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Abstract: A semicircular section tubular photoreactor has been constructed, characterized and applied to the treatment of groundwater contaminated with As(V) by means of the SORAS (solar oxidation and removal of arsenic) technique, using ferrous and citrate salts. The solar concentrator was built with recyclable waste materials: glass tubes from fluorescent lamps and 6-inch diameter PVC pipes cut in half and covered by aluminum foil. The reactor concentrates solar radiation up to 2.8 times its natural intensity. Batch irradiation experiments followed by controlled agitation (shear rate = 30-33 s⁻¹; 20 min agitation period) showed that the photoreactor accelerates the formation of settleable floccules $(D_p > 0.5$ mm), compared with a fluorescent lamp glass tube alone and a 2 L PET (polyethylene terephthalate) bottle. Irradiation times necessary for floccule formation in the photoreactor, the fluorescent lamp tube and the PET bottle were 15 min, 25 min and 60 min, respectively. Continuous flow experiments using a photoreactor with a photo-collection area of 0.9 m² and a hydraulic retention time (equal to the irradiation time) of 15 min showed that immediate formation of floccules of good settleability occurs when the solution is subjected to moderate agitation (33 s^{-1}) . An efficiency of 98.36% for As(V) removal was obtained with a final concentration of 16.5 μ g/L in decanted waters. In accordance to these results, the photoreactor is able to treat approximately 130 L/m² within a 5-h period with UVA irradiation intensities of 50-70 W/m².

Key words: Tubular photoreactor, arsenic removal, SORAS (solar oxidation and removal of arsenic), groundwater, flocculation.

1. Introduction

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High concentrations of naturally occurring arsenic in ground and well water poses a health hazard in many parts of the world. Among the most affected countries are Bangladesh, India, Chile and Argentina [1, 2]. Recently, it has been reported that shallow wells located at rural areas—with scarce water sources—around the southern shore of Poopó Lake in the Bolivian Altiplano have arsenic contents in the range of $5.6 - 242 \mu g/L$ [3].

Long-term daily consumption of water with a high

arsenic content has caused severe diseases in humans such as skin and lung cancer, hyperkeratosis and damage to the central nervous system [4, 5].

To address this problem, several research groups have worked on the development of easy to use technologies using UVA (ultraviolet A) solar radiation. Hug et al. [6] developed the so-called SORAS (solar oxidation and removal of arsenic) technique—a simple household treatment that uses PET (polyethylene terephthalate) bottles and lemon juice drops—which was applied with relative success in Bangladesh, where concentrations vary in the range of 100-150 μ g/L, obtaining As(III) removal efficiencies of 50%-70%.

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The SORAS technique uses the interaction of UVA solar radiation, the natural iron, Fe(II-III), contained in Bangladesh groundwater and the citrates in the lemon juice to form Fe(III) oxide precipitates which adsorb As(III-V) ions. The precipitates can then be filtered and removed from the solution.

In Chile, the SORAS technique, modified by addition of ferrous sulfate and sodium citrate salts, has been tested by Lara et al*.* [7] to obtain arsenic removals greater than 95% for waters from Camarones River which has very high concentrations of As(V) $(1,250 \mu g/L)$. Excellent results for the removal of As(III) and As(V) have been reported by Cornejo et al. [8] for Camarones river waters. Utilizing lemon juice as the citrate source and steel shavings as Fe(0) source, they obtained arsenic removals higher than 99.5% and total arsenic concentrations below 10 μ g/L, satisfying the WHO (World Health Organization) recommended value [9].

Despite promising results, these batch processes are slow (they last 3-6 hours), their treatment capacities are small, making them unsuitable for the larger consumption needs of rural populations such as crop irrigation, domestic animal breeding and water use in community health centers and schools. To satisfy these needs, the development of a practical and low-cost treatment system of larger capacity is desirable. To date, no studies have been reported on the treatment characteristics and performance of continuous flow photoreactors for arsenic removal using the SORAS approach.

The research efforts reported here were focused on the development and application of a semicircular section tubular solar concentrator for arsenic removal from groundwater by means of a modified SORAS method using ferrous and citrate salts. The concentration of solar UVA radiation up to several times the usual solar intensity would permit the increase of the reaction rates involved in the arsenic removal, thus increasing treatment capacity. The reactor was constructed with locally available, low cost, recycled materials. In this study, the optical attributes of the built photoreactor were characterized and its performance for arsenic removal was evaluated for both batch and continuous mode operation.

2. Optical Characterization

2.1 The Photoreactor

PVC (polyvinyl chloride) pipes of 16 cm internal diameter and 1 m long were used as optical collectors. They were split at the axis to obtain the chosen semicircular cross section as shown in Fig. 1.

Spent fluorescent light tubes made of clear glass were proposed as reaction containers due to their higher transmittance in the UVA (ultraviolet A) range (290-390 nm) as compared to PET bottles (0.3 mm thickness). This can be seen in Fig. 2, which depicts the transmission spectra of both materials in the range of 270-400 nm. Table 1 shows the values of transmittance measured by a UVA radiometer for both the PET bottle films and the glass tube walls. These measurements confirm the higher transmittance for spent fluorescent glass tubes.

Fig. 1 Design schematics of the built tubular photoreactor.

Fig. 2 Transmittance spectra for PET bottle and spent fluorescent glass tube.

	PET wall, 0.3 mm			Fluorescent glass tube wall, 0.9 mm			
Sample No.	Incident UVA radiation $(W/m2)$	UVA radiation transmitted $(W/m2)$	Transmittance percentage $(\%)$	Incident UVA radiation (W/m^2)	UVA radiation transmitted $(W/m2)$	Transmittance percentage (%)	
1	49.6	35.2	71.0	52.6	47.0	89.4	
$\overline{2}$	49.8	36.6	73.5	52.5	47.3	90.1	
3	50.0	35.6	71.2	52.3	47.4	90.6	
$\overline{4}$	50.1	36.3	72.5	53.0	43.0	81.1	
5	51.5	38.8	75.3	53.3	43.0	80.7	
6	51.4	36.2	70.4	53.4	40.0	74.9	
7	51.8	38.6	74.5	53.7	42.5	79.1	
8	52.0	38.4	73.8	53.6	40.1	74.8	
9	52.2	39.0	74.7				
10	52.3	37.5	71.7				
Media	51.07	37.22	72.78	53.1	43.8	82.6	
SD.	1.07	1.41	1.80	0.53	3.08	6.6	
$RSD(\%)$	2.10	3.80	2.47	1.00	7.04	7.98	

Table 1 UVA transmittance percentages for PET bottle wall (0.3 mm thickness) and spent fluorescent glass tube walls (0.9 mm thickness).

The tubes of 3.6 cm external diameter and 0.9 mm thickness were located at the focus of the semicircular section.

The inner surface of the tubes was covered by aluminum foil in order to increase the reflectance of solar UVA radiation. Aluminum has a high reflectance index (0.92) in the UVA range of the solar electromagnetic spectrum, as shown in Fig. 3 [10].

2.2 Effective Power Received by Solution within Glass Tubes

As an approximation, the effect of UVA reflection obtained over the shadowed area was considered (Fig. 1). The effective UVA power received by water solution within glass tubes is:

$$
P_{tube}^{collector} = I_0 (A_0 \cdot T_{tube} + 2A_1 \cdot R_{Al} \cdot
$$

$$
T_{tube} + A_0 \cdot T_{As} T_{tube} R_{Al} T_{tube})
$$
 (1)

where, I_0 : incident solar radiation intensity (W/m²), A_0 , A_I : area perpendicular to incident radiation (m²), R_{AI} : aluminum reflectance (-), T_{tube} : glass tube transmittance (-), $P_{tube}^{collector}$: power received by the tube in solar concentrator (W) and T_{As} : transmittance of water solution containing As, Fe (II) and sodium citrate (-). The term $(A_0 \cdot T_{As}T_{tube}R_{Al}T_{tube})$ results

from the product of the area perpendicular to incident radiation, A_0 , by transmittance of the two layers of glass, transmittance of water solution containing As, Fe(II) and sodium citrate and reflectance of the aluminum foil.

The radiation intensity received by the water solution becomes:

$$
I = P_{tube}^{collector} / A_{tube}
$$
 (2)

These parameters were measured experimentally at noon on a clear day. The results are shown in Table 2.

The effective perimeter, *leffective*, corresponding to the area of collector that receives solar radiation when the solar radiation is perpendicular to the collector axis, was measured via laser and can be approximated by the following relation:

$$
l_{effective} = \left(\frac{1}{2.5}\right) \cdot 2\pi r \tag{3}
$$

where, *r*: inner radius of PVC tube (m).

Area A_2 (Fig. 1) does not make any contribution in reflecting radiation towards the glass tube when radiation is perpendicular to the collector. However, this area does contribute effectively when radiation has an incident angle with respect to the perpendicular.

Due to the semicircular geometry of the collector,

Fig. 3 Reflectance spectra for different vacuum electrodeposited metals [10].

Table 2 Experimental data for the calculation of the effective power received by water solution inside glass tubes.

Parameter	Value
Tube wall transmittance (T_{tube})	0.851
Aluminum foil reflectance (R_{Al})	0.80
A_0 (m ²)	0.0400
$2A_{I}$ (m ²)	0.0913
T_{As}	0.70

the effective perimeter is constant from approximately 11:00 a.m. to 2:00 p.m., and thus the reflected radiation reaching the glass tube remains the same over this time period without adjusting the position of the collector. Before 11:00 a.m. and after 2:00 p.m., the collector wall can cast shadows that reduce the amount of effective radiation. However, the useful time period of the collector can be taken from 9:00 a.m. to 3:00 p.m. with some loss during the initial and final hours.

Using the experimental data from Table 1 in the 290-390 nm range, Eq. (1) can be evaluated to obtain the intensity of UVA radiation received by the solution within the glass tube:

$$
P_{tube}^{collector} = 0.112 \times I_0 \tag{4}
$$

The ratio of power received by collector-tube to the power received by the tube alone is given by

$$
\frac{P_{tube}^{collector}}{P_{tube}} = \frac{0.112 \times I_0}{0.04 \times I_0}
$$
 (5)

where, $P_{tube} = 0.04 \times I_0$ is the power received by the tube alone.

Thus:

$$
P_{tube}^{collector} = 2.8 P_{tube} \tag{6}
$$

and
$$
I_{tube}^{collection} = 2.8I_0
$$
 (7)

Eq. (7) implies that the intensity of the radiation received by the solution contained inside the glass tube is 2.8 times the incident solar radiation. In Cochabamba (the location of all data collected), the maximum solar UVA radiation in November is approximately 70 W/m^2 . Thus, the effective UVA radiation received by the solution is 196 W/m².

Higher intensities of UVA radiation inside the solar concentrator are expected to increase photochemical reactions rates. This is confirmed by experimentally studying removal rates of As(V) using the modified SORAS method as discussed below.

3. Experimental Methods

3.1 Chemical Reagents

All chemical compounds were analytical grade (Merck and Sigma, Aldrich), except commercial grade sodium citrate $(C_6H_5Na_3O_7.2H_2O$, 98.68% purity). Solutions for analytic determination of As(V) were prepared with distilled and deionized water. Dibasic sodium arsenate $(Na_2HAsO_4·7H_2O)$ was used for As(V) standards. Ferrous sulfate (FeSO₄ $·7H₂O$) was the iron source for the photochemical reaction.

3.2 Determination for As(V)

The spectrophotometric method modified by Johnson [11] was used at 905 nm using a Shimadzu UV-1601 model. The mixed reagent for color development was optimized adding L-ascorbic mass with twice the dosage recommended by Strickland [12]. A $0-1,000 \mu g/L$ range calibration curve was constructed, which showed to be linear within the interval of interest, as shown in Fig. 4.

For distilled waters spiked with As(V), samples were immediately filtered through 0.45 μ m membranes before performing analytical tests. Absorbances were measured after exactly 2 h, using quartz 1.0 cm light path cells.

For ground waters spiked with As(V), pentavalent phosphorous interferes the analytical determination of As(V), so FAAS (flame atomic absorption spectrometry) with hydride generation was used for total arsenic determination.

3.3 Arsenic Removal Tests

3.3.1 Batch Irradiation without Agitation—Distilled Water Spiked with As(V), $1,000 \mu g/L$

Simultaneous tests were carried out using the photoreactor, a fluorescent tube alone and a 2 L transparent PET bottle, attached to an aluminum frame which was placed on a rotating table. The apparatus enables variation of the inclination from 0° to 45° and the azimuthal angle from 0° to 360°, to facilitate sun position tracking. Fig. 5 shows the experimental apparatus with the three reactors in two positions.

The procedure for arsenic removal (used in batch tests) was as follows: iron sulfate and sodium arsenate salts were added to 1 L or 2 L of distilled water spiked with As(V) $(1,000 \mu g/L)$ to obtain a molar relation of As:Citrate:Fe(II) = 1:5:19, as recommended by Lara et al. [7]. The solution was aerated without solar radiation to guarantee sufficient dissolved oxygen and

Fig. 4 Calibration curve for As(V) analytical determination in distilled water solutions.

Fig. 5 Experimental apparatus tracking sun position during irradiation.

then divided among the three photoreactors: the glass-tube of the solar concentrator, the glass-tube alone and a 2 L PET bottle. The three photoreactors were exposed to sunlight without agitation during a 2-4 h period. Samples were taken at 2.5 min intervals during the first 15 min to 25 min of exposure. Moreover, times for visible floccules appearance were measured for each of the reactors. Integral UV radiation was measured every 10 min with a PCE-UV34 radiometer (290-390 nm interval).

3.3.2 Batch Irradiation Followed by Controlled Agitation—Distilled Water Spiked with As(V), 1,000 g/L

A similar procedure was carried out with the following modification: 2 L of solution (distilled water spiked with 1,000 μ g-As(V)/L) were expose to sunlight during 5 min, 10 min and 15 min for the solar collector; 5 min, 20 min and 25 min for the glass-tube alone and 30 min, 40 min, 50 min and 60 min for the 2 L PET bottle.

Immediately following irradiation, the solutions were submitted to controlled agitation inside laboratory (RUVA = 0.1 W/m²), using a 2 L beaker without deflectors and a stirrer provided with 3 inch \times 1 inch blade located at 2.25 inch from the bottom. Angular velocity was 50-52 rpm, to obtain a shear rate of 30-33 s^{-1} at 30 °C (Fig. 6) [13].

Times of visible floccules appearance were

measured during the agitation period of 30 min. Floccule size and settleability were evaluated using the Willcomb index (Table 3) [13].

3.3.3 Continuous Flow Irradiation Followed by Controlled Agitation—Groundwater Spiked with As(V), $1,000 \mu$ g/L

50 L of groundwater were spiked with 1,000 μ g-As(V)/L. A continuous flow of 440 mL/min corresponding to 15 min hydraulic residence time (equal to the irradiation time) was run through the photoreactor. The system was exposed to sunlight during approximately 1 h. After a steady-state was obtained, the solution was collected. The solution was then agitated at 33 s^{-1} shear rate inside laboratory $(RUVA = 0.1 W/m²)$, as described previously. Times of visible floccules appearance were measured during the agitation period of 30 min. Floccule size and settleability were evaluated using the Willcomb index, as before. Fig. 7 shows the tubular photoreactor of 6.6 L effective volume.

4. Results and Discussion

4.1 Batch Irradiation without Agitation—Distilled Water Spiked with As(V), 1,000 $\mu g/L$

Fig. 8 shows concentration versus irradiation time relationships for the three photoreactors. It can be seen that As(V) removals higher than 90% are achieved within the first 10 min in the solar concentrator.

Fig. 6 Relationships between rpm and shear rate at different temperatures for a 2 L beaker, without deflectors [13].

Table 3 Willcomb index [13].

Fig. 7 Continuous flow solar concentrator.

Fig. 8 As(V) concentration-time profiles for solar concentrator, fluorescent glass tube and 2 L PET bottle. Spiked distilled water (1,000 ppb As(V)).

During the same period of exposure, the tube alone and the PET bottle only removed approximately 10%. After an exposure of 15-16 min, As(V) concentrations at the latter photoreactors decrease dramatically, although more slowly in the case of the PET bottle. This behavior is explained as follows: colloidal floccules $(< 0.45 \text{ µm})$ that already adsorb As(V) have higher growth rates in the solar concentrator in which UVA effective radiation intensity is 2.8 times incident radiation. In the case of both the tube alone and the PET bottle, the effective radiation intensity is lower than the incident radiation due to the wall (glass tube transmittance = 0.85 ; PET bottle transmittance = 0.73) and solution transmission resistances.

As shown in Table 4, similar behavior was observed for the appearance of visible floccules. Within the solar concentrator, floccules appear at 44 min compared to 54 min within the glass tube alone and 105 min in the PET bottle. In all cases, after 4 h irradiation time, the floccules agglomerate achieving particle sizes larger than 2 mm and excellent Willcomb index values (floccules settle completely leaving the water clear).

The observations show that floccule growth rate depends on the effective UVA radiation intensity reaching the solutions. Larger quantities of photons, produced by larger UVA intensities, accelerate the series of reactions proposed by Hug et al. [6]: photolysis of citrate—Fe(III) complex, the formation of hydroxyl (OH^{*}), superoxide (O_2^*) free radicals and H_2O_2 and, finally, the Fe(OH)₃ precipitate formation.

4.2 Batch Irradiation Followed by Controlled Agitation—Distilled Water Spiked with As(V), 1,000 μ g/L

Table 5 shows the results obtained for floccule appearance times and floccule sizes in tests using the solar concentrator, the tube alone and the 2 L PET bottle. In all cases, floccule appearance times decreased due to the effect of controlled agitation when compared to the times obtained without agitation. It is also observed that longer irradiation times generate larger and better quality floccules. Times for the formation of good quality floccules with good settleability were: 60 min for the PET bottle, 25 min for the fluorescent glass tube and 15 min for the

Reactor	Incident UVA $\rm (W/m^2)$	Effective UVA $\rm (W/m^2)$	Appearance time of visible floccules (min)	Willcomb index after 4 h irradiation (-)	Floccule size (mm)
	62.6	27.3	95	10	>2
2 L PET bottle	63.0	27.5	115	10	>2
Ne tube	62.6	39.0	50	10	>2
	63.0	39.2	59	10	>2
	62.6	175.3	45	10	>2
Solar concentrator	63.0	176.4	43	10	>2

Table 4 Batch irradiation of spiked distilled water without agitation.

Table 5 Batch irradiation of spiked distilled water followed by controlled agitation (shear rate = 33 s^{-1}).

Reactor	Incident UVA		Effective UVA Irradiation time	Floccule appearance time	Willcomb	Floccule size
	(W/m ²)	(W/m ²)	(min)	during agitation (min)	index	(mm)
	34.9	12.5	31	No flocculation	$\mathbf{0}$	$\overline{0}$
	57.5	25.1	30	21	$0 - 2$	< 0.1
2 L PET bottle	60.6	26.5	40	12	$2 - 3$	$0.1 - 0.3$
	61.4	26.8	50		$4-6$	$0.5 - 0.75$
	63.9	27.9	60	0.5	8	$0.75 - 1.5$
	52.3	32.6	15	No flocculation	Ω	θ
Ne tube	62.1	38.7	20	2	$6 - 8$	$0.75 - 1.0$
	64.3	40.1	25		8	$1.0 - 1.5$
	61.8	38.5	26		$6 - 8$	$0.75 - 1.0$
	67.3	188.4	5	No flocculation	Ω	θ
	63.3	177.2	10	2	4-6	$0.5 - 0.75$
	53.7	150.4	10	26	$\overline{2}$	< 0.3
Solar concentrator	50.5	141.4	10	60	$0 - 1$	< 0.1
	44.4	124.3	15		$8 - 10$	$0.75 - 1.0$
	64.9	181.7	15		$8 - 10$	$0.75 - 1.5$

solar concentrator. Finally, for the same irradiation times, when solar UVA radiation increases, the times for floccule appearance decrease and their quality and size improve.

These results show that greater UVA radiation intensities accelerate not only the formation kinetics of the $Fe(OH)_{3}$ precipitates, but also the floccule growth rates. Thus, the increase of radiation intensities causes the acceleration of the As(V) removal global process.

4.3 Continuous Flow Irradiation Followed by Controlled Agitation—Groundwater Spiked with As(V), $1,000 \mu g/L$

Once the optimum irradiation time of 15 min was determined for the solar concentrator, a calculated volumetric water flow of 0.440 L/min for an effective volume of 6.6 L/m^2 (yielding a hydraulic retention time of 15 min) was introduced to the photoreactor. Groundwater spiked with $1,000 \mu$ g-As(V)/L was used. Its physical and chemical characteristics are shown in Table 6.

It can be observed that bicarbonate (a major constituent of total alkalinity) is predominant as compared with other anions (nitrate, chloride and sulphate), implying a favorable water quality condition for the SORAS process, due to the following reasons: the alkalinity provided by bicarbonate ions probably enhances iron hydroxide precipitation [14] and bicarbonate may generate new oxidizing species that favor As(V) removal as suggested by Hug, et al. [15]. On the contrary, nitrate and sulphate may compete with arsenate species for the adsorption sites in the iron hydroxide surface [16]; however, their concentrations are very small in comparison with those reported in other studies [8, 14, 1080

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17].

The collected waters were agitated in 2 L beakers at 50-52 rpm (shear rate = $30-33$ s⁻¹) during 20 min. The results obtained in two runs are presented in Table 7. Floccules with a Willcomb index of 6-8 (size of $0.75-1.0$ mm) are shown in Fig. 9, within a 2 L beaker.

Floccule appearance times during agitation were almost immediate and floccule qualities were sufficient for an appropriate sedimentation. Settled waters achieved 98.36% As(V) removal and the residual pentavalent arsenic concentration was 16.45

Table 7 Continuous flow irradiation of spiked ground water (1,000 μ g-As(V)/L), followed by controlled agitation.

Fig. 9 Flocculation (Willcomb Index = 6-8) under controlled agitation (shear rate = 33 s⁻¹). Spiked groundwater (1,000 **g-As(V)/L).**

 μ g/L, a value low enough to be easily reduced to satisfy the WHO guide value of $10 \mu g/L$ by filtration devices.

With a volumetric flow rate of 440 mL/min, one photoreactor square meter is able to treat approximately 130 L of contaminated water per day, assuming a 5 h daily operation period (10:00 a.m.-3:00 p.m.). This daily capacity can supply safe water to a 4-person family at a consumption rate of 30-35 L/person/d, a typical value in rural areas in the Bolivian Altiplano.

5. Conclusions

A semicircular section tubular photoreactor has been constructed and characterized. The device has been applied to the treatment of groundwater spiked with pentavalent arsenic, using the SORAS (Solar UV Radiation Assisted Removal of Arsenic) technique. The solar concentrator was built reusing disposable materials: fluorescent glass tubes, 6 inch diameter sewage PVC tubes covered with aluminum foil. The photoreactor concentrates solar radiation intensity up to 2.8 times the incident value.

Simultaneous batch runs carried out under integral UVA irradiation (50 W/m² and 70 W/m²) on distilled water spiked with $1,000 \mu g/L$ As(V) showed that colloidal floccules $($0.45 \mu m$) that already adsorb$ As(V) have higher growth rates in the solar concentrator, compared with fluorescent glass tubes and the 2 L PET bottles, in that order. The same behavior is observed for visible floccules appearance times. Again, within the solar concentrator, floccules appear at 44 min compared to 54 min within the glass tube alone and 105 min in the PET bottle.

Controlled agitation (shear rate = $30-33$ s⁻¹) after solar UVA irradiation increased floccule growth rates until they become visible. The required irradiation periods of time were 15 min, 25 min and 60 min for the solar concentrator, fluorescent glass tubes and 2 L PET bottles, respectively.

It can be concluded that greater effective UVA solar

radiation intensities and controlled agitation at typical shear rates increase floccule growth rates as well as the overall As(V) removal process rates.

Continuous flow regime experiments, using a photoreactor of approximately 0.9 m^2 irradiation area in which an As(V) spiked $(1,000 \mu g/L)$ groundwater containing typical cation and anion concentrations was treated, demonstrated excellent removal efficiencies for As(V) $(> 98\%$; remnant As(V) concentration in decanted waters = 16.5 µg/L when the hydraulic retention time was similar to the irradiation time of 15 min. It follows that approximately 130 L/m^2 of contaminated groundwater can be treated, assuming a 5 h daily operation period (10:00 a.m.-3:00 p.m.) with 50-70 W/ m^2 solar UVA radiation intensities.

The semicircular section tubular photoreactor can be used for removal of As(V) contained in groundwater from many countries including Bolivia and Bangladesh, where similar UVA radiation intensities occur.

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