GRAFT COPOLYMERIZATION OF ACRYLONITRILE ONTO RECYCLED NEWSPAPER-CELLULOSE BASED ADSORBENT FOR HEAVY METALS REMOVAL

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DEDICATION

This thesis is dedicated to my father, who taught me that the best kind of knowledge to have is that which is learned for its own sake. It is also dedicated to my mother, who taught me that even the largest task can be accomplished if it is done one step at a time.

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ABSTRACT

Finding a good selection of solid sorbent becomes one of the greatest problems in the adsorption technique. In this study, adsorptive recycled newspaper (RNP)-based adsorbent was prepared with acrylonitrile initiated by ceric ammonium nitrate by graft copolymerization. The grafted recycled newspaper was used to remove heavy metals, such as chromium, Cr(VI); nickel, Ni(II); and copper, Cu(II), from aqueous solutions. The effects of grafting process of acrylonitrile onto the RNP, such as acrylonitrile concentration, ceric ammonium nitrate concentration, and reaction time, were investigated. The physicochemical properties of the grafted RNP was characterized by using Fourier transform infrared (FTIR), scanning electron microscope coupled with energy dispersive X-ray spectroscope (SEM-EDX), thermal gravimetric analysis (TGA), differential thermogravimetry (DTG), and X-ray diffraction (XRD) analysis. The adsorption capacity of the heavy metals was evaluated as a function of pH, initial concentrations of Cr(VI), Ni(II) and Cu(II), and contact time. Under pH 2 conditions, adsorption of Cr(VI) was optimal at 60 ppm for 360 minutes, with 15.1 mg/g of adsorption capacity. Meanwhile, for the adsorption of Cu(II), the optimal conditions were observed at pH 7 for 80 ppm over a period of 180 minutes, achieving 25.4 mg/g of adsorption capacity. The highest adsorption capacity for Ni(II) adsorption was 317.5 mg/g, which was achieved at pH 5 for a period of 60 minutes at 100 ppm of the initial Ni(II) concentration. Then, the equilibrium isotherms were conducted using Langmuir and Freundlich models. The findings showed that both Cr(VI) and Cu(II) were best fitted with the Langmuir model, which reflected the monolayer adsorption mechanism. Meanwhile, for the adsorption of Ni(II), the equilibrium coefficient was well suited with the Freundlich model, indicating that the adsorption had occurred based on the multilayer adsorption mechanism. The kinetic studies have been conducted by measuring the Pseudo-first-order and Pseudo-second-order for three types of heavy metals. The result showed that the adsorption kinetics of Cu(II) and Ni(II) were best fitted with the Pseudo-first-order, indicating the physisorption reaction. Meanwhile, the adsorption of Cr(VI) fitted with the Pseudo-second-order related to the chemisorption. This study concluded that the graft copolymerization of acrylonitrile on RNP improved efficiency by 52%, 57% and 56% for Cu(II), Ni(II) and Cr(VI) respectively relative to the pristine RNP.

ABSTRAK

Pemilihan penyerap pepejal yang baik menjadi salah satu masalah terbesar dalam teknik penjerapan. Di dalam kajian ini, penjerapan berasaskan kertas kitar semula (RNP) telah disediakan melalui pengkopolimeran cangkuk akrilonitril dengan kehadiran serium ammonia nitrat melalui pengubahsuaian kimia. RNP yang telah melalui proses pengkopolimeran cangkuk telah digunakan sebagai penjerap untuk mengasingkan sebatian logam berat seperti kromium, Cr(VI); nikel, Ni(II); dan tembaga, Cu(II) dari larutan berair. Kesan semasa proses pengkopolimeran cangkuk akrilonitril ke atas RNP telah dikaji termasuk kepekatan akrilonitril, kepekatan serium ammonia nitrat dan masa tindak balas. Sifat kimiafizikal RNP yang telah melalui proses pengkopolimeran cangkuk diperiksa menggunakan spektroskopi inframerah transformasi fourier (FTIR), pengimbas mikroskopi elektron dengan analisis x-ray penyebaran tenaga (SEM-EDX), analisis termogravimetrik (TGA), termogravimetrik derivatif (DTG) dan analisis difraksi x-ray (XRD). Kapasiti penjerapan logam berat dinilai berdasarkan pH, kepekatan awal Cr(VI), Cu(II) dan Ni(II) dan hubungan masa. Dalam keadaan pH 2, penjerapan C (VI) optimum pada 60 ppm selama 360 minit, dengan kapasiti penjerapan 15.1 mg/g. Sementara itu, untuk penjerapan Cu(II), keadaan optimum diperhatikan pada pH 7 untuk 80 ppm dalam jangka masa 180 minit, mencapai 25.4 mg/g kapasiti penjerapan. Kapasiti penjerapan tertinggi untuk penjerapan Ni(II) adalah 317.5 mg/g, yang dicapai pada pH 5 untuk jangka masa 60 minit pada 100 ppm kepekatan Ni(II) awal. Keseimbangan isoterma dijalankan menggunakan model Langmuir dan Freundlich. Hasil kajian menunjukkan bahawa kedua-dua Cr(VI) dan Cu(II) paling sesuai dengan model Langmuir, yang mencerminkan mekanisme penjerapan ekalapisan. Penjerapan Ni(II) pula menunjukkan keseimbangan isoterma Freundlich di mana proses penjerapan berlaku secara mekanisma multilapisan. Kajian secara kinetik telah dijalankan melalui Pseudo peringkat-pertama dan Pseudo peringkat-kedua untuk menyesuaikan data penjerapan di dalam kajian kinetik. Keputusan kajian menunjukkan penjerapan kinetik untuk Cu(II) dan Ni(II) adalah lebih tepat diterangkan dengan Pseudo peringkat-pertama masing-masing yang menunjukkan tindakbalas secara penjerapan fizikal. Manakala, penjerapan Cr(VI) lebih tepat diterangkan dengan Pseudo peringkat-kedua yang dikatkan dengan tindakbalas penjerapan kimia. Melalui kajian ini dapat disimpulkan bahawa pengkopolimeran cangkuk akrilonitril ke atas RNP telah meningkatkan kecekapan penyingkiran sebanyak 52%, 57% dan 56% untuk Cu(II), Ni(II) dan Cr(VI) berbanding dengan RNP asli.

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LIST OF ABBREVIATIONS

AAS	Atomic adsorption spectroscopy
ATR	Attenuated total reflection
Cr(VI)	Chromium(VI)
Cu(II)	Copper(II)
DTG	Differential gravimetric analysis
EDX	Energy dispersive X-ray
FTIR	Fourier Transform Infrared
GE	Grafting Efficiency
GP	Grafting Percentage
Ni(II)	Nickel(II)
RNP	Recycled Newspaper
SEM	Scanning electron microscopy
t-RNP	Treated Recycled Newspaper
TGA	Thermal gravimetric analysis
XRD	X-ray Diffraction

LIST OF SYMBOLS

C_0	Initial concentration of metal ions
C_e	Final concentration of metal ions
k_1	Rate constant of Pseudo-first-order model (min^{-1})
k_2	Rate constant of Pseudo-second-order model (min^{-1})
K_F	Equilibrium adsorption constant for Freundlich model (Lg^{-1})
K_L	Equilibrium adsorption constant for Freundlich model (Lmg^{-1})
n	Heterogeneity factor in Frreunlich isotherm
q_e	Equilibrium adsorption capacity (mg/g)
$q_{e,cal}$	Calculated equilibrium adsorption capacity (mg/g)
$q_{e,exp}$	Experimental equilibrium adsorption capacity (mg/g)
$q_{e,max}$	Maximum adsorption capacity (mg/g)
q_t	Adsorption capacity at time, t (mg/g)
R_L	Separation factor in Langmuir isotherm
t	Time
W_1	Weight of original RP (g)
W_2	Weight of grafted-RP after copolymerization (g)
W_3	Weight of grafted-RP after copolymerization and extraction (g)

CHAPTER 1

INTRODUCTION

1.1 Research Background

The outcome from the industrialisation and urbanisation contributes to environmental problems, especially those related to the release of heavy metals. Common industries that discharge these heavy metals are industrials fields of fertiliser, mining operation, metal plating facilities, paper, et cetera [1]. Effluents, such as Cu, Cr and Ni, not only harm the aquatic organisms, but in others ways, have a high capability to accumulate in food chain, since these heavy metals are non-biodegradable. These heavy metals also possess high toxic content, leading to its ability to give a greater threat to the human health and to the environment [2]. Although plants at the micronutrient level need many of the heavy metals, higher concentration will yield toxic effects. Common heavy metal such as Cr is positioned at the top of notorious effluent list, since Cr is categorised in Group 1 by the International Agency for Research on Cancer (IARC) [3]. The main industries that mostly discharge hexavalent chromium, Cr(VI), include metallurgy, leather tanning, electroplating, and wood preservation industries [4], [5].

Generally, Cr can be found in three common types or forms of oxidation states in its compound, which are Cr^{2+}, Cr^{3+} and Cr^{6+}. Both Cr^{3+} and $Cr^{6+}$are commonly detected in Cr compounds. Meanwhile, Cr^{+}, Cr^{4+} and Cr^{5+} are rarely noticed in its compounds. The toxicity of Cr(VI) is higher compared to other forms of Cr. It can directly penetrate into the human body via skin contact, inhalation or oral intake. As the Cr enters the human body, it will be directly transferred to parts of the human body via blood, which can cause damage to the DNA due to its carcinogenetic properties [6]. The allowable concentration of Cr species in the surface or potable water must be less than 0.05 mg/L and obliged.

Meanwhile, the presence of the Ni that are majorly detected in the steel production industries, non-ferrous alloy production, mineral processing, electroplating and batteries manufacture are known to be toxic and too carcinogenic if it is present at very high levels. In human body, the optimum dosage of the Ni(II) ions is necessary as it involved in the metabolism of human protein and regulation of hormone. The purpose of the Ni also responsible in stimulating hematopoietic function and increase the rate of the regeneration of red blood cell [7]. Nevertheless, uncontrollable intake of the nickel boosts the abnormal effect towards human metabolism as it will irritate respiratory system, causes tissue damage, gene toxicity and increase the risk of cancer [8]. According to the World Health Organization (WHO), the maximum acceptance limit of nickel is 0.02 mg/L in potable water [9]. In addition, an excessive exposure of Cu(II) initiates the production of reactive free oxygen in human blood systems which will give harmful effects towards protein, lipids and DNA. The effects of the over exposure of Cu also can damage the marine ecosystems as it will result the harm towards gills, liver, the nervous systems and changing the sexual life of fishes [10]. Due to deleterious effect, the allowable limit of Cu in drinking water and industrial effluents is restricted at 2.0 mg/L and 1.3 mg/L, respectively [11].

Due to its carcinogenicity to humans, the removal of these heavy metals is extremely vital before they are expelled into the aquatic system. Since the nature of these heavy metals is harmful towards the environment, including to the human health and aquatic lives, numerous progresses on conventional treatment technologies have been made. Technologies that are generally applied to treat heavy metals are chemical precipitation [12]–[14], membrane separation [15], adsorption [16]–[18], and filtration [9]. Among these approaches, the adsorption technology is noticed as a promising and most effective method because of its simple operation and low cost [19], [20]. The adsorption technique offers high efficiency and flexibility due to the availability of various adsorbents, effectiveness in the removal of organic and inorganic pollutants, and insensitivity to toxic substances [21], [22].

Previously, most of the adsorbents are carbon-based materials. As cited in most of the literature, the application of carbon-based materials, such as activated carbon, carbon nanotubes, aerogels, and graphene, have undoubtedly been proven as good adsorbents. Although the capability of carbon-based materials is excellent for the removal of heavy metals, these materials possess some disadvantages because they have low density of functional groups and they are high in their operational cost, thus, limiting the application of carbon-based materials in real industrial fields [23]. As an excellent alternative, applications related to heavy metals removal is narrowly focused on the usage of biopolymers as bio sorbents.

Low-cost adsorbent with high adsorption capacity is still under development in order to diminish the usage of adsorbent dosage and reduce post-adsorption disposal problems. Moreover, environmental sustainability, industrial ecology, eco-efficiency, and green chemistry have become another crucial point to pursue the development of new materials and product from renewable resources. Naturally derived and renewable materials have become the main focus among researchers in many applications due to its environmentally friendly and naturally available features. Cellulose is perceived as a capable candidate in the removal of environmental pollutants [14], [24]. The adaptability of cellulose for various applications is due to its low cost, ease of availability, biocompatibility, derivable properties, and high content of hydroxyl group in the chain of the cellulose structure that can be modified for special or specific application [25]–[27]. Due to these advantages, cellulose has been majorly applied in wastewater treatment, reinforcing composite materials and additionally, in bio-derived and bio-inspired materials [20]. Previously, there has been a study on the preparation of cellulose from various natural resources, such as sisal fibre, cotton linen, bamboo, populus fibre, wood pulp, banana peels, cassia tora gum, pineapple leaves, and torch ginger [19], [28]–[31].

In Malaysia, the production of paper is over 1 million tons per year. There are 67 paper mills producing more than 50 tons per day, ranging from traditional, recycled and specific type of papers. Among these listed mills, six are producing less than 10,000 tons per year, nine are producing between 11,000 to 80,000 tons per year and only six mills can produce from 100,000 up to 300,000 tons per year [32]. Although cellulose and its derivative have been used in heavy metal sorption, some limitations were found due to its low stability and poor interaction in water.

However, improvements can be made by employing modifications via cross-linking, surfactant adsorption and grafting methods. The adsorption capacity of modified adsorbents is believed to be much better compared to the unmodified ones. This is due to the presence of active groups that are capable of improving the ability of the adsorbent to attract the metal ions. Among all modifications of cellulosic materials, grafting copolymerisation is most effective compared to other methods due to its chemical resistance and radiation stability characteristics and low operating cost [19], [33].

Grafting process will introduce branches at the cellulosic backbones without destroying the original properties of the cellulose. Chemical modification by graft copolymerisation of monomer onto the cellulosic biofibres offers a large number of functional groups, which will enhance the adsorption capacity of the cellulosic fibres [2]. The physical and chemicals properties, as well as the functionality of the adsorbent can be improved by introducing acrylonitrile as the monomer. The monomer was covalently grafted onto the cellulose backbones. The process of applying chemical initiation grafting via direct oxidation of ceric ions (Ce^{4+}) has been chosen as a method for graft copolymerisation due to its wide range of monomers, tolerance with water and operational simplicity. Ce^{4+} ions were used in this study as initiator to initiate polymerisation, since it is proven to shorten the reaction time while having a high rate of grafting and ease of application [34], [35].

1.2 Problem Statement

The adsorption method is a promising technique among all the available approaches in heavy metal removal treatment due to its high efficiency, low cost and simplicity in design and operation. A good selection of solid sorbent becomes one of the greatest problems in the adsorption technique. The properties of the adsorbent should have characteristics of adsorbing maximum pollutant economically and biodegradable, and the sources are copious in nature. These factors have increased the interest of many researchers to explore for alternative adsorbent that are efficient and low in cost. In recent years, the “green” comprehensive utilisation of cellulose resources has drawn much attention from the government and researchers. Cellulose is unquestionably the most abundant, naturally occurring, reproducible organic compound; and it will become the main chemical resource in the future. Thus, in this study, cellulose is selected due to its abundance in nature and being an effective adsorbent for handling heavy metals.

Cellulose can be sourced from many kinds of origins, including plant sources such as pineapple, banana stalk, bamboo, wheat straw, potato peels, et cetera. Among all cellulose derivatives presented, recycled newspaper (RNP) is chosen as a source of cellulose to remove heavy metals, since it possesses the capability to adsorb, and is renewable, abundant and cheaper in Malaysia. This would be an environmentally friendly approach, since tons of RNP is discharged daily in Malaysia. Therefore, it is important to access the feasibility of RNP as a source of cellulose in the preparation of green and low-cost adsorbent. The RNP consists of more than 60 wt% of cellulose, where the cellulose is the main constituent, having low density and exhibiting better processing flexibility. The RNP has potential functional groups such as hydroxyl, which has great affinity towards metal ions. However, RNP show low stability and have a poor interaction in water which will limit the performance as the adsorbent.

Based on the literatures, the adsorption capacities of chemically modified cellulose-based adsorbent are much higher compared to that of pristine cellulose-based adsorbent. Chemical modification through graft copolymerisation is covalently introduced onto the cellulose backbones without destroying the whole original structure of the cellulose. Grafting is the most common approach used in modifying cellulose to produce grafted adsorbent for the removal of heavy metals from aqueous solution. Through this research, the grafting copolymerisation of acrylonitrile by using ceric ammonium nitrate as initiator onto the cellulose-based adsorbent sourced from RNP is believed to have maximum adsorption capacity with a high potential to become a biodegradable adsorbent.

1.3 Objectives of Study

The main research objectives of this analysis are listed as follows:

1. To graft cellulose sourced from RNP with acrylonitrile, initiated by ceric ammonium nitrate via chemical modification.

2. To investigate the effect of reaction conditions (e.g., monomer concentration, initiator concentration and reaction time) on the physicochemical properties of the RNP-based adsorbent.

3. To assess the effect of adsorption conditions (e.g., contact time, initial concentration of heavy metal concentration, and pH of the solution) towards the adsorption capacity of RNP-based adsorbent for the removal of heavy metal ions.

1.4 Scope of Study

In order to achieve the listed objectives, the scope of study has been drawn as follows:

1. Extracting cellulose from RNP via 5 wt% alkali and 2% w/v $NaClO_2$ as a bleaching treatment process to obtain white pulp samples;

2. Grafting the RNP with acrylonitrile monomer at different monomer concentrations (0.5g, 1.0g, 1.5 g, and 2.0g), ceric ammonium nitrate concentrations (0.4 g, 0.8 g, 1.2 g, and 1.5 g), and reaction times (30 min, 45 min, 60 min, and 75 min);

3. Characterising the grafted RNP via the Fourier transform infrared spectroscopy (FTIR), scanning electron microscopy (SEM) coupled with

energy dispersive X-ray spectrometer (EDX), X-ray diffraction (XRD), thermal gravimetric analysis (TGA), and differential gravimetric analysis (DTG);

4. Evaluating the adsorption capacity at different operation conditions of Cr(VI), Cu(II) and Ni(II): initial heavy metal concentrations (20–100 mg/L), pH of the solution (pH 2–7) and contact time (30–360 min) via batch adsorption experiments with fixed dosage of grafted RNP adsorbent of 0.02 g;

5. Evaluating the adsorption kinetics study by analysing the obtained results of the batch adsorption experiments with two types of models, such as pseudo-first-order and pseudo-second-order models; and

6. Determining the adsorption isotherms for the adsorption experiment between the grafted RNP with heavy metal ions via adsorption isotherms model of Langmuir and Freundlich.

1.5 Significance of Study

The hazardousness of the heavy metals containing Cr(VI), Ni(II) and Cu(II) effluent have led to the critical eco-system problems. Therefore, the removals of these effluents become the main concern in recent years by many researchers, and many ways have been loomed for minimizing these problems. The adsorption have been advocated and selected since it is straightforward and own great ability, and finally the feasibility of many kinds of adsorbents. The selection of adsorbent to be implemented in the adsorption process will improve the capability of the adsorbent especially in terms of the adsorption capacity to provide a minimum content of metals effluent before being discharged to the water systems. Relying on the needs of effective and capability of the adsorbent to remove effluent having heavy metals, this proposal have been carried out.

The alternative way to conserve the environment becomes pivotal. The fabrication of cellulose microfiber sourced from RNP seems to be green ways for environmental issues. The utilization of RNP as the raw materials for the adsorbent is reviewed as cut-cost and economically to industrial field. The preparation of this cellulose-based adsorbent that utilized recycled supplies is rated as “green” process.

As a result, it will maintain the safe mood environment since the adsorbent derived from recycled materials. The optimum parameter for the grafting process will be determined throughout this research. The cost for this adsorbent itself was assumed to be low and only needed to purchase the monomer in order to enhance the adsorption process of the heavy metals. Therefore, this research can be categorized as economic approach. In future, this research can be a reference for a commercial and industrial production and application. This study is expected to be a solver to the problem related to the heavy metals issues.

1.6 Thesis Outline

This thesis consists of five main chapters, all of which represent the sequence of this research work. Chapter 1 briefly explains the overview related to the research background, problem statement, objectives, as well as the scope and significance of the study. Chapter 2 details on recent publications and literature review on the removal of heavy metals using cellulose-based adsorbent and modified-based cellulose adsorbent. This chapter also reviews the preferable methods for the removal of hazardous metal ions, including the advantages and disadvantages. The modification of cellulose-based adsorbent sourced from the RNP and its application are also discussed in Chapter 2. Next, Chapter 3 covers the research methodology, including the extraction of the cellulose, grafting technique of acrylonitrile onto RNP, characterisations of the adsorbent, and experimental studies for the adsorption of the three types of heavy metals, such as Cr(VI), Ni(II) and Cu(II). In brief, Chapter 4 clarifies on the results and discussion regarding the research objectives. All outcomes from the chapters are concluded in Chapter 5, including the future work and recommendations.

REFERENCES

1. F. Fu and Q. Wang, "Removal of heavy metal ions from wastewaters: A review," *J. Environ. Manage.*, vol. 92, no. 3, pp. 407–418, 2011.
2. A. S. Singha and A. Guleria, "Chemical modification of cellulosic biopolymer and its use in removal of heavy metal ions from wastewater," *Int. J. Biol. Macromol.*, vol. 67, pp. 409–417, 2014.
3. M. H. Dehghani, D. Sanaei, I. Ali, and A. Bhatnagar, "Removal of chromium(VI) from aqueous solution using treated waste newspaper as a low-cost adsorbent: Kinetic modeling and isotherm studies," *J. Mol. Liq.*, vol. 215, pp. 671–679, 2016.
4. S. Zhang, H. Lyu, J. Tang, B. Song, M. Zhen, and X. Liu, "A novel biochar supported CMC stabilized nano zero-valent iron composite for hexavalent chromium removal from water," *Chemosphere*, vol. 217, pp. 686–694, 2019.
5. Y. Zhang, X. Jiao, N. Liu, J. Lv, and Y. Yang, "Enhanced removal of aqueous Cr(VI) by a green synthesized nanoscale zero-valent iron supported on oak wood biochar," *Chemosphere*, vol. 245, no. Vi, 2020.
6. Y. Shi, T. Zhang, H. Ren, A. Kruse, and R. Cui, "Polyethylene imine modified hydrochar adsorption for chromium (VI) and nickel (II) removal from aqueous solution," *Bioresour. Technol.*, vol. 247, no. September 2017, pp. 370–379.
7. J. He, H. Shang, X. Zhang, and X. Sun, "Synthesis and application of ion imprinting polymer coated magnetic multi-walled carbon nanotubes for selective adsorption of nickel ion," *Appl. Surf. Sci.*, vol. 428, pp. 110–117.
8. H. Guo *et al.*, "Nickel chloride (NiCl2) in hepatic toxicity: Apoptosis, G2/M cell cycle arrest and inflammatory response," *Aging (Albany. NY).*, vol. 8, no. 11, pp. 3009–3027, 2016.
9. A. A. Palliparambil, S. Bethireddy, P. Hariprasad Reddy, and S. B. Rajulapati, "Removal of nickel by adsorption from aqueous solution of cellulose acetate-zero valent iron nanoparticle mixed matrix membrane," *Indian J. Sci. Technol.*, vol. 9, no. 41, 2016.
10. M. A. Hossain, H. H. Ngo, W. S. Guo, and T. V Nguyen, "Removal of Copper from Water by Adsorption onto Banana Peel as Bioadsorbent," pp. 227–234,

2012.
11. Y. H. Teow, L. M. Kam, and A. W. Mohammad, “Synthesis of cellulose hydrogel for copper (II) ions adsorption,” *J. Environ. Chem. Eng.*, vol. 6, no. 4, pp. 4588–4597, 2018.
12. Q. Chen, Y. Yao, X. Li, J. Lu, J. Zhou, and Z. Huang, “Comparison of heavy metal removals from aqueous solutions by chemical precipitation and characteristics of precipitates,” *J. Water Process Eng.*, vol. 26, no. November, pp. 289–300, 2018.
13. M. A. Barakat, “New trends in removing heavy metals from industrial wastewater,” *Arab. J. Chem.*, vol. 4, no. 4, pp. 361–377, 2011.
14. X. Sun *et al.*, “Amino-functionalized magnetic cellulose nanocomposite as adsorbent for removal of Cr(VI): Synthesis and adsorption studies,” *Chem. Eng. J.*, vol. 241, pp. 175–183, 2014.
15. M. R. Adam *et al.*, *Adsorptive Membranes for Heavy Metals Removal From Water*. Elsevier Inc., 2019.
16. J. Li, L. Cao, Y. Yuan, R. Wang, Y. Wen, and J. Man, “Comparative study for microcystin-LR sorption onto biochars produced from various plant- and animal-wastes at different pyrolysis temperatures: Influencing mechanisms of biochar properties,” *Bioresour. Technol.*, vol. 247, no. July 2017, pp. 794–803, 2018.
17. J. Xiong, D. Zhang, H. Lin, and Y. Chen, “Amphiprotic cellulose mediated graphene oxide magnetic aerogels for water remediation,” *Chem. Eng. J.*, vol. 400, no. April, p. 125890, 2020.
18. H. Pang *et al.*, “Adsorptive and reductive removal of U(VI) by Dictyophora indusiate-derived biochar supported sulfide NZVI from wastewater,” *Chem. Eng. J.*, vol. 366, no. February, pp. 368–377, 2019.
19. A. Ali and K. Saeed, “Phenol removal from aqueous medium using chemically modified banana peels as low-cost adsorbent,” *Desalin. Water Treat.*, vol. 57, no. 24, pp. 11242–11254, 2016.
20. M. Li, Y. Gong, A. Lyu, Y. Liu, and H. Zhang, “The applications of populus fiber in removal of Cr (VI) from aqueous solution,” *Appl. Surf. Sci.*, vol. 383, pp. 133–141, 2016.
21. S. Ghorai, A. Sarkar, M. Raoufi, A. B. Panda, H. Schönherr, and S. Pal, “Enhanced removal of methylene blue and methyl violet dyes from aqueous

solution using a nanocomposite of hydrolyzed polyacrylamide grafted xanthan gum and incorporated nanosilica," *ACS Appl. Mater. Interfaces*, vol. 6, no. 7, pp. 4766–4777, 2014.

22. M. Zhang, Q. Yao, C. Lu, Z. Li, and W. Wang, "Layered double hydroxide-carbon dot composite: High-performance adsorbent for removal of anionic organic dye," *ACS Appl. Mater. Interfaces*, vol. 6, no. 22, pp. 20225–20233, 2014.
23. A. Kundu, G. Redzwan, J. N. Sahu, S. Mukherjee, B. Sen Gupta, and M. A. Hashim, "Hexavalent Chromium Adsorption by a Novel Activated Carbon Prepared by Microwave Activation," *BioResources*, vol. 9, no. 1, pp. 1498–1518, 2014.
24. S. Hokkanen, E. Repo, L. J. Westholm, S. Lou, T. Sainio, and M. Sillanpää, "Adsorption of Ni^{2+}, Cd^{2+}, PO_4^{3-} and NO_3^- from aqueous solutions by nanostructured microfibrillated cellulose modified with carbonated hydroxyapatite," *Chem. Eng. J.*, vol. 252, pp. 64–74, 2014.
25. V. K. Gupta, S. Agarwal, P. Singh, and D. Pathania, "Acrylic acid grafted cellulosic Luffa cylindrical fiber for the removal of dye and metal ions," *Carbohydr. Polym.*, vol. 98, no. 1, pp. 1214–1221, 2013.
26. S. H. Hasan, K. K. Singh, O. Prakash, M. Talat, and Y. S. Ho, "Removal of Cr(VI) from aqueous solutions using agricultural waste 'maize bran,'" *J. Hazard. Mater.*, vol. 152, no. 1, pp. 356–365, 2008.
27. M. A. Mohamed, W. N. W. Salleh, J. Jaafar, A. F. Ismail, M. Abd. Mutalib, and S. M. Jamil, "Feasibility of recycled newspaper as cellulose source for regenerated cellulose membrane fabrication," *J. Appl. Polym. Sci.*, vol. 132, no. 43, pp. 1–10, 2015.
28. T. Hajeeth, K. Vijayalakshmi, T. Gomathi, and P. N. Sudha, "Removal of Cu(II) and Ni(II) using cellulose extracted from sisal fiber and cellulose-g-acrylic acid copolymer," *Int. J. Biol. Macromol.*, vol. 62, pp. 59–65, 2013.
29. R. Khullar, V. K. Varshney, S. Naithani, and P. L. Soni, "Grafting of acrylonitrile onto cellulosic material derived from bamboo (Dendrocalamus strictus)," *Express Polym. Lett.*, vol. 2, no. 1, pp. 12–18, 2008.
30. R. M. dos Santos, W. P. Flauzino Neto, H. A. Silvério, D. F. Martins, N. O. Dantas, and D. Pasquini, "Cellulose nanocrystals from pineapple leaf, a new approach for the reuse of this agro-waste," *Ind. Crops Prod.*, vol. 50, pp. 707–

714, 2013.

31. B. R. Sharma, V. Kumar, and P. L. Soni, "Graft copolymerization of acrylonitrile onto Cassia tora gum with ceric ammonium nitrate-nitric acid as a redox initiator," *J. Appl. Polym. Sci.*, vol. 90, no. 1, pp. 129–136, 2003.
32. J. Roda, "Malaysia," no. December 2006, 2016.
33. F. E. Okieimen, C. E. Sogbaike, and J. E. Ebhoaye, "Removal of cadmium and copper ions from aqueous solution with cellulose graft copolymers," *Sep. Purif. Technol.*, vol. 44, no. 1, pp. 85–89, 2005.
34. N. A. Rosli, I. Ahmad, I. Abdullah, F. H. Anuar, and F. Mohamed, "Hydrophobic modification of cellulose isolated from Agave angustifolia fibre by graft copolymerisation using methyl methacrylate," *Carbohydr. Polym.*, vol. 125, pp. 69–75, 2015.
35. M. L. Rahman, N. Rohani, N. Mustapa, and M. M. Yusoff, "Synthesis of polyamidoxime chelating ligand from polymer-grafted corn-cob cellulose for metal extraction," *J. Appl. Polym. Sci.*, vol. 131, no. 19, pp. 1–8, 2014.
36. D. W. O'Connell, C. Birkinshaw, and T. F. O'Dwyer, "Heavy metal adsorbents prepared from the modification of cellulose: A review," *Bioresour. Technol.*, vol. 99, no. 15, pp. 6709–6724, 2008.
37. S. Hokkanen, A. Bhatnagar, and M. Sillanpää, "A review on modification methods to cellulose-based adsorbents to improve adsorption capacity," *Water Res.*, vol. 91, pp. 156–173, 2016.
38. T. Riaz *et al.*, "Synthesis and characterization of polyurethane-cellulose acetate blend membrane for chromium (VI) removal," *Carbohydr. Polym.*, vol. 153, pp. 582–591, 2016.
39. S. Wan *et al.*, "Accelerated antimony and copper removal by manganese oxide embedded in biochar with enlarged pore structure," *Chem. Eng. J.*, vol. 402, no. May, p. 126021, 2020.
40. H. Gong, J. Chi, Z. Ding, F. Zhang, and J. Huang, "Removal of lead from two polluted soils by magnetic wheat straw biochars," *Ecotoxicol. Environ. Saf.*, vol. 205, no. July, p. 111132, 2020.
41. J. H. Kwak *et al.*, "Biochar properties and lead(II) adsorption capacity depend on feedstock type, pyrolysis temperature, and steam activation," *Chemosphere*, vol. 231, pp. 393–404, 2019.

42. S. Gupta, S. Sireesha, I. Sreedhar, C. M. Patel, and K. L. Anitha, "Latest trends in heavy metal removal from wastewater by biochar based sorbents," *J. Water Process Eng.*, vol. 38, no. October, p. 101561, 2020.
43. D. V. Cuong, P. Wu, L. Chen, and C. Hou, "Active MnO2/biochar composite for efficient As(III) removal: Insight into the mechanisms of redox transformation and adsorption," *Water Res.*, no. Iii, p. 116495, 2020.
44. M. Imran *et al.*, "Synthesis, characterization and application of novel MnO and CuO impregnated biochar composites to sequester arsenic (As) from water: Modeling, thermodynamics and reusability," *J. Hazard. Mater.*, vol. 401, no. June, p. 123338, 2021.
45. Y. Liu, H. Ma, B. Liu, B. S. Hsiao, and B. Chu, "High-performance nanofibrous membrane for removal of Cr(VI) from contaminated water," *J. Plast. Film Sheeting*, vol. 31, no. 4, pp. 379–400, 2015.
46. T. Hajeeth, P. N. Sudha, K. Vijayalakshmi, and T. Gomathi, "Sorption studies on Cr (VI) removal from aqueous solution using cellulose grafted with acrylonitrile monomer," *Int. J. Biol. Macromol.*, vol. 66, pp. 295–301, 2014.
47. S. Maleki and A. Karimi-Jashni, "Effect of ball milling process on the structure of local clay and its adsorption performance for Ni(II) removal," *Appl. Clay Sci.*, vol. 137, pp. 213–224, 2017.
48. Z. Huang *et al.*, "Modified cellulose by polyethyleneimine and ethylenediamine with induced Cu(II) and Pb(II) adsorption potentialities," *Carbohydr. Polym.*, vol. 202, no. April, pp. 470–478, 2018.
49. R. Z. Wang *et al.*, "Synergistic removal of copper and tetracycline from aqueous solution by steam-activated bamboo-derived biochar," *J. Hazard. Mater.*, vol. 384, no. October 2019, p. 121470, 2020.
50. S. Tangtubtim and S. Saikrasun, "Adsorption behavior of polyethyleneimine-carbamate linked pineapple leaf fiber for Cr(VI) removal," *Appl. Surf. Sci.*, vol. 467–468, no. July 2018, pp. 596–607, 2019.
51. M. M. Nasef and O. Güven, "Radiation-grafted copolymers for separation and purification purposes: Status, challenges and future directions," *Prog. Polym. Sci.*, vol. 37, no. 12, pp. 1597–1656, 2012.
52. A. Martín-Domínguez *et al.*, "Chromium removal from drinking water by redox-assisted coagulation: Chemical versus electrocoagulation," *Sep. Purif. Technol.*, vol. 200, no. February, pp. 266–272, 2018.

53. H. Polat and D. Erdogan, “Heavy metal removal from waste waters by ion flotation,” *J. Hazard. Mater.*, vol. 148, no. 1–2, pp. 267–273, 2007.
54. P. Gao, X. Chen, F. Shen, and G. Chen, “Removal of chromium(VI) from wastewater by combined electrocoagulation- electroflotation without a filter,” *Sep. Purif. Technol.*, vol. 43, no. 2, pp. 117–123, 2005.
55. Y. A. El-Taweel, E. M. Nassef, I. Elkheriany, and D. Sayed, “Removal of Cr(VI) ions from waste water by electrocoagulation using iron electrode,” *Egypt. J. Pet.*, vol. 24, no. 2, pp. 183–192, 2015.
56. R. Dobrowolski, A. Szcześ, M. Czemierska, and A. Jarosz-Wikołazka, “Studies of cadmium(II), lead(II), nickel(II), cobalt(II) and chromium(VI) sorption on extracellular polymeric substances produced by Rhodococcus opacus and Rhodococcus rhodochrous,” *Bioresour. Technol.*, vol. 225, pp. 113–120, 2017.
57. T. Sathvika, Manasi, V. Rajesh, and N. Rajesh, “Adsorption of chromium supported with various column modelling studies through the synergistic influence of Aspergillus and cellulose,” *J. Environ. Chem. Eng.*, vol. 4, no. 3, pp. 3193–3204, 2016.
58. D. Duranoĝlu, A. W. Trochimczuk, and U. Beker, “Kinetics and thermodynamics of hexavalent chromium adsorption onto activated carbon derived from acrylonitrile-divinylbenzene copolymer,” *Chem. Eng. J.*, vol. 187, pp. 193–202, 2012.
59. S. O. Owalude and A. C. Tella, “Removal of hexavalent chromium from aqueous solutions by adsorption on modified groundnut hull,” *Beni-Suëf Univ. J. Basic Appl. Sci.*, vol. 5, no. 4, pp. 377–388, 2016.
60. M. K. Rai *et al.*, “Removal of hexavalent chromium Cr (VI) using activated carbon prepared from mango kernel activated with H3PO4,” *Resour. Technol.*, vol. 2, pp. S63–S70, 2016.
61. A. B. Wassie and V. C. Srivastava, “Teff straw characterization and utilization for chromium removal from wastewater: Kinetics, isotherm and thermodynamic modelling,” *J. Environ. Chem. Eng.*, vol. 4, no. 1, pp. 1117–1125, 2016.
62. S. Habibi, A. Nematollahzadeh, and S. A. Mousavi, “Nano-scale modification of polysulfone membrane matrix and the surface for the separation of chromium ions from water,” *Chem. Eng. J.*, vol. 267, pp. 306–316, 2015.

63. S. Z. N. Ahmad, W. N. Wan Salleh, A. F. Ismail, N. Yusof, M. Z. Mohd Yusop, and F. Aziz, "Adsorptive removal of heavy metal ions using graphene-based nanomaterials: Toxicity, roles of functional groups and mechanisms," *Chemosphere*, vol. 248, p. 126008, 2020.
64. V. K. Gupta and Suhas, "Application of low-cost adsorbents for dye removal - A review," *J. Environ. Manage.*, vol. 90, no. 8, pp. 2313–2342, 2009.
65. A. E. Burakov *et al.*, "Adsorption of heavy metals on conventional and nanostructured materials for wastewater treatment purposes: A review," *Ecotoxicol. Environ. Saf*, vol. 148, no. November 2017, pp. 702–712, 2018.
66. M. Lu, X. H. Guan, X. H. Xu, and D. Z. Wei, "Characteristic and mechanism of Cr(VI) adsorption by ammonium sulfamate-bacterial cellulose in aqueous solutions," *Chinese Chem. Lett.*, vol. 24, no. 3, pp. 253–256, 2013.
67. C. Li, Y. Zhang, J. Peng, H. Wu, J. Li, and M. Zhai, "Adsorption of Cr(VI) using cellulose microsphere-based adsorbent prepared by radiation-induced grafting," *Radiat. Phys. Chem.*, vol. 81, no. 8, pp. 967–970, 2012.
68. P. Devi and A. K. Saroha, "Utilization of sludge based adsorbents for the removal of various pollutants: A review," *Sci. Total Environ.*, vol. 578, pp. 16–33, 2017.
69. S. Pourfadakari, S. Jorfi, M. Ahmadi, and A. Takdastan, "Experimental data on adsorption of Cr(VI) from aqueous solution using nanosized cellulose fibers obtained from rice husk," *Data Br.*, vol. 15, pp. 887–895, 2017.
70. J. R. Memon, S. Q. Memon, M. I. Bhanger, A. El-Turki, K. R. Hallam, and G. C. Allen, "Banana peel: A green and economical sorbent for the selective removal of Cr(VI) from industrial wastewater," *Colloids Surfaces B Biointerfaces*, vol. 70, no. 2, pp. 232–237, 2009.
71. P. Kumar and M. S. Chauhan, "Adsorption of chromium (VI) from the synthetic aqueous solution using chemically modified dried water hyacinth roots," *J. Environ. Chem. Eng.*, vol. 7, no. 4, p. 103218, 2019.
72. T. Qian, L. Wang, C. Le, and Y. Zhou, "Low-temperature-steam activation of phosphorus in biochar derived from enhanced biological phosphorus removal (EBPR) sludge," *Water Res.*, vol. 161, pp. 202–210, 2019.
73. S. Salehi, S. Alijani, and M. Anbia, "Enhanced adsorption properties of zirconium modified chitosan-zeolite nanocomposites for vanadium ion removal," *Int. J. Biol. Macromol.*, vol. 164, pp. 105–120, 2020.

74. J. Xiao, R. Hu, G. Chen, and B. Xing, "Facile synthesis of multifunctional bone biochar composites decorated with Fe/Mn oxide micro-nanoparticles: Physicochemical properties, heavy metals sorption behavior and mechanism," *J. Hazard. Mater.*, vol. 399, no. May, p. 123067, 2020.
75. V. G. Georgieva, L. Gonsalvesh, and M. P. Tavlieva, "Thermodynamics and kinetics of the removal of nickel (II) ions from aqueous solutions by biochar adsorbent made from agro-waste walnut shells," *J. Mol. Liq.*, vol. 312, p. 112788, 2020.
76. M. H. Gonzalez *et al.*, "Coconut coir as biosorbent for Cr(VI) removal from laboratory wastewater," *J. Hazard. Mater.*, vol. 159, no. 2–3, pp. 252–256, 2008.
77. M. E. El-Naggar *et al.*, "Synthesis, characterization and adsorption properties of microcrystalline cellulose based nanogel for dyes and heavy metals removal," *Int. J. Biol. Macromol.*, vol. 113, pp. 248–258, 2018.
78. C. Shi, F. Tao, and Y. Cui, "Evaluation of nitriloacetic acid modified cellulose film on adsorption of methylene blue," *Int. J. Biol. Macromol.*, vol. 114, pp. 400–407, 2018.
79. K. Gobi, M. D. Mashitah, and V. M. Vadivelu, "Adsorptive removal of Methylene Blue using novel adsorbent from palm oil mill effluent waste activated sludge: Equilibrium, thermodynamics and kinetic studies," *Chem. Eng. J.*, vol. 171, no. 3, pp. 1246–1252, 2011.
80. R. A. K. Rao and U. Khan, "Adsorption of Ni(II) on alkali treated pineapple residue (Ananas comosus L.): Batch and column studies," *Groundw. Sustain. Dev.*, vol. 5, no. June, pp. 244–252, 2017.
81. Mardiah, R. Fathoni, P. Pudyaningtyas, H. Gamu, and Rinaldy, "Reuse of newspaper as an adsorbent for Cu (II) removal by citric acid modification," *MATEC Web Conf.*, vol. 156, 2018.
82. H. Chen, Q. Han, R. A. Venditti, and H. Jameel, "Enzymatic hydrolysis of pretreated newspaper having high lignin content for bioethanol production," *BioResources*, vol. 10, no. 3, pp. 4077–4098, 2015.
83. S. R. Hassan, H. M. Zwain, N. Q. Zaman, and I. Dahlan, "Recycled paper mill effluent treatment in a modified anaerobic baffled reactor: Start-up and steady-state performance," *Environ. Technol. (United Kingdom)*, vol. 35, no. 3, pp. 294–299, 2014.

84. D. Haldar and M. K. Purkait, “Micro and nanocrystalline cellulose derivatives of lignocellulosic biomass: A review on synthesis, applications and advancements,” *Carbohydr. Polym.*, vol. 250, no. August, p. 116937, 2020.
85. Z. N. Garba, I. Lawan, W. Zhou, M. Zhang, L. Wang, and Z. Yuan, “Microcrystalline cellulose (MCC) based materials as emerging adsorbents for the removal of dyes and heavy metals – A review,” *Sci. Total Environ.*, vol. 717, p. 135070, 2020.
86. A. Zaman, F. Huang, M. Jiang, W. Wei, and Z. Zhou, “Preparation, Properties, and Applications of Natural Cellulosic Aerogels: A Review,” *Energy Built Environ.*, vol. 1, no. 1, pp. 60–76, 2020.
87. C. Chen, J. Luo, W. Qin, and Z. Tong, “Elemental analysis, chemical composition, cellulose crystallinity, and FT-IR spectra of Toona sinensis wood,” *Monatshefte fur Chemie*, vol. 145, no. 1, pp. 175–185, 2014.
88. T. M. Sampath Udeni Gunathilake, Y. C. Ching, C. H. Chuah, N. A. Rahman, and N. S. Liou, “Recent advances in celluloses and their hybrids for stimuli-responsive drug delivery,” *Int. J. Biol. Macromol.*, vol. 158, pp. 670–688, 2020.
89. L. Pang, Z. Gao, H. Feng, S. Wang, and Q. Wang, “Cellulose based materials for controlled release formulations of agrochemicals: A review of modifications and applications,” *J. Control. Release*, vol. 316, no. August, pp. 105–115, 2019.
90. Z. Zhu, S. Fu, N. Lavoine, and L. A. Lucia, “Structural reconstruction strategies for the design of cellulose nanomaterials and aligned wood cellulose-based functional materials – A review,” *Carbohydr. Polym.*, vol. 247, no. April, p. 116722, 2020.
91. A. Farooq *et al.*, “Cellulose from sources to nanocellulose and an overview of synthesis and properties of nanocellulose/zinc oxide nanocomposite materials,” *Int. J. Biol. Macromol.*, vol. 154, pp. 1050–1073, 2020.
92. G. Morandi, L. Heath, and W. Thielemans, “Cellulose nanocrystals grafted with polystyrene chains through Surface-Initiated Atom Transfer Radical Polymerization (SI-ATRP),” *Langmuir*, vol. 25, no. 14, pp. 8280–8286, 2009.
93. H. K. Lim, T. T. Teng, M. H. Ibrahim, A. Ahmad, and H. T. Chee, “Adsorption and Removal of Zinc (II) from Aqueous Solution Using Powdered Fish Bones,” *APCBEE Procedia*, vol. 1, no. January, pp. 96–102,

2012.

94. R. Saha, K. Mukherjee, I. Saha, A. Ghosh, S. K. Ghosh, and B. Saha, "Removal of hexavalent chromium from water by adsorption on mosambi (Citrus limetta) peel," *Res. Chem. Intermed.*, vol. 39, no. 5, pp. 2245–2257, 2013.
95. N. Tazrouti and M. Amrani, "Chromium (VI) adsorption onto activated kraft lignin produced from alfa grass (Stipa tenacissima)," *BioResources*, vol. 4, no. 2, pp. 740–755, 2009.
96. M. Iqbal, A. Saeed, and I. Kalim, "Characterization of adsorptive capacity and investigation of mechanism of Cu2+, Ni2+ and Zn2+ adsorption on mango peel waste from constituted metal solution and genuine electroplating effluent," *Sep. Sci. Technol.*, vol. 44, no. 15, pp. 3770–3791, 2009.
97. L. Semerjian, "Removal of heavy metals (Cu, Pb) from aqueous solutions using pine (Pinus halepensis) sawdust: Equilibrium, kinetic, and thermodynamic studies," *Environ. Technol. Innov.*, vol. 12, pp. 91–103, 2018.
98. R. Davarnejad, M. K. Moraveji, and M. Havaie, "Integral technique for evaluation and optimization of Ni (II) ions adsorption onto regenerated cellulose using response surface methodology," *Arab. J. Chem.*, vol. 11, no. 3, pp. 370–379, 2018.
99. M. H. Dehghani *et al.*, "Investigating the equilibrium and adsorption kinetics for the removal of Ni (II) ions from aqueous solutions using adsorbents prepared from the modified waste newspapers: A low-cost and available adsorbent," *Microchem. J.*, vol. 146, no. February, pp. 1043–1053, 2019.
100. S. Pourfadakari, S. Jor, and M. Ahmadi, "Data in Brief Experimental data on adsorption of Cr (VI) from aqueous solution using nanosized cellulose fi bers obtained from rice husk," vol. 15, pp. 887–895, 2017.
101. H. A. Hegazi, "Removal of heavy metals from wastewater using agricultural and industrial wastes as adsorbents," *HBRC J.*, vol. 9, no. 3, pp. 276–282, 2013.
102. J. Matusiak, E. Grządka, and A. Bastrzyk, "Stability, adsorption and electrokinetic properties of the chitosan/silica system," *Colloids Surfaces A Physicochem. Eng. Asp.*, vol. 554, no. June, pp. 245–252, 2018.
103. F. Ji, C. Li, B. Tang, J. Xu, G. Lu, and P. Liu, "Preparation of cellulose acetate/zeolite composite fiber and its adsorption behavior for heavy metal

ions in aqueous solution," *Chem. Eng. J.*, vol. 209, pp. 325–333, 2012.

104. R. Srinivasan, "Advances in application of natural clay and its composites in removal of biological, organic, and inorganic contaminants from drinking water," *Adv. Mater. Sci. Eng.*, vol. 2011, 2011.

105. A. M. Farag, H. H. Sokker, E. M. Zayed, F. A. Nour Eldien, and N. M. Abd Alrahman, "Removal of hazardous pollutants using bifunctional hydrogel obtained from modified starch by grafting copolymerization," *Int. J. Biol. Macromol.*, vol. 120, pp. 2188–2199, 2018.

106. B. M. Ibrahim and N. A. Fakhre, "Crown ether modification of starch for adsorption of heavy metals from synthetic wastewater," *Int. J. Biol. Macromol.*, vol. 123, pp. 70–80, 2019.

107. H. Dogan and N. D. Hilmioglu, "Zeolite-filled regenerated cellulose membranes for pervaporative dehydration of glycerol," *Vacuum*, vol. 84, no. 9, pp. 1123–1132, 2010.

108. S. Ummartyotin and H. Manuspiya, "A critical review on cellulose: From fundamental to an approach on sensor technology," *Renew. Sustain. Energy Rev.*, vol. 41, pp. 402–412, 2015.

109. X. He, L. Cheng, Y. Wang, J. Zhao, W. Zhang, and C. Lu, "Aerogels from quaternary ammonium-functionalized cellulose nanofibers for rapid removal of Cr(VI) from water," *Carbohydr. Polym.*, vol. 111, pp. 683–687, 2014.

110. G. Abdul, X. Zhu, and B. Chen, "Structural characteristics of biochar-graphene nanosheet composites and their adsorption performance for phthalic acid esters," *Chem. Eng. J.*, vol. 319, pp. 9–20, 2017.

111. L. Wojnárovits, C. M. Földváry, and E. Takács, "Radiation-induced grafting of cellulose for adsorption of hazardous water pollutants: A review," *Radiat. Phys. Chem.*, vol. 79, no. 8, pp. 848–862, 2010.

112. L. O. Ekebafe, M. O. Ekebafe, F. A. O. Akpa, G. Erhuanga, and B. W. Etiobhio, "Graft Polimerizacija Akrilonitrila Na Delignifikovanim Celuloznim Materijalima Dobijenim Iz Bambusa (Bambusa Vulgaris) I Njihova Primena Za Uklanjanje Teških Metala Iz Vodenih Rastvora," *Chem. Ind. Chem. Eng. Q.*, vol. 17, no. 2, pp. 133–140, 2011.

113. A. Bhattacharya and B. N. Misra, "Grafting: A versatile means to modify polymers: Techniques, factors and applications," *Prog. Polym. Sci.*, vol. 29, no. 8, pp. 767–814, 2004.

114. A. S. Singha and R. K. Rana, "Functionalization of cellulosic fibers by graft copolymerization of acrylonitrile and ethyl acrylate from their binary mixtures," *Carbohydr. Polym.*, vol. 87, no. 1, pp. 500–511, 2012.
115. A. Y. Kulkarni and P. C. Mehta, "Ceric ion-induced redox polymerization of acrylonitrile on cellulose," *J. Appl. Polym. Sci.*, vol. 12, no. 6, pp. 1321–1342, 1968.
116. V. Kumar, S. Naithani, and D. Pandey, "Optimization of reaction conditions for grafting of α-cellulose isolated from Lantana camara with acrylamide," *Carbohydr. Polym.*, vol. 86, no. 2, pp. 760–768, 2011.
117. A. Ali, "Removal of Mn(II) from water using chemically modified banana peels as efficient adsorbent," *Environ. Nanotechnology, Monit. Manag.*, vol. 7, pp. 57–63, 2017.
118. L. Liu, J. P. Xie, Y. J. Li, Q. Zhang, and J. M. Yao, "Three-dimensional macroporous cellulose-based bioadsorbents for efficient removal of nickel ions from aqueous solution," *Cellulose*, vol. 23, no. 1, pp. 723–736, 2016.
119. A. A. Yakout, R. H. El-Sokkary, M. A. Shreadah, and O. G. Abdel Hamid, "Removal of Cd(II) and Pb(II) from wastewater by using triethylenetetramine functionalized grafted cellulose acetate-manganese dioxide composite," *Carbohydr. Polym.*, vol. 148, pp. 406–414, 2016.
120. X. Qiu, X. Ren, and S. Hu, "Fabrication of dual-responsive cellulose-based membrane via simplified surface-initiated ATRP," *Carbohydr. Polym.*, vol. 92, no. 2, pp. 1887–1895, 2013.
121. I. Kaur, R. Kumar, and N. Sharma, "A comparative study on the graft copolymerization of acrylic acid onto rayon fibre by a ceric ion redox system and a γ-radiation method," *Carbohydr. Res.*, vol. 345, no. 15, pp. 2164–2173, 2010.
122. H. M. Abd El Salam, S. M. Sayyh, and E. H. M. Kamal, "Enhancing both the mechanical and chemical properties of paper sheet by graft co-polymerization with acrylonitrile/methyl methacrylate," *Beni-Suef Univ. J. Basic Appl. Sci.*, vol. 3, no. 3, pp. 193–202, 2014.
123. I. Kaur, N. Sharma, and V. Kumari, "Modification of fiber properties through grafting of acrylonitrile to rayon by chemical and radiation methods," *J. Adv. Res.*, vol. 4, no. 6, pp. 547–557, 2013.
124. T. T. Hanh, H. T. Huy, and N. Q. Hien, "Pre-irradiation grafting of

acrylonitrile onto chitin for adsorption of arsenic in water," *Radiat. Phys. Chem.*, vol. 106, pp. 235–241, 2015.

125. N. A. Awang, W. N. Wan Salleh, A. F. Ismail, N. Yusof, F. Aziz, and J. Jaafar, "Adsorption Behavior of Chromium(VI) onto Regenerated Cellulose Membrane," *Ind. Eng. Chem. Res.*, vol. 58, no. 2, pp. 720–728, 2019.
126. M. A. Mohamed, W. N. W. Salleh, J. Jaafar, S. E. A. M. Asri, and A. F. Ismail, "Physicochemical properties of 'green' nanocrystalline cellulose isolated from recycled newspaper," *RSC Adv.*, vol. 5, no. 38, pp. 29842–29849, 2015.
127. E. S. Abdel-Halim, "Chemical modification of cellulose extracted from sugarcane bagasse: Preparation of hydroxyethyl cellulose," *Arab. J. Chem.*, vol. 7, no. 3, pp. 362–371, 2014.
128. C. A. Hubbell and A. J. Ragauskas, "Effect of acid-chlorite delignification on cellulose degree of polymerization," *Bioresour. Technol.*, vol. 101, no. 19, pp. 7410–7415, 2010.
129. E. S. Abdel-Halim, H. H. Alanazi, and A. A. Alghamdi, "Extraction and bleaching of olive tree branch cellulose," *BioResources*, vol. 10, no. 4, pp. 7136–7150, 2015.
130. A. A. M. A. Nada, M. Y. Alkady, and H. M. Fekry, "Synthesis and characterization of grafted cellulose for use in water and metal ions sorption," *BioResources*, vol. 3, no. 1, pp. 46–59, 2008.
131. H. T. Sahin and M. B. Arslan, "A study on physical and chemical properties of cellulose paper immersed in various solvent mixtures," *Int. J. Mol. Sci.*, vol. 9, no. 1, pp. 78–88, 2008.
132. M. K. Mohamad Haafiz, S. J. Eichhorn, A. Hassan, and M. Jawaid, "Isolation and characterization of microcrystalline cellulose from oil palm biomass residue," *Carbohydr. Polym.*, vol. 93, no. 2, pp. 628–634, 2013.
133. A. Sonia and K. Priya Dasan, "Chemical, morphology and thermal evaluation of cellulose microfibers obtained from Hibiscus sabdariffa," *Carbohydr. Polym.*, vol. 92, no. 1, pp. 668–674, 2013.
134. G. Siqueira, J. Bras, and A. Dufresne, "Cellulosic bionanocomposites: A review of preparation, properties and applications," *Polymers (Basel).*, vol. 2, no. 4, pp. 728–765, 2010.
135. A. Mandal and D. Chakrabarty, "Isolation of nanocellulose from waste

sugarcane bagasse (SCB) and its characterization," *Carbohydr. Polym.*, vol. 86, no. 3, pp. 1291–1299, 2011.

136. D. Liu, T. Zhong, P. R. Chang, K. Li, and Q. Wu, "Starch composites reinforced by bamboo cellulosic crystals," *Bioresour. Technol.*, vol. 101, no. 7, pp. 2529–2536, 2010.
137. P. Lu and Y. Lo Hsieh, "Preparation and characterization of cellulose nanocrystals from rice straw," *Carbohydr. Polym.*, vol. 87, no. 1, pp. 564–573, 2012.
138. A. Mukherjee *et al.*, "Free radical induced grafting of acrylonitrile on pre-treated rice straw for enhancing its durability and flame retardancy," *J. Adv. Res.*, vol. 8, no. 1, pp. 73–83, 2017.
139. G. Xia *et al.*, "Cellulose-based films prepared directly from waste newspapers via an ionic liquid," *Carbohydr. Polym.*, vol. 151, pp. 223–229, 2016.
140. V. K. Thakur, M. K. Thakur, and R. K. Gupta, "Rapid synthesis of graft copolymers from natural cellulose fibers," *Carbohydr. Polym.*, vol. 98, no. 1, pp. 820–828, 2013.
141. M. Müller, C. Riekel, R. Vuong, and H. Chanzy, "Skin/core micro-structure in viscose rayon fibres analysed by X-ray microbeam and electron diffraction mapping," *Polymer (Guildf).*, vol. 41, no. 7, pp. 2627–2632, 2000.
142. Z. Sun and F. Chen, "Homogeneous grafting copolymerization of methylmethacrylate onto cellulose using ammonium persulfate," *Cellul. Chem. Technol.*, vol. 48, no. 3–4, pp. 217–223, 2014.
143. S. Wadhawan, A. Jain, J. Nayyar, and S. K. Mehta, "Role of nanomaterials as adsorbents in heavy metal ion removal from waste water: A review," *J. Water Process Eng.*, vol. 33, no. October 2019, p. 101038, 2020.
144. J. Anandkumar and B. Mandal, "Removal of Cr(VI) from aqueous solution using Bael fruit (Aegle marmelos correa) shell as an adsorbent," *J. Hazard. Mater.*, vol. 168, no. 2–3, pp. 633–640, 2009.

145 T. S. Anirudhan, J. Nima, and P. L. Divya, "Adsorption of chromium(VI) from aqueous solutions by glycidylmethacrylate- grafted-densified cellulose with quaternary ammonium groups," *Appl. Surf. Sci.*, vol. 279, pp. 441–449, 2013.

146. Y. Lin *et al.*, "Synthesis of poly(Acrylic acid)-grafted carboxymethyl cellulose for efficient removal of copper ions," *J. Renew. Mater.*, vol. 7, no. 12, pp.

1403–1414, 2019.
147. E. Heraldy, Wireni, and W. W. Lestari, "Biosorption of Cu(II) ions by cellulose of cabbage waste as biosorbent from agricultural waste," *AIP Conf. Proc.*, vol. 1710, no. Ii, 2016.
148. S. Ben Cheikh, R. Ben Cheikh, E. Cunha, P. E. Lopes, and M. C. Paiva, "Production of cellulose nanofibers from Alfa grass and application as reinforcement for polyvinyl alcohol," *Plast. Rubber Compos.*, vol. 47, no. 7, pp. 297–305, 2018.
149. G. Z. Kyzas, N. K. Lazaridis, and M. Kostoglou, "On the simultaneous adsorption of a reactive dye and hexavalent chromium from aqueous solutions onto grafted chitosan," *J. Colloid Interface Sci.*, vol. 407, pp. 432–441, 2013.
150. R. Yang *et al.*, "Thiol-modified cellulose nanofibrous composite membranes for chromium (VI) and lead (II) adsorption," *Polymer (Guildf).*, vol. 55, no. 5, pp. 1167–1176, 2014.
151. I. M. Kenawy, M. A. H. Hafez, M. A. Ismail, and M. A. Hashem, "Adsorption of Cu(II), Cd(II), Hg(II), Pb(II) and Zn(II) from aqueous single metal solutions by guanyl-modified cellulose," *Int. J. Biol. Macromol.*, vol. 107, pp. 1538–1549, 2018.

LIST OF PUBLICATIONS

Journal with Impact Factor

1. **Nor Asikin Awang,** Wan Norharyati Wan Salleh, Ahmad Fauzi Ismail, Norhaniza Yusof, Farhana Aziz & Juhana Jaafar, "Adsorption Behavior of Chromium(VI) onto Regenerated Cellulose Membrane," *Industrial Engineering Chemical Research*. (vol. 58, no. 2, pp. 720-728, 2019) (Q1, IF: 3.375)

Indexed Journal

1. **Nor Asikin Awang,** Wan Norharyati Wan Salleh, Hasrinah Hasbullah, Norhaniza Yusof, Farhana Aziz, Juhana Jaafar, & Ahmad Fauzi Ismail, "Graft Copolymerization of Acrylonitrile onto Recylced Newspapers Cellulose Pulp," *AIP Conference Proceedings*. (vol. 1885, no. 1, pp.1-8, 2017).
2. **Nor Asikin Awang,** Wan Norharyati Wan Salleh, Hasrinah Hasbullah, & Ahmad Fauzi Ismail, "Synthesis, Characterization and Adsorption Properties of Grafted Cellulose for Cr(VI) Removal," *Material Today Proceedings*. (vol. 19, no. 4, pp.1777-1786, 2019).
3. **Nor Asikin Awang,** Wan Norharyati Wan Salleh, Hasrinah Hasbullah, Siti Zu Nurain Ahmad, Nor Hafiza Ismail, Nurafiqah Rosman, & Norhaniza Yusof, "Modified Recycled Paper-Based Adsorbent for Nickel Removal," *IOP Conference Series: Materials Science and Engineering*. (vol. 736, no. 072001, 2020).
4. **Nor Asikin Awang,** Wan Norharyati Wan Salleh, Mohamad Fakrudin Mustakim Alisah Afiqah, Ahmad Fauzi Ismail, Nurafiqah Rosman, & Norhaniza Yusof, "Adsorption of Cesium from Aqueous Solution using Chitosan Beads," *Jurnal Teknologi*. (vol. 81, no. 2, pp. 135-140, 2019).

Book Chapter

1. **Nor Asikin Awang,** Mohamad Azuwa Mohamed, &Wan Norharyati Wan Salleh, "Polyvinyl alcohol/cellulose-based bio-composites and bio-nanocomposites" *Wiley and Sons (Scrivener Publishing)*. **(Status: Under Review)**