Solar Energy in the Treatment of Arsenic-Contaminated Surface Waters

Llerlin Chapa Tomanguilla, Ing., Carlos A. Castañeda Olivera, Dr. and Elmer Benites-Alfaro, Dr.

Universidad César Vallejo, Campus Los Olivos, Lima, Peru
lchapat@ucvvirtual.edu.pe, caralcaso@gmail.com, ebenitesa@ucv.edu.pe

Abstract– Surface water contaminated with arsenic as a result of anthropogenic activities or naturally occurring events is harmful to health and the environment. Thus, the research proposed to use solar energy as a technology for the treatment of arsenic-contaminated water from the Rimac river in Lima, Peru. The treatment consisted of using solar radiation with an average rate of 550.18 W/m², with the presence of iron wire as an absorbent and lemon juice as a catalyst. The results showed that arsenic was reduced from 1.64 mg/L to 0.128 mg/L (92.2% efficiency), using 4g of iron wire and 6 mL of lemon juice. The resulting treatment data were statistically processed by multiple linear regression, and the linear model was found to be: Y = 0.417 + 0.0930X₁ + 0.0234X₂ - 0.0014X₃; where X₁ is the amount of iron wire (g), X₂ is the lemon juice (mL) and X₃ is the solar radiation (W/m²). Finally, it is concluded that the method is effective in the reduction of arsenic in surface water with the advantage of being easy to use, low cost and environmentally friendly.

Keywords: arsenic; contaminated water; water treatment; solar radiation; solar energy.

I. INTRODUCTION

Arsenic exposure continues to be a prevalent human health problem worldwide [1]. The presence of arsenic in water has been reported in several places: In Bangladesh, 50 to 60 million people were reported to be consuming arsenic-contaminated groundwater [2]. In India it was also reported that approximately 6 million people were exposed to arsenic contamination, as well as in Taiwan, China and in the USA more than 350 thousand people consume water with 0.5 mg/L of arsenic [3]. In Peru, research indicates that 86% of groundwater exceeds the maximum permissible limits (MPL) of 10 μg/L stipulated by the World Health Organization [4].

Arsenic is commonly found in water in the trivalent and pentavalent form [5]. The arsenic (III) removal process takes place after it is transformed into arsenic (V), and in the presence of iron hydroxide it precipitates and facilitates the separation. This precipitation occurs because the iron metal has a positive oxidation number that attracts the negatively charged arsenic, and consequently its precipitation [6].

Advanced photocatalytic oxidation with TiO2 is a technology to reduce gaseous pollutants, and it could also be used in combination with other technologies such as biodegradation to be more successful in reducing arsenic [7],

as is the case of the biomass of the fungus *Aspergillus niger* that removed Arsenic (V) up to 69% in a contact time of 24 hours [8], and the biomass of the bacteria *Pseudomonas aeruginosa* that achieved an arsenic reduction of 60% in 3 months [9]. This technology was also applied to reduce pharmaceutical contaminants in water, achieving better efficiency when solar photocatalytic oxidation was combined with ozonation [10]. Other methods used to decrease arsenic are iron-based metal filters called ZVI filter [11].

An applicable method to propose as a remediation alternative for arsenic in surface water is solar oxidation. This method has been previously studied in groundwater treatment using continuous flow processes with both artificial UV radiation [12] and sunlight [13], reducing arsenic concentrations from 1000 μg/L to less than 10 μg/L. This technology was also used in the treatment of water for human consumption using solar radiation of 586 W/m² for 6 hours, obtaining arsenic reduction percentages greater than 88% [14]. Therefore, this research promotes the use of solar oxidation as a simple technology to effectively reduce arsenic concentration in water. It is a simple, practical and applicable treatment in areas where it is difficult or expensive to install large water treatment structures such as the Rimac river basin. This basin is located in the San Mateo district in Lima, Peru (11° 46' 6.503" S, 76° 18' 14.962" W). It is also mentioned that the samples collected from the Rimac River presented arsenic concentrations above the MPLs, and as a consequence there is a danger of environmental damage and damage to people's health. The presence of arsenic in the waters of the Rimac River is related to the different mining activities carried out in the area, which due to climatic and meteorological conditions produce acid mine drainage, tailings leachate and sulfate mineralization, causing contamination by heavy metals such as arsenic in receiving bodies such as soil and water in the Rimac River basin [15].

II. MATERIALS AND METHODS

2.1. Materials and equipment used

- DAVIS portable meteorological station, model VANTAGE PRO 2 PLUS
- YSI digital potentiometer, model pH100
- Water sampling kit

2.2. Research design

The design was experimental with pre- and post-test [16]. The research was conducted *in situ*, specifically in the area characterized by good exposure to solar radiation and close to the Rimac River (San Mateo, Lima). Fifteen tests were carried out with homogeneous samples that were collected from the sampling point.

Samples coded as M1, M6 and M11 were used for the blank tests and samples coded as M2, M3, M4, M5, M7, M8, M9, M10, M12, M13, M14 and M15 were subjected to the solar oxidation tests, with different doses of iron (iron wire), lemon juice and time of solar exposure. After treatment, the arsenic concentration was verified by atomic absorption spectroscopy analysis.

2.3. Stages of the research

2.3.1. Field work

Water samples from the Rimac River were collected following the National Protocol for Monitoring the Quality of Surface Water Resources [17], and the amount taken was 5 liters per day for 3 consecutive days.

For the solar oxidation of arsenic, the methodology of Litter et al. [18] was followed:
a. The collected water was immediately deposited in polyethylene terephthalate containers, prior measurement of pH and temperature.
b. The doses of iron (iron wire) and lemon juice were then added in the amounts foreseen for the test.

c. Then, the samples were exposed to a variable period of time to solar radiation, starting at 7:00 AM until 5:00 PM to capture the highest solar radiation.
d. After the exposure and resting time, the solutions were filtered to separate the precipitate. Subsequently, pH was measured and arsenic analysis was performed.

2.3.2. Cabinet work

The results of the experiment were systematized and analyzed to determine the most optimal operating conditions for reducing arsenic with solar oxidation. In addition, an inferential analysis was performed using Minitab software. For this, multiple linear regression was used to fit a linear model between several variables such as lemon concentration, iron concentration and solar radiation to estimate the level of arsenic reduction.

III. RESULTS

3.1. Physicochemical parameters

The results of the physicochemical parameters of the water samples both before and after the solar oxidation process are presented in Table 1. The values of the initial and final arsenic concentration of the 16 worked samples are also shown.

Table 1 shows that sample M4 obtained the highest reduction in arsenic concentration (optimum value), reaching a value of 92.2%, using 4 g of iron (iron wire), 6 mL of lemon juice, and a solar radiation index of 550.18 W/m^2.

Table 1 Result of the physicochemical parameters and concentration of arsenic.

Days	Samples	Iron wire (g)	Lemon juice (mL)	Time (h)	Solar radiation (W/m²)	pH		Temperature		Arsenic		Reduction rate (%)
						Initial	Final	Initial	Final	Initial	Final	
1	M1	0	0	0	0	0	0	0	0	1.64	0	0
	M2	2	2	2	303.83	6.06	5.89	14.2	27.5	1.64	0.283	82.74
	M3	3	4	4	444.46	6.8	4.58	13.7	31.4	1.64	0.213	87.01
	M4	4	6	6	550.18	7.08	4.12	13.7	26.2	1.64	0.128	**92.2**
	M5	5	8	8	590.21	6.03	3.91	14	26.5	1.64	0.472	71.22
2	M6	0	0	0	0	0	0	0	0	2.2	0	0
	M7	3	2	2	293.24	5.89	5.63	14.3	26.7	2.2	0.465	78.86
	M8	4	4	4	428.14	5.87	4.72	14.3	33.8	2.2	0.242	89
	M9	5	6	6	523.72	6.03	4.03	13.8	24.7	2.2	0.418	81
	M10	6	8	8	540.25	6.1	5.57	14	26.9	2.2	0.404	81.64

Days	Samples	Iron wire (g)	Lemon juice (mL)	Time (h)	Solar radiation (W/m²)	pH		Temperature		Arsenic		Reduction rate (%)
						Initial	Final	Initial	Final	Initial	Final	
3	M11	0	0	0	0	0	0	0	0	0.835	0	0
	M12	2	8	2	292.24	5.59	3.84	14.7	30	0.835	0.248	70.3
	M13	3	6	4	407.69	5.73	4.11	14.4	31.5	0.835	0.316	62.16
	M14	5	4	6	463.34	5.78	4.01	14.3	24.5	0.835	0.293	56.65
	M15	6	2	8	392.44	5.83	3.85	14.2	24.2	0.835	0.362	64.91

3.2. Solar radiation

The results shown in the graph refer to the measurement of solar radiation the analysis period was three days, where we found a maximum temperature of 16.5 ° C. These monitored values regarding solar radiation were found between 11:00 am and 12:00 pm, see Figure 1.
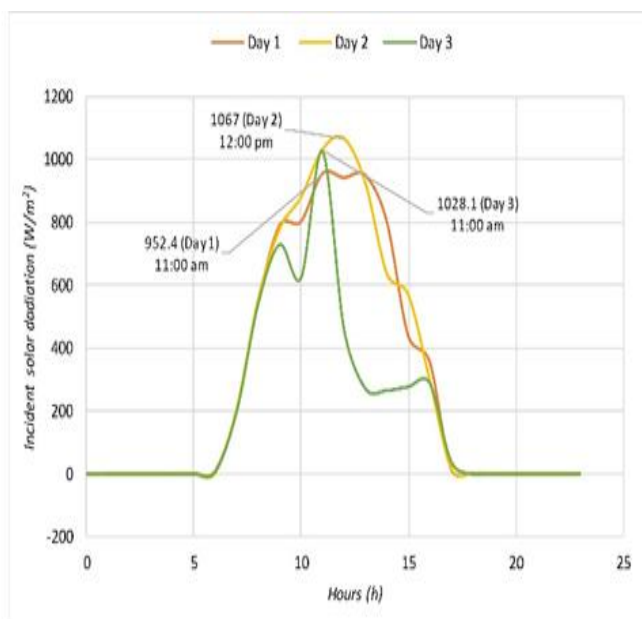

Figure 2. Arsenic concentration and solar radiation index as a function of exposure time: Sample M4.

Figure 1. Hourly variation of solar radiation index: Sample M4.

The reduction of arsenic with respect to the solar radiation index as a function of the exposure time of the M4 sample is shown in Figure 2. It was observed that after 6 hours of solar exposure with a value of 550.10 W/m² of solar radiation the concentration initial arsenic from 1.64 mg/L decreases to 0.128mg/L.
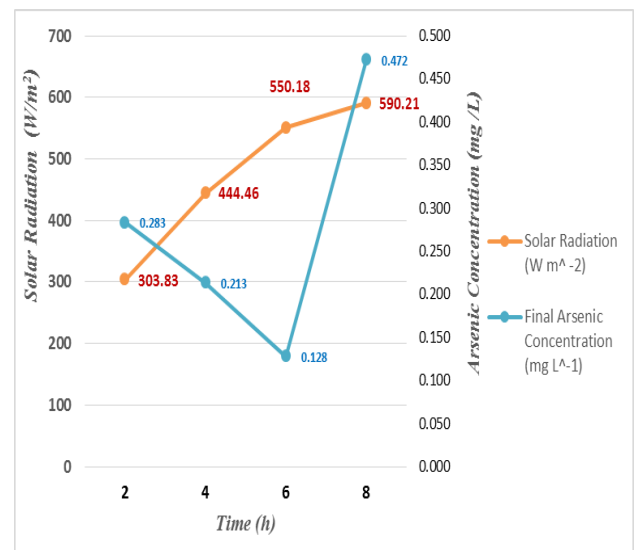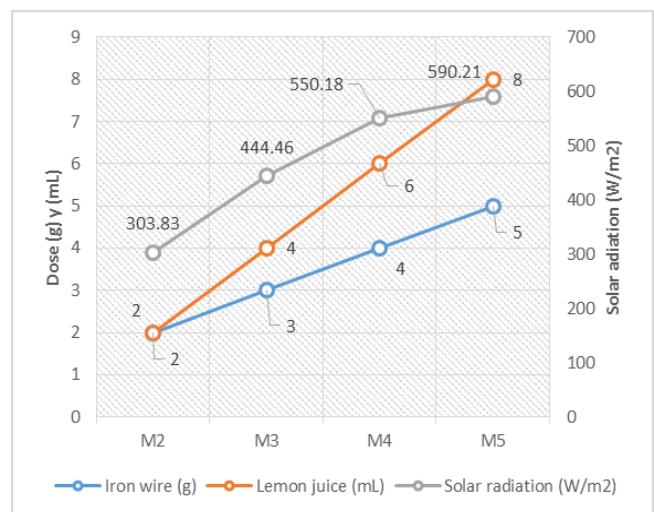

Figure 3. Best control group analysis: Sample M4.

Figure 3 shows the data for day 2, which is considered to be the most representative day and where the greatest arsenic reduction was obtained (sample M4). The optimal treatment doses to reduce the arsenic concentration were 4g of iron wire, 6mL of lemon juice and a solar exposure period of 6 hours reaching a solar radiation value of 550.15 W/m^2.

3.3. Influence of iron, lemon juice and solar radiation in the oxidation process

In order to establish the influence of iron, lemon juice and the radiation index in the solar oxidation process for the reduction of arsenic in the contaminated waters of the Rimac River, inferential statistics were used through multiple linear regression, which allows estimating the involvement of several independent variables in a dependent variable of interest (arsenic concentration) [19]. The multiple linear regression model is shown in Equation 1, and their respective coefficient values are shown in Table 2.

$$Y = a + b_1 X_1 + b_2 X_2 - b_3 X_3 \qquad (1)$$

Where: X_1 is the amount of iron, X_2 corresponds to the volume of lemon juice, X_3 represents the solar radiation and Y is the approximation corresponding to the concentration of arsenic.

Table 2 Coefficients of multiple linear regression model

Coefficients	Value
a	0.417
b_1	0.0930
b_2	0.0234
b_3	0.0014

In Table 3, Y_{real} is the result of arsenic concentration after solar oxidation treatment, and $Y_{theoretical}$ is the result of arsenic concentration calculated by the model (Eq. 1).

Table 3 Result of the real and theoretical arsenic concentration

X_1	X_2	X_3	Y_{real}	$Y_{theoritical}$
3	2	292.24	0.465	0.339
4	4	428.14	0.242	0.291
5	6	523.72	0.418	0.299
6	8	540.25	0.404	0.416
2	2	303.83	0.283	0.23
3	4	444.46	0.213	0.176

X_1	X_2	X_3	Y_{real}	$Y_{theoritical}$
4	6	550.18	0.128	0.17
5	8	590.21	0.472	0.254
2	8	292.24	0.248	0.387
3	6	407.69	0.316	0.274
5	4	463.34	0.293	0.336
6	2	392.44	0.362	0.48
4	6	625.7	0.002	0.066
4	6	625.7	0.002	0.066
4	6	625.7	0.002	0.066

The multiple linear regression model presented an R^2 equal to 63.35%, that is, a significant percentage indicating that the dependent variable (arsenic concentration) is influenced by the independent variables. On the other hand, it is also important to indicate that it is not necessary to maximize the R^2 value to be able to say or interpret that the model is good, but rather that the purpose of the model is to find estimators that are exact to the coefficients of the original regression [20]. This model helps to predict the responses, the selection of the variables that influence the results, as well as to discard the variables that do not provide information.

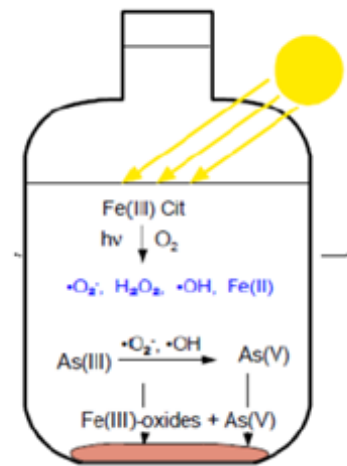3.4. Solar oxidation operability

Figure 4. Basic principle of the solar oxidation method. Source: [23]

The arsenic is removed by the iron wire which hydrolyzes forming hydroxides, in which the arsenic adsorbs and precipitates with other metal ions, i.e. the iron wire oxidizes

and traps the arsenic when it comes in contact with water. This happens because arsenic has a negative charge so that by using the iron wire which has a positive charge, it will attract the arsenic to precipitate continuously, see Figure 4.

Lemon juice facilitates the oxidation process, such as the generation of oxidants due to the action of solar radiation.

As for the solar radiation, it is the one that generates very strong oxidants, which are the ones that oxidize the arsenic and bring it to an oxidized state, where it has the ability to adhere to the iron oxides, which then precipitate to the bottom of the vessel.

IV. DISCUSSION

The maximum arsenic concentration reduction of 1.64 for 0.128 mg/L is corroborated by Rojas-Chaves [21], who obtained arsenic reductions from 0.2 to 0.01 mg/L using the solar radiation technique (1200 W/m^2) during 2 hours of exposure. With the same technology, S. Duarte et al [22] achieved arsenic reduction percentages of 90%. The differences in the results indicate the influence of operational conditions such as solar radiation index, exposure time and others considered for the development of each experiment.

Regarding the values of iron dose, lemon juice and solar radiation, it was found that there is a relationship between them. This effect could be corroborated with the statistical treatment of the data by means of multiple linear regression. In the investigation, the maximum (92.2%) arsenic reduction was achieved with 4g of iron, 6mL of lemon juice and an average solar radiation of 550.18 W/m^2. With these same inputs, Chávez Q. and Miglio T. [14] achieved arsenic reductions between 88 and 98.5%, with a solar radiation of 586 W/m^2.

From the statistical analysis of the data by means of multiple linear regression, it is suggested that there is a significant linear effect between arsenic reduction and the doses of iron wire, lemon juice and solar radiation indexes. It is also established that the applied model implies an acceptability percentage of 63.35%, resulting in a good model for future predictions in solar oxidation for arsenic reduction.

The results found indicate that the solar oxidation process reduces the level of arsenic concentration in water, as verified by the works of Gill and O'Farrell [12], Santos-Domínguez et al. [8], F. Almomani et al. [10] and Litter et al. [18].

V. CONCLUSIONS

It was determined that the application of solar energy as an oxidation technology reduced the arsenic concentration level of the analyzed water samples. The maximum arsenic reduction was 92.2% with 4 g of iron wire and 6 mL of lemon juice under solar radiation index of 550.18 W/m^2 for 6 hours of exposure.

The treatment results were evaluated by multiple linear regression, obtaining the model: $Y = 0.417 + 0.0930X_1 + 0.0234X_2 - 0.0014X_3$; where: X_1 = Iron wire (g), X_2 = Lemon juice (mL) and X_3 = Solar radiation (W/m^2). This indicated that at least one of the worked variables (X_1, X_2 and X_3) significantly influences the reduction of arsenic concentration.

REFERENCES

[1] E. A. Hull *et al.*, "Human health risk from consumption of aquatic species in arsenic-contaminated shallow urban lakes," *Sci. Total Environ.*, vol. 770, p. 145318, May 2021.

[2] S. J. Hug, L. Canonica, M. Wegelin, D. Gechter, and U. Von Gunten, "Solar oxidation and removal of arsenic at circumneutral pH in iron containing waters," *Environ. Sci. Technol.*, vol. 35, no. 10, pp. 2114–2121, May 2001.

[3] M. L. Castro de Esparza, "Arsénico en el agua de bebida en América Latina y su efecto en la salud pública," 2006.

[4] OMS-Organización Mundial de la Salud, "Guías para la calidad de agua potable: Anexo 3 - Cuadro de Información resumida sobre sustancias químicas," 2008.

[5] A. A. Duker, E. J. M. Carranza, and M. Hale, "Arsenic geochemistry and health," *Environ. Int.*, vol. 31, no. 5, pp. 631–641, Jul. 2005.

[6] R. Chang and K. A. Goldsby, *Química*, 11a edició. China: McGraw-Hill Interamericana de España S.L., 2013.

[7] F. A. Almomani, R. R. Bhosale, M. A. M. M. Khraisheh, A. Kumar, and C. Kennes, "Mineralization of dichloromethane using solar-oxidation and activated TiO2: Pilot scale study," *Sol. Energy*, vol. 172, pp. 116–127, Sep. 2018.

[8] E. E. Santos-Domínguez, J. M. Vargas-Morales, J. F. Cárdenas-González, and I. Acosta-Rodríguez, "Remoción de arsénico (V) en solución acuosa por biomasa modificada del hongo aspergillus Niger," *Inf. Tecnol.*, vol. 28, no. 6, pp. 45–52, 2017.

[9] E. E. Pellizzari, L. G. Marinich, S. A. Flores, and C. M. Giménez, "Degradación de arsénico por pseudomonas aeruginosa para bioremediación de agua. Estudio preliminar," *Av. en Ciencias e Ing. ISSN-e 0718-8706, Vol. 6, N°. 1, 2015, págs. 1-5*, vol. 6, no. 1, pp. 1–5, 2015.

[10] F. Almomani, R. Bhosale, A. Kumar, and M. Khraisheh, "Potential use of solar photocatalytic oxidation in removing emerging pharmaceuticals from wastewater: A pilot plant study," *Sol. Energy*, vol. 172, pp. 128–140, Sep. 2018.

[11] B. Casentini, F. T. Falcione, S. Amalfitano, S. Fazi, and S. Rossetti, "Arsenic removal by discontinuous ZVI two steps system for drinking water production at household scale," *Water Res.*, vol. 106, pp. 135–145, Dec. 2016.

[12] L. W. Gill and C. O'Farrell, "Solar oxidation and removal of arsenic - Key parameters for continuous flow applications," *Water Res.*, vol. 86, pp. 46–57, Dec. 2015.

[13] C. O'Farrell, J. Mac Mahon, and L. W. Gill, "Development of a continuous flow solar oxidation process for the removal of arsenic for sustainable rural water supply," *J. Environ. Chem. Eng.*, vol. 4, no. 1, pp. 1181–1190, Mar. 2016.

[14] M. L. Chávez Quijada and M. Miglio Toledo, "Remoción de arsénico por oxidación solar en aguas para consumo humano," *Rev. la Soc. Química*

del Perú, vol. 77, no. 4, pp. 307–314, 2011.

[15] C. M. George *et al.*, "Arsenic exposure in drinking water: an unrecognized health threat in Peru," *Bull World Heal. Organ*, 2014.

[16] R. Hernández S., C. Fernández C., and M. del P. Baptista L., *Metodología de la investigación*, 6 Ed. México: McGRAW-HILL, 2014.

[17] ANA-Autoridad Nacional del Agua, "Protocolo Nacional para el Monitoreo de la Calidad de los Recursos Hídricos Superficiales. R.J. 010-2016-ANA," Lima-Perú, 2016.

[18] M. I. Litter, H. D. Mansilla, and R. Gettar, *Remoción de arsénico asistida por luz solar en comunidades rurales de América Latina.Proyecto OEA AE 141/2001*. Agencia Interamericana para la Cooperación y el Dearrollo, 2003.

[19] F. javier Barón López and F. Téllez Montiel, "Apuntes de bioestadística: Tercer ciclo de en ciencias de la Salud y Medicina," España, 2004.

[20] M. Martínez Rodríguez, "Errores frecuentes en la interpretación del coeficiente de determinación lineal," *Anu. Juridico y Econ. Escur.*, no. 38, pp. 315–331, 2005.

[21] P. Rojas-Chaves, M. J. Vargas-Benavides, A. Araya-Obando, J. Valverde-Cerdas, and L. G. Romero-Esquivel, "Estudio de remoción de arsénico en agua potable a nivel domiciliar mediante oxidación solar y coagulación-floculación," *Rev. Tecnol. en Marcha*, vol. 28, no. 4, pp. 54–65, 2015.

[22] A. A. L. S. Duarte, S. L. C. Oliveira, and T. Amorim, "Arsenic removal from drinking water by advanced filtration processes," 2011.

[23] Wegelin,Martin G,Daniel et al, "SORAS- a simple arsenic removal process.Bangladesh: 26th WERD Conference Dhaka, Bangladesh," 2000.