cm at 25°C	450-550
Turbidity, NTU	3
Hardness, mg CaCO ₃ /L	145-195
Alkalinity, mg CaCO ₃ /L	240-250
TOC, mg/L	0.2
TDS, mg/L	604
Calcium, mg CaCO ₃ /L	45-70
Magnesium, mg CaCO ₃ /L	100-125
Sodium, mg CaCO ₃ /L	116
Potassium, mg CaCO ₃ /L	3
Ammonium, mg/L	2.6
Iron, mg/L	0.144-0.168
Nitrate, mg/L	0.302
Phosphate, mg/L	0.02-0.07
Fluoride, mg/L	0.65-0.75
Arsenic, mg/L	Not detected

minimum assay 99%-102.0%, B. N.: D18 S48703; E. Merck India Ltd., Mumbai - 400018) and Sodium tartrate ($[\text{CHOH.COONa}]_2 \cdot 2\text{H}_2\text{O}$) (molecular weight 230.08 g, minimum assay 99.5%, Burgoyne Burbidges & Company India Pvt. Ltd.) were used to produce photocatalyst. The iron concentration in the ground water at desired level was obtained by Ferrous sulphate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$) (molecular weight 278.01 g, minimum assay 99.5%, C. N.: 7782-63-0; Loba Chemica Pvt. Ltd., P. O. Box - 2042, Mumbai - 400002, India). Sodium phosphate monobasic ($\text{NaH}_2\text{PO}_4 \cdot 2\text{H}_2\text{O}$) (molecular weight 156.01 g, B. N.: 08800131; Sarabhai M. Chemicals, Gorwa Road, Boroda, India) and sodium metasilicate nonahydrate ($\text{Na}_2\text{SiO}_3 \cdot 2\text{H}_2\text{O}$) (molecular weight 284.2 g, minimum assay 95%, Product No. - 40220, B. N.: 1096/1496/091013; S. d. fine-chem Ltd., Mumbai - 400025, India) were used in the experiment to increase the phosphate and silicate concentrations in the ground water respectively.

Determination of arsenic

To determine arsenic in water at low concentration, the molybdenum blue method²³ was used. One mL of a 1 N hydrochloric acid and two drops of 50% saturated potassium iodate were added successively to the untreated aliquots to oxidize As(III) to As(V). Thereafter, 4 mL of mixed reagent were added to each of the 'untreated' and 'oxidised' aliquots with thorough mixing, followed by two drops of 50% saturated potassium iodate solution and 4 mL of 1 N hydrochloric acid with thorough mixing after each addition. After that to develop the blue colour all the flasks were placed in a 40°C water bath for 2 h, following which the absorbance was read in a 40-mm cell at a wavelength of 865 nm. By the above procedure, suitable blanks were run, along with the samples, and the samples were analysed in duplicates. $\text{As(III)} = (\text{absorbance of the 'oxidised' aliquots} - \text{absorbance of the 'untreated' aliquots})$ and $\text{As(V)} = \text{absorbance of the 'untreated' aliquots}$. A standard curve for arsenic was prepared and from the absorbance values the concentrations of the samples were determined.

Preparation of tamarind extract

Tamarind (fruit of the tree *Tamarindus indica*) is a natural source of tartrate and citrate and contains ca. 10% each of tartaric and citric acid^{24,25}. A set procedure was followed for preparation of tamarind extract. Twenty grams of deseeded ripe tamarind were

placed in 100 mL of water and hand mixed thoroughly. The mixture was allowed to settle overnight and the supernatant filtered through four layers of fine cotton cloth. The concentration of natural acids (tartaric and citric) in μM of the tamarind extract was estimated by alkalimetric titration²⁶ using sodium hydroxide to raise the pH of the extract to pK_{a1} value (≈ 3.1) of tartaric and citric acids.

Experimental procedure

One litre of the arsenic-bearing water (IIT, Kanpur tap water spiked with As(III) using sodium arsenite) was placed in a 1-L borosilicate glass beaker, and after 2-5 min ferrous sulphate was added to the water to make the Fe(II) concentration to the desired level, and mixed with a magnetic stirrer. After 1-2 min of iron addition, the photocatalyst (sodium tartrate and sodium citrate in combination or only sodium citrate) was added and mixed for further 10 min. Then the beakers were placed under sunlight, with continuous stirring. At predetermined time intervals, suspension samples were withdrawn and analyzed for arsenic [(total and As(III))]. Suspension samples were also filtered through 0.45 μm membrane filter and analyzed for the same.

Results and Discussion

Kinetics of oxidation and removal of arsenic from ground water with tartrate and citrate in combination or citrate as photocatalysts

Kinetics tests, using 5 mg/L Fe(II), 50 μM catalyst, and 250 $\mu\text{g/L}$ As(III)-spiked IITK ground

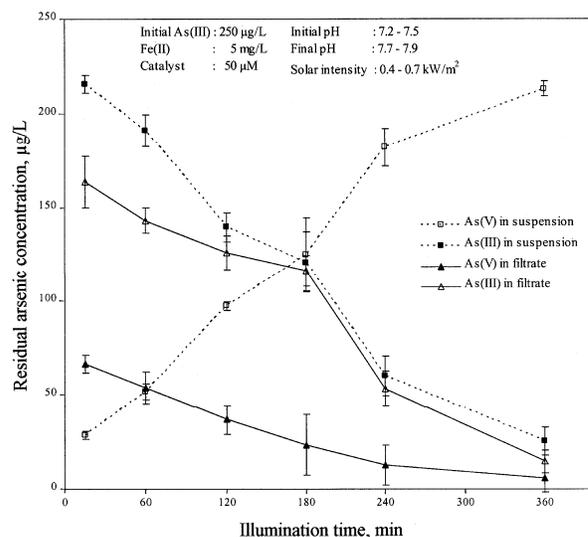


Fig. 1 — Oxidation and removal kinetics of arsenic from ground water with tartrate and citrate in combination as photocatalyst

water were conducted for 6 h under 0.3-0.7 kW/m² solar intensity and the results are shown in Fig. 1. With tartrate and citrate as photocatalyst, 30-40% of As(III) was oxidized to As(V) in 15 min and 92-95% As(III) oxidation was achieved in 6 h (residual arsenic concentration as 17-28 µg/L, in suspension). The As(V) concentration in suspension increased correspondingly with time and after 6 h, residual As(V) concentration in suspension was observed as 210-218 µg/L. From the results it was evident that tartrate and citrate in combination was an effective photocatalyst in oxidizing As(III) to As(V). Arsenic removal from ground water was studied by analyzing the filtrate. About 90-94% of arsenic was removed in 6 h, and the residual total arsenic concentration in the filtrate was 14-24 µg/L. This result indicated the efficiency of tartrate and citrate in combination for arsenic oxidation and removal from ground water.

Mechanism of arsenic removal

About 90% of the As(III) was oxidized by Fe(III)(5 mg/L)-tartrate/citrate(50 µM) complex in 6 h (Expt. 1). Negligible oxidation of As(III) to As(V) was observed in absence of photooxidation (Expt. 2). Without any added photocatalyst nearly 20% of As(III) was oxidized in 6 h on exposure to sunlight in presence of 5 mg/L Fe(II) (Expt. 3). After 6 h illumination, oxidation was rather poor (ca. 67%) in absence of any added iron and the removal was not significant (residual total arsenic concentration in filtrate 168 µg/L) (Expt. 4). In presence of 16.6 mM

2-propanol, a scavenger of OH[•], more than 90% of As(III) oxidation and removal occurred (Expt. 5), similar to Expt. 1. The precipitation of Fe(III)(hydr)oxides was observed to be enhanced by the presence of Fe(III)-tartrate/citrate complex (data not shown). The results of the Exp.1-5 are given in Table 2.

During oxidation in dark, the Fe(III)-tartrate/citrate photocatalyst had no measurable effect as the oxidants were presumably formed by photooxidation of Fe(II). Without any added iron, As(III) oxidation occurred due to the background concentration of iron in ground water. Addition of 2-propanol (quantitative scavenger of OH[•] radicals) had no influence on the As(III) oxidation, indicating that OH[•] radicals were not involved in oxidation of As(III) and that a more selective oxidant that was not scavenged by 2-propanol was involved. A schematic of As(III) photooxidation in presence of Fe(II) and tartrate citrate photocatalyst, and removal is shown in Fig. 2.

Effect of phosphate and silicate on arsenic removal

Phosphate and silicate ions present in ground water compete with As(V) for Fe(III)(hydr)oxides sorption sites^{27,28}. Photooxidation/removal of arsenic from ground water in presence of 2 mg/L phosphate or 50 mg/L silicate was studied and the data are shown in the Table 3. Silicate moderately decreased the removal of arsenic, while phosphate had more drastic effect on arsenic removal. The results also

Table 2 — Residual arsenic concentrations in suspensions and filtrates under different conditions

Expt. No.	Experimental conditions	Solar intensity (kW/m ²)	pH		Residual total arsenic after 6 h illumination (µg/L)					
			Initial	Final	In suspension			In filtrate		
					As(III)	As(V)	Total	As(III)	As(V)	Total
1	With catalyst, iron, and illumination	0.4-0.7	7.7	7.9	25 (17-31)	213 (210-218)	239 (235-243)	15 (8-20)	6 (4-8)	21 (14-24)
2	With catalyst, iron, and no illumination	—	7.2	7.4	232 (232-235)	16 (15-18)	248 (247-250)	226 (223-229)	12 (9-15)	238 (237-240)
3	With iron, illumination, and no catalyst	0.3-0.7	7.2	8.1	187 (181-190)	50 (48-54)	237 (235-240)	138 (123-155)	36 (32-40)	175 (160-195)
4	With catalyst, illumination, and no iron	0.3-0.8	7.3	7.9	48 (43-53)	167 (195-205)	248 (245-250)	36 (28-40)	132 (130-133)	168 (160-175)
5	With catalyst, iron, illumination, and 2-propanol	0.5-0.6	7.6	8.3	42 (37-50)	206 (200-210)	248 (247-250)	19 (14-28)	12 (11-14)	32 (25-40)

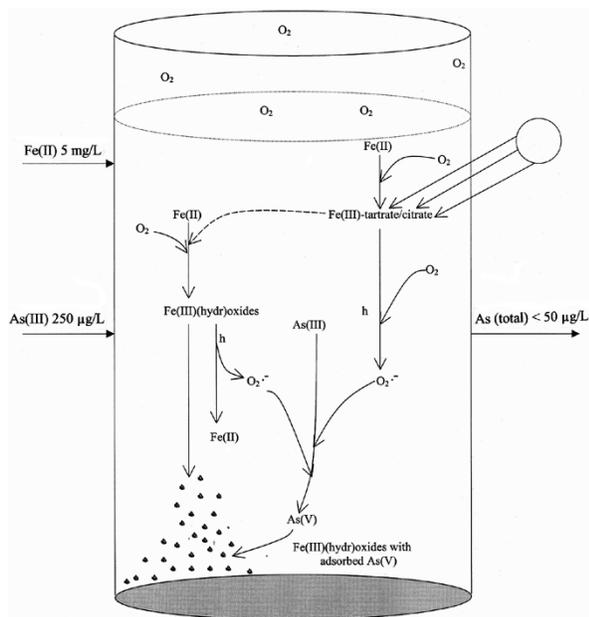


Fig. 2 — Schematic of As(III) photooxidation and removal

suggested that the affinity of phosphate for Fe(III)(hydr)oxides was much greater than that of silicate.

Effect of initial As(III) and Fe(II) concentrations on arsenic removal

Oxidation/removal of arsenic with varying initial As(III) concentration (250, 500 and 1000 µg/L) and Fe(II) concentration (2, 3, 4 and 5 mg/L) and with 50 µM photocatalyst, was studied and shown in the Table 4. It is observed that when the initial As(III) concentration in the ground water is 250 µg/L, at least 4 mg/L Fe(II) is required to achieve total arsenic removal to below 50 µg/L. Under same conditions, 3 mg/L and 2 mg/L Fe(II) reduce residual total arsenic concentration to 72 and 80 µg/L, respectively. When initial As(III) concentration is 500 µg/L or more, no given Fe(II) concentration achieves the removal to below 50 µg/L. This is presumably due to scarcity of Fe(II) to form photocatalyst and precipitate to adsorb As(V).

Table 3 — Residual total arsenic concentrations in filtrates in presence of phosphate and silicate anions (with catalyst and iron)

Expt. No.	Anion	Solar intensity (kW/m ²)	pH		Residual total arsenic in filtrate after 6 h illumination (µg/L)
			Initial	Final	
1	—	0.4-0.5	7.7	7.9	14-24
2	Phosphate (2 mg/L)	0.4-0.5	7.4	7.8	61-72
3	Silicate (50 mg/L)	0.4-0.5	7.5	8.1	45-57

Potential of tamarind extract as a natural photocatalyst for solar photocatalytic oxidation and removal of arsenic

Two stirred beaker experiments (in duplicate) – one with the arsenic bearing water [(As(III) 250 µg/L)] and other with a natural arsenic-bearing water [As(III) 235 µg/L; As(V) 25 µg/L], collected from A.G. Colony, North 24 Parganas, West Bengal, and with 5 mL of tamarind extract (equivalent to 50 µM tartrate and citrate) were conducted to study the potential of tamarind extract as photocatalyst for oxidation and removal of arsenic from ground water. The results shown in the Table 5 indicate the

Table 4 — Residual total arsenic concentrations in filtrates for different initial As(III) and Fe(II) concentrations

Expt. No.	Run No.	Solar intensity (kW/m ²)	pH		Initial As(III) (µg/L)	Fe(II) (mg/L)	Residual total arsenic in filtrate after 6 h illumination (µg/L)
			Initial	Final			
I	1(a)		7.3	8.0	250	2	80
	1(b)	0.3-0.4	7.3	8.0		3	72
	1(c)		7.2	7.8		4	30
II	2(a)		7.0	7.8	500	3	178
	2(b)	0.3-0.5	7.2	7.8		4	120
	2(c)		7.3	8.0		5	105
III	3(a)		7.2	8.0	1000	3	230
	3(b)	0.3-0.4	7.1	7.7		4	207
	3(c)		7.2	8.2		5	180

Table 5 — Residual total arsenic concentrations in IITK ground water and in A.G. Colony ground water after stirred-beaker experiment with natural photocatalyst

Expt. No.	Water	Solar intensity (kW/m ²)	pH		Residual total arsenic in filtrate after 6-8 h illumination (µg/L)
			Initial	Final	
1(a)		0.2-0.7	6.8	8.5	43
1(b)	Arsenic-bearing water with Fe(II) 5 mg/L (added)		6.9	8.2	25
2(a)		0.3-0.7	7.3	8.1	35
2(b)	A.G.colony ground water with Fe(II) 4.48 mg/L		7.1	8.3	39

Table 6 — Residual total arsenic concentrations in A.G. Colony ground water after experiment in a closed PET bottle with tamarind extract

Expt. No.	Solar intensity (kW/m ²)	pH		Residual total arsenic in filtrate after 6-8 h illumination (µg/L)
		Initial	Final	
1	0.3-0.7	7.4	8.1	40
2		7.3	8.2	47

effectiveness of tamarind extract as photocatalyst for oxidation and removal of arsenic from actual arsenic-bearing ground water.

Oxidation and removal of arsenic with tamarind extract as photocatalyst in closed PET bottle

Arsenic oxidation and removal experiment was also performed in closed polyethylene terephthalate (PET) bottle so as to apply the process for treatment of small quantities of drinking water at the household level. A 1-L PET bottle was filled with 750 mL of the A.G. colony ground water [As(III) 235 µg/L; As(V) 25 µg/L], followed by addition of one teaspoon (5 mL) of tamarind extract, giving a total photocatalyst concentration of about 50 µM. Aeration was done by shaking the bottle manually for 30 s, and the bottle was exposed horizontally to sunlight for 6-8 h. After illumination, the bottle was slowly rotated for 10 min and was placed vertically overnight. Thereafter, the supernatant was filtered through four layers of fine

cotton cloth and the filtrate was analyzed for residual total arsenic. The results given in the Table 6 indicate that solar oxidation and removal of arsenic from ground water in closed PET bottle with tamarind extract as photocatalyst is a feasible method that can be used at the household level to treat small quantities of drinking water.

Conclusions

Tartrate and citrate in combination appeared to be an equally effective photocatalyst and decreased the residual total arsenic concentration to below 50 µg/L. Solar illumination, Fe(II) and photocatalyst are the three necessary conditions for oxidation and removal of arsenic. In absence of any added photocatalyst, Fe(III)(hydr)oxides in ground water plays the role of photocatalyst. Arsenic removal efficiency was dependent upon the concentration of initial As(III) and Fe(II) in the ground water. With increase in initial As(III) concentration up to 1000 µg/L, removal efficiency decreased although the Fe(II) concentration in the ground water was 5 mg/L. Competing ions, such as phosphate and silicate reduced the removal of arsenic. Tamarind extract (equivalent to 50 µM of tartrate-citrate) applied to the arsenic-bearing water removed arsenic to below 50 µg/L. Experiment within a closed PET bottle with tamarind extract under 6-8 h solar illumination and then filtration by four layers of fine cotton cloth showed the feasibility of the method to treat small quantity of drinking water at the household level.

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