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Adsorption Efficiency of UV-Irradiated Polyvinyl Alcohol-Alginate Adsorbent Beads for Chromium (VI) Removal

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Abstract

Due to the risk of chromium (Cr) contamination, industrial waste emissions have become a global concern, necessitating an effective adsorption approach. Polyvinyl alcohol (PVA) – alginate beads are a promising material for overcoming this problem. The present work aims to investigate the effect of ultraviolet (UV) light irradiation on the preparation of PVA-alginate beads throughout a range of UV irradiation durations, as well as its Cr (VI) removal rate. In this work, a homogeneous gel made of PVA and sodium alginate (SA) was produced and then radiated to UVA light for varying periods of time (5-20 minutes). Subsequently, the PVA-alginate adsorbent beads were made by crosslinking with 6% boric acid and 2% calcium chloride. PVA-alginate adsorbent beads were fabricated easily after UVA irradiation because the viscosity of gel PVA-alginate was reduced. The Cr (VI) adsorption experiment employed 15 g of PVA-alginate adsorbent beads, 180 minutes of adsorption time, and 50 mg/L Cr (VI) initial concentration. The size analysis revealed that the average diameter of the adsorbent beads after Cr (VI) adsorption was approximately 4.00 mm. The UV-Vis results showed that sample A prepared by exposing the PVA-alginate adsorbent beads to UV light for 5 minutes has an optimum Cr (VI) removal rate of 71.63% after in contact for 180 minutes. This study's findings were critical for establishing a new knowledge of the production of PVA-alginate beads irradiated with UV light and moving forward to mitigate heavy metal pollution in waste water, therefore enhancing the quality of water and the environment.

1. Introduction

Water pollution has arisen as a major worldwide problem, threatening aquatic ecosystems as well as human health. Heavy metals and dyes have received special attention among contaminants due to their persistence and possible toxicity [1]. Heavy metals, known for their non-biodegradability and persistence over time in aquatic ecosystems, are a particularly dangerous component of water pollution. Heavy metals' negative impacts on the aquatic environment have sparked concern regarding toxicity and carcinogenicity [2]. Water pollution by harmful heavy metals like chromium (Cr), which is predominantly caused by industrial activity, is of special concern [3]. This bleak outlook is worsened by increasing urbanisation and unrestrained manufacturing emissions, resulting in a troubling decrease in clean water availability [4].

Heavy metals, such as trivalent chromium [Cr (III)] and hexavalent chromium [Cr (VI)], have gained notoriety for their harmful impacts on human health and the environment. Cr (VI), with its high toxicity and possible carcinogenicity, endangers both aquatic creatures and human health [3,5]. Taking in Cr (VI) can have serious consequences for important organs such as the kidneys and liver [5]. Industrial discharges, notably those from the chemical, painting, and mining sectors, are the primary sources of chromium contamination in soil and water systems. Conventional wastewater treatment procedures frequently fail to adequately mitigate Cr (VI) pollution, spurring a quest for novel solutions to this environmental hazard [3].

Adsorption methods have emerged as a viable route in the quest for effective and sustainable pollution cleanup [6]. The combination of polyvinyl alcohol (PVA) and marine biopolymer alginate has significant beneficial towards heavy metal removal in wastewater. Because of its non-toxic, biocompatible, and rapidly gelling characteristics, alginate derived from brown seaweed has a remarkable affinity for heavy metals [7]. PVA-alginate beads demonstrate outstanding adsorption capacities when coupled with PVA, a water-soluble polymer recognised for its powerful mechanical qualities and flexibility. These beads have a distinct advantage due to their high mechanical strength, durability, and hydraulic qualities [8]. This novel technique corresponds to the growing demand for ecologically acceptable and cost-effective heavy metal pollution mitigation methods [6].

The combination of UV light exposure with polymers has received a lot of attention in the wastewater treatment industry. This method has been demonstrated to be successful as an adsorbent for removing different pollutants from water sources [9,10]. On top of that, UVA irradiation plays a major role in this process because it initiates the photodegradation of contaminants, increases polymer adsorption capacity, and promotes the breakdown of organic molecules [11]. Previous research has looked at the beneficial interactions of UV light exposure with polymers, and the results have been positive. Al-Ghouti et al. [12] revealed that combining UV irradiation with graphene oxide enhanced phenol removal from contaminated water substantially, with the maximum adsorption capacity rising with UV exposure from 70.43% to 90.82% for GO1 and from 86.75% to 95.95% for GO2. These findings highlight the possibility of UV light exposure and polymers as a practical and efficient technique for environmental remediation in water treatment.

Therefore, the current study investigates the intriguing role of UVA light in the manufacturing of PVA-alginate beads and their capacity to reduce heavy metal pollution. By subjecting these beads to varied lengths of UV light radiation, this study focuses on the influence of this unique characteristic on bead production and performance. This finding is a big step towards closing a critical research gap concerning the interaction of UV radiation with polymer bead manufacturing. Through extensive testing and analysis, the study hopes to shed light on the delicate relationship between UV light exposure and the creation of PVA-alginate beads with improved heavy metal adsorption capacities.

2. Experimental Procedures

2.1 Preparation of Polyvinyl Alcohol-Alginate Adsorbent Beads

Prior to forming the beads, 12 g of PVA (R&M Chemicals) was dissolved into 53 mL of distilled water, while 2.5 g of SA (QRëC) was added to the other beaker, which had 20 mL of distilled water. Both solvents were microwaved for 2.5 minutes at 100°C. Then, the PVA and SA gels were mixed via stirring method until a uniform gel was obtained. The homogeneous gel was then irradiated for 5-20 minutes. The parameters of sample preparation are shown in Table 1. In the present study, an ultraviolet light type A (UVA) was employed, and the distance between the samples and the UVA lamp was kept constant at 26 cm.

Table 1 *PVA-alginate adsorbent beads and their UV irradiation duration*

Sample	UV irradiation time (minutes)
CS	0 (control sample)
A	5
B	10
C	15
D	20

Subsequently, a syringe was used for producing the PVA-alginate beads, which were then put into a crosslinking solution containing 6% boric acid (QRëC) and 2% calcium chloride (HmbG) while stirring constantly. The bead mixture was then allowed to rest for 24 hours to allow the gelation process. Following the gelation process, the PVA beads were washed with distilled water to remove any leftover borate ions. PVA beads were treated with 0.5 M sodium sulphate (QRëC) after 30 minutes to improve their properties and chemical stability. Prior to the Cr (VI) removal test, the PVA-alginate adsorbent beads were maintained at 4°C.

2.2 Cr (VI) Removal Test

In this present study, 50 mg/L of potassium dichromate ($K_2Cr_2O_7$) was prepared. Then, 50 mL of Cr (VI) solution and 15 g of PVA-alginate adsorbent beads were added. Subsequently, the adsorption experiment was conducted using UVA light at 365 nm and at room temperature. The UVA lamp maintained a 15 cm distance between the UVA light and the beaker. The removal rate of Cr (VI) by the PVA-alginate adsorbent was calculated using Equation 1, where, C_o (mg/L) denotes the initial Cr (IV) concentration and C_e (mg/L) denotes the equilibrium Cr (IV) concentration in solution.

$$\text{Adsorption } (\%)=\frac{(C_o- C_e)}{C_o} \text{ x } 100 \tag{1}$$

2.3 Characterisation

Following the Cr (VI) adsorption test, 20 PVA-alginate beads were picked randomly for each of the designated parameters in order to measure the diameter of the adsorbent with a vernier calliper. Each reading was repeated five times, and the average was calculated to obtain the readings' mean. The adsorbent's structural properties were determined using Fourier transform infrared spectroscopy (FTIR) at wavelengths ranging from 400 to 4000 cm^{-1}. Additionally, the concentration of Cr (VI) was determined using ultraviolet-visible spectroscopy (UV-Vis) with wavelength at 540 nm. Prior to the UV-Vis test, the concentration of Cr (VI) was examined using the diphenyl carbazide (DPC) method with a detection limit of 5 g/L. In a volumetric flask, 1 mL of each experiment's sample was combined with 0.2 mL of newly made 0.25% (w/v) DPC (QRC) in acetone (Merck Schuchardt OHG) and 9 mL of 0.2 M H_2SO_4 (Merck Schuchardt OHG). To ensure complete colour development, the mixture was vortexed for 15-30 seconds and left to stand for 10-15 minutes.

3. Results and Discussion

3.1 Size Analysis

Adsorbent bead size was identified by measuring the diameter of 20 beads of PVA-alginate adsorbent from each sample using vernier calliper. The average diameter of the PVA-alginate adsorbent beads (sample CS) before UV treatment was 4.20 mm. Meanwhile, the average diameter for PVA-alginate adsorbent beads after being irradiated with UV light for sample A, sample B, sample C, and sample D, are 4.10, 4.07, 4.05, and 4.00 mm, respectively. Fig. 1 depicts the average diameter of the PVA-alginate adsorbent beads before and after being irradiated with UVA light of present study and comparison with previous works.

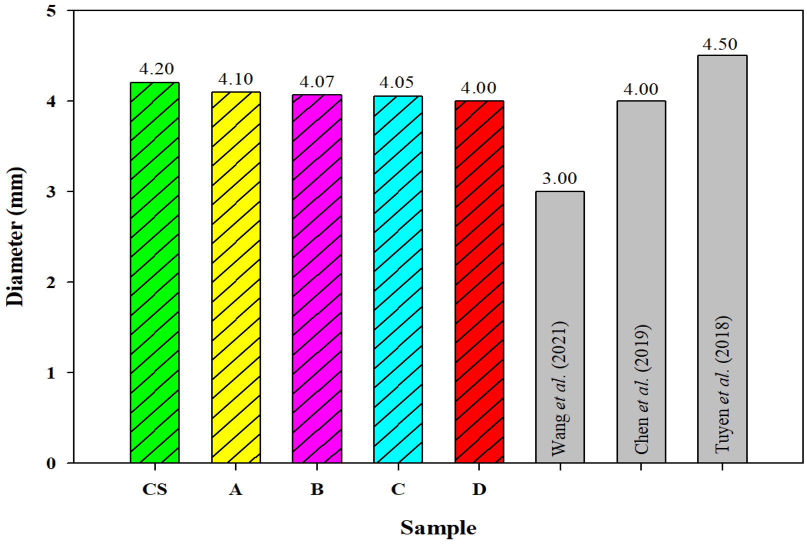

Fig. 1 *Diameter of PVA-alginate adsorbent beads with or without UV irradiation and comparison with previous works [13-15]*

The average diameter of the PVA-alginate adsorbent beads in the present study was around 4 mm. Meanwhile, the length of UV irradiation appears to have an effect on the size of the adsorbent beads. The longer the UVA irradiation is exposed to the PVA-alginate adsorbent beads, the smaller the diameter of the beads. Based on Fig. 1, the average diameter of PVA-alginate beads was 4.50 mm in the previous studies, and the findings of this study were comparable with the prior studies, where the average diameter of PVA-alginate beads was about 4.00 mm. UVA radiation promotes photooxidative degradation, which breaks polymer chains, creates free radicals, and lowers molecular weight, causing the mechanical properties to deteriorate and leading to chemical and physical instability [16]. It is noteworthy to mention that after being exposed to UVA light, the PVA-alginate adsorbent beads changed colour from white to transparent. It is thought that UVA radiation causes the PVA to lose its ability to keep the rigidity of the beads. As a result, the longer the PVA-alginate adsorbent beads are exposed to UVA radiation, the smaller they get.

The current study discovered that the diameter of PVA-alginate beads might vary depending on the material utilised in producing them. The diameter of PVA-alginate beads measured by Wang *et al.* [13] was less than that of the current study. It is because different materials were employed and several preparation variables were modified in the current study.

3.2 FTIR Analysis

Adsorbent functional groups such as hydroxyl and carboxyl groups play a significant part in Cr (VI) removal. The FTIR spectra of PVA-alginate adsorbent beads are shown in Fig. 2. The peaks of the PVA-alginate spectra at 3300, 2909, 1600, 1418, 1333, and 1094 cm^{-1} matched to hydroxyl and carboxyl stretching. The broad peak formed between 3500 cm^{-1} and 3300 cm^{-1} is due to the strong hydroxyl (O-H) stretching vibration of cellulose molecules from the UVA irradiation [17]. On the other hand, C-H stretching has a high of 2906 cm^{-1} [18]. However, after more than 5 minutes of UV irradiation, the peak of 2906 cm^{-1} disappeared in samples B, C, and D. The dissolved peak (C-H component) impacted the PVA-alginate adsorption performance. The presence of O-H bending of adsorbed water was indicated by the band at 1600 cm^{-1}, while the presence of C-H bending caused vibrational peaks at 1418 cm^{-1} and 1333 cm^{-1}. The narrow shoulder peak at 1094 cm^{-1} corresponds to the alcohol, C-OH stretching.

UV radiation increased the effectiveness of PVA breakdown by increasing the formation of free radicals. Meanwhile, the exposure duration of UV light had an effect on the functional groups of the adsorbent. The FTIR results of this analysis match those of Jiang *et al.* [19] work.

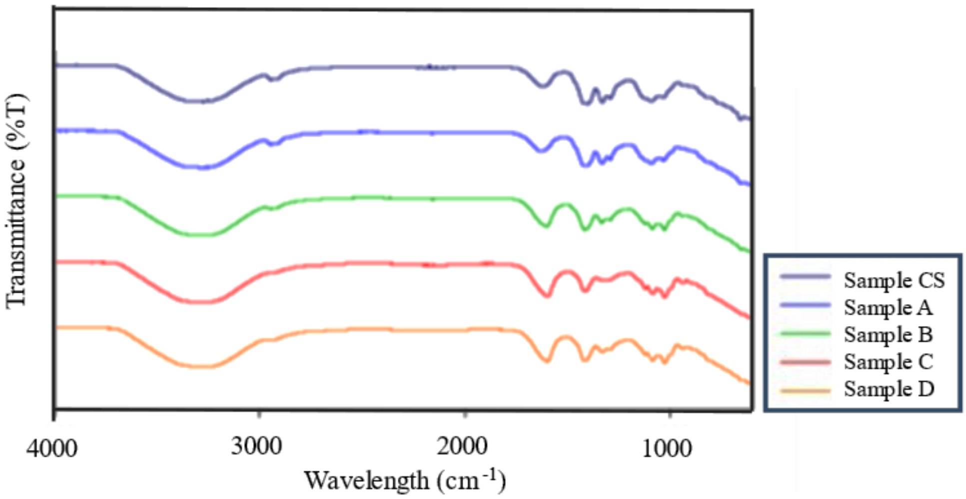

Fig. 2 *FTIR spectrum of PVA-alginate adsorbent beads*

3.3 Cr (VI) Removal Analysis

The study assessed the removal rate of PVA-alginate adsorbent beads in a 50 mg/L Cr (VI) solution. After 180 minutes of contact time, the concentration of Cr (VI) solution decreased from 50 mg/L to 14.88 mg/L, according to the removal rate of sample CS in Fig. 3. When the contact periods of adsorbent beads were extended, the removal rate rose.

In the case of sample A, the findings revealed that the concentration of Cr (VI) solution was lowered from 50 mg/L to 14.19 mg/L after 180 minutes of contact time. Furthermore, the maximum removal rate in the adsorption was 71.63% after 180 minutes. The removal rate for 30, 60, 90, 120, 150, and 180 minutes was 43.31, 46.40, 49.45, 50.39, 63.11, and 71.63%, respectively. The concentration of Cr (VI) solution in sample B decreased from 50 mg/L to 19.03 mg/L after 180 minutes of contact time. Moreover, after 180 minutes, the greatest removal rate in the adsorption was 61.95%. The removal rate was 30.64, 35.56, 39.60, 41.21, 44.97, and 61.95% after 30, 60, 90, 120, 150, and 180 minutes, respectively.

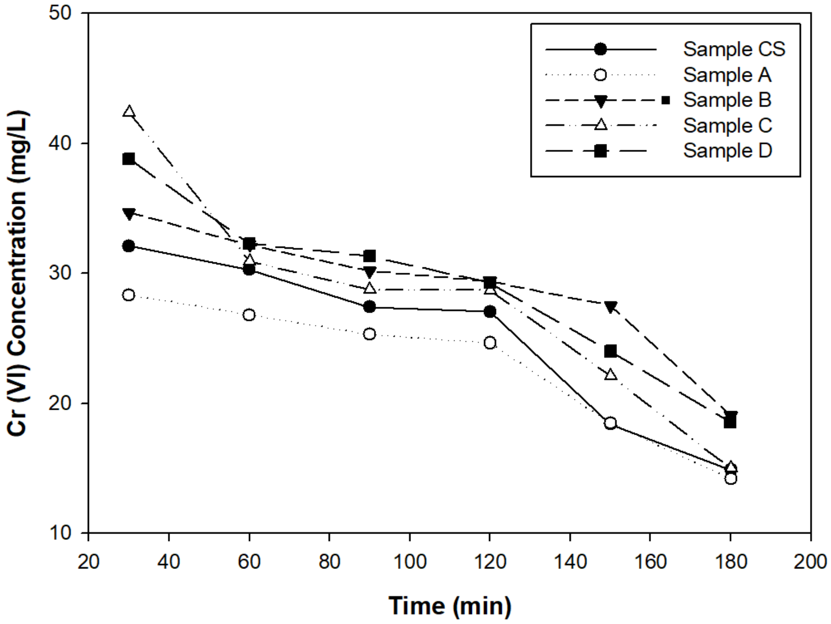

Fig. 3 *Cr (VI) removal performance of PVA-alginate adsorbent beads*

Meanwhile, after 180 minutes of contact time, the concentration of Cr (VI) solution dropped from 50 mg/L to 14.99 mg/L in sample C. Furthermore, the maximum removal rate for sample C was 70.02% after 180 minutes of treatment. The removal rate was 15.43, 38.11, 42.48, 42.59, 55.75, and 70.02% after 30, 60, 90, 120, 150, and 180 minutes of treatment, respectively. After 180 minutes of contact time, the concentration of Cr (VI) solution decreased from 50 mg/L to 18.53 mg/L in sample D. In addition, the maximum removal rate was obtained after 180 minutes of Cr (VI) solution treatment with sample D. The removal rate after 30, 60, 90, 120, 150, and 180 minutes was 22.35, 35.40, 37.28, 41.43, 51.99, and 62.94%, respectively.

In line with Fig. 3, the removal rate of Cr (VI) employing PVA-alginate adsorbent beads improved with increasing adsorption contact times. Sample A has the highest efficiency in removing Cr (VI) among other PVA-alginate absorbent beads samples, whereas sample B has the lowest. As a result, the adsorption rate of PVA-alginate beads was reduced when the UV radiation duration was more than 5 minutes prior to the Cr (VI) removal test. Consequently, UV light was identified as a factor influencing the Cr (VI) adsorption of the PVA-alginate adsorbent. To adsorb the Cr (VI), Li *et al.* [20] produced adsorbent beads out of PVA, SA, and polyaniline (PPS). After 48 hours of adsorption duration, the investigations revealed the greatest Cr (VI) removal rate of PPS adsorbent beads (99.67%) in the 50 mg/L Cr (VI) solution. The UV light enhances the hydrophilicity characteristic of the PVA-alginate adsorbent beads, which improves the adsorbent's adsorption performance.

The present study employed the same experimental conditions as earlier studies but only used a combination of PVA and SA to produce the adsorbent [20,21]. This study produced greater outcomes than prior studies because it was possible to decrease the period of Cr (VI) removal. Despite the fact that the removal performance is 71.63%, it is lower than in the previous study [22]. This work established the importance of UV light treatment in the production of PVA-alginate beads. It may have an effect on the adsorption rate of PVA-alginate adsorbents as well as the rate of Cr (VI) removal. An ideal UV irradiation time is 5 minutes (sample A) resulting in critical for increasing the PVA-alginate adsorption rate and accelerating the Cr (VI) removal rate.

3.4 Discussion

PVA is a synthetic polymer having a simple chemical structure and a pendant hydroxyl group. This hydroxyl group is important in Cr (VI) removal performance because the hydroxyl group (OH-) is capable of removing Cr (VI) ions by attach to Cr (VI) metal ions in solution through replacing hydrogen ions with metal ions [23]. The obtained results clearly show that PVA alginate beads have potential as an effective adsorbent, for removing Cr (VI). This excellent performance can be attributed primarily to the chemical structure and functional groups in both polyvinyl alcohol (PVA) and sodium alginate. SA, a derived linear from marine algae and certain bacteria consists of mannuronate (M) and guluronate (G) [24]. The presence of M residues plays important role in facilitating cation exchange with Cr (VI). These adjacent chains of M residues enable effective ionic crosslinking with various di- and trivalent metal ions associated with Cr (VI) [25-27].

Also, both PVA and sodium alginate possess hydroxyl groups, which play vital role in their capability to exchange cations. This characteristic enhances the efficiency of adsorbing Cr (VI). Cr (VI) exists in forms of oxyanions including chromate (CrO_4^{2-}), dichromate ($Cr_2O_7^{2-}$), chromic acid ($HCrO_4^-$), and chromium trioxide (CrO_3) [28]. It is believed that the interaction between PVA, sodium alginate and Cr (VI) facilitates the reduction of Cr (VI) through ion exchange processes resulting in the conversion of Cr (VI) into Cr (III). In the nutshell, the successful adsorption of Cr (VI) by PVA alginate beads can be primarily attributed to the to the intricate interplay of chemical structures and functional groups present in both PVA and sodium alginate. This interaction enables effective cation exchange reactions and the reduction of Cr (VI) to less toxic Cr (III) species.

It is important to note that, after a 5-minute exposure to UVA irradiation, the rate of Cr (VI) adsorption showed only a slight increase, rising from 70.24% to 71.63%. Despite this modest improvement in adsorption efficiency, it is essential to mention that UVA irradiation significantly simplifies the fabrication process of PVA-alginate beads. Our observations indicate that PVA-alginate gel, when not subjected to UV irradiation, displays elevated gel viscosity and poses challenges in achieving uniform spherical bead shapes during fabrication. Conversely, when the PVA-alginate gel is exposed to UV irradiation for a duration of 5 minutes, the viscosity of the gel markedly decreases, thus facilitating the bead fabrication process. Consequently, the resulting beads exhibit a higher degree of uniformity in both size and shape. In summary, while the increase in Cr (VI) adsorption efficiency after 5 minutes of UVA irradiation is relatively minor, the notable advantage lies in the simplification of the PVA-alginate bead fabrication process. This leads to the fabrication of beads that exhibit greater uniformity in both size and shape, thereby enhancing the practicality and reproducibility of the adsorption process.

Additionally, it is also observed that prolonged UV irradiation, lasting for 10 minutes or more, leads to a substantial reduction in the adsorption rate. This occurrence can be explained through the findings obtained from FTIR spectroscopy, which indicate that after prolonged exposure to UVA irradiation samples B, C and D exhibit the disappearance of C-H stretching vibrations. The presence of UVA irradiation, as a factor can trigger the degradation of compounds via photodegradation. As a consequence of this degradation process caused by UVA exposure diminished the C-H stretching vibrations [29].

As mentioned by Das et al. [30] the stretching vibrations of C-H bonds may have an impact, on the process of adsorption in relation to the presence of functional groups that contain C-H bonds, like alkyl groups (CH_3) and aromatic groups (benzene rings). These functional groups can act as anchor points for metal ions during adsorption utilizing bonding mechanisms such as coordination bonds, electrostatic interactions and hydrogen bonding. Additionally, the C-H bonds found in PVA alginate beads can undergo reactions with metal ions [31]. In this study we propose that Cr (VI) transforms into Cr (III) due to reactions initiated by the stretching vibrations of C-H bonds. Furthermore, these vibrations can cause changes in the conformation and orientation of molecules on the adsorbent surface thereby affecting the accessibility of adsorption sites for Cr (VI) ions [32].

Based on these findings it becomes evident why the rate, at which samples B, C and D absorb decreases significantly after being exposed to UV light for 10 minutes or more. This decrease can be mainly attributed to the disappearance of C H stretching vibrations, which have been shown to have an impact, on the adsorption process of Cr (VI).

4. Conclusion

The adsorption effectiveness of UV-irradiated PVA-alginate adsorbent beads for Cr (VI) removal was examined in this study. The size evaluation of PVA-alginate adsorbent beads revealed results that were consistent with past studies, with the average diameter of the beads ranging between 4.00 and 4.20 mm. The FTIR spectrum of PVA-alginate adsorbent beads reveals substantial hydroxyl (O-H) stretching vibrations of C-H stretching, O-H bending of adsorbed water, C-H bending, and O-H bending vibrations. These are the crucial hydroxyl and carboxyl functional groups for the effective performance of heavy metal pollutant adsorption in aqueous solution. After 180 minutes of contact time, the UV-Vis evidence indicated that PVA-alginate adsorbent beads obtained the maximum Cr (VI) removal rate of 71.63% for sample A (irradiate with UVA for 5 minutes). Hence, UV-irradiated PVA-alginate adsorbent beads have a reasonable adsorbent characteristic for removing Cr (VI) in water and may be completely used for heavy pollutant removal in water.

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Conflict of Interest

Authors declare that there is no conflict of interests regarding the publication of the paper.

Author Contribution

The authors confirm contribution to the paper as follows: ***study conception and design:*** *W.G. Lau, M.A. Selimin, T.C. Lee;* ***data collection:*** *W.G. Lau;* ***analysis and interpretation of results:*** *W.G. Lau, M.A. Selimin, T.C. Lee;* ***draft***

manuscript preparation: *W.G. Lau, M.A. Selimin, T.C. Lee. All authors reviewed the results and approved the final version of the manuscript.*

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