Removal of pollutants from aqueous environment by using hydrogels and their composites: Review on recent development

[1]Ankur H. Gor & [1,†,*]Pragnesh N Dave
[1]Department of Chmeistry, K. S. K. V. Kachchh University, Mundra Road, Bhuj-370 001 (Kachchh), Gujarat, India
[†] Present address: Department of Chemsitry,Sardar Patel University,
Vallabh Vidyangar-388 120,Gujarat,India

Abstract : Hydrogels and hydrogel composites are the potential candidates for removal of pollutant environmental remediation because of the high surface area, hydrophilic and high swelling capacity. In review, first we have discussed the classification, methods of preparation of hydrogels. In addition, we have also discussed the source of water pollution and its separation techniques, mainly adsorption discussed in brief. We have discussed the applications of hydrogel and hydrogel composite for adsorption of pollutant and effect of diversified factors on it. Finally, we have discussed the recent development in hydrogels for removal of heavy metal, dyes, anions, organic pollutant, antibiotics, and pesticides.

Index Terms **- Wastewater, Hydrogel, Adsorption, Heavy metals, pesticides.**

I. Introduction

On earth, two-thirds part is cover by water, but the availability of clean and containment free water still issues for human society. Nature has its own mechanism to recycle the water, but the natural process disturbed by growth in industrial operation, human population [1]. While growth in metal processing, leather tanning, pesticide industries, pharmaceutical industries and textile industries always consumes a large amount of water for different operations and they discharge of large amounts of wastewater in an environment that contain the various organic and inorganic chemical compounds. Most of the pollutants exist in the wastewater creates issues for human health and the environment. Therefore, It is necessary to scale down or restraint assets in the environment. The draining of wastewater without any treatment leads to disparate environmental complication such as diminish the dissolved oxygen in aquatic systems, the presence of microorganisms, pathogens and toxic compound threatening to human health, increase the nutrient content in the lakes and decomposition of organic compound leads to the formation of malodorous gases [2]. Therefore, industry should expunge the harmful chemical bodies from polluted wastewater before draining into an environment. Polluted water has an effect on the human society, aquatic living system, and durability of the environment. Different technologies have been developed for wastewater treatment but these technologies are expensive and require high energy, large area. Wastewater treatment and reuse of the water is crucial topic and researchers are still focusing to develop the inexpensive, easily applicable technologies. In order to address these problems, it is the challenge for scientists to develop special technology which breaks down the barrier of the traditional technologies which were used for wastewater treatment. Hydrogel technology offers the immanent properties for wastewater treatment with low cost and environment-friendly polymeric material.

A hydrogel defined as a cross linked hydrophilic polymer network with great capacity to absorb water and remain insoluble due to crosslinks [3]. Hydrogel also defined as polymeric systems which have ability to swell in the water and preserve significant fraction (>20%) of water inside their network without dissolving in water [4]. Due to water absorbing capacity, hydrogel gained interest of researchers. In the fabrication of hydrogels, various type of natural, synthetic and altered polymers used.

Swelling property of hydrogel has played a crucial role in wide range applications and especially in wastewater treatment. Last few years, assemble of hydrogels with of chelating functional groups for treatment of wastewater had gained the interest of researchers. Synthetic polymers such as acrylamide, acrylic acid, N-is opropylacrylamide, Itaconic acid, 2-Hydroxyethyl-methacrylate, succinic acid, polyvinylpyrollidine, and Polyvinyl alcohol have been used in the preparation of hydrogels and explored their applications for the wastewater treatment [6-10]. Hydrogels with synthetic polymers carried certain disadvantages like biodegradability and cost consuming. To address these problems, hydrogel formation with natural polymers has numerous properties like high swelling capacity, biodegradability and cheap. Hydrogel based on the natural polymer such as polypeptides collagen, and polysaccharides have been designed for wastewater treatment [11-14]. To enhance the hydrogel property, composite and nanocomposite form of a hydrogel is also reliable technologies.

In this chapter, we have discussed in the brief overview of recent developments on hydrogel and hydrogel based composite for wastewater treatment. First, we have discussed the classification of hydrogel and different synthesis methods. In addition, we have also discussed the water pollution and separation technology for wastewater treatment including adsorption and photocatalysis. We have discussed the application of hydrogel as an adsorbent for wastewater treatment and application of hydrogel composite as photocatalyst material for wastewater treatment and the effect of various factors on adsorption and photocatalysis. We have discussed the recent development in the removal of metal ion, dyes, anions, pesticides, antibiotics, and some organic pollutant. Finally, we have discussed the conclusion of the review in hydrogel and hydrogel composite for wastewater treatmentt.

II. Wastewater treatment by adsorption

Adsorption technique is the most favorable method for the treatment of wastewater because of it is cheap, simple, and universal method. Adsorption can use to remove the soluble and insoluble pollutants. Adsorption technique also applied for source reduction, reclamation for potable, industrial, and other purposes [15]. Sometimes adsorption is reversible and it should be regenerate by using a suitable process [16]. Adsorption is a surface phenomenon. Adsorption is a procedure, which involves the boom of molecules on the surface of the adsorbent. The substance being adsorb is the adsorbate and the adsorbing material is termed as the adsorbent. Surface forces play a key role in the adsorption process. Depend On the type of interaction between the

adsorbate molecule and adsorbent, adsorption process have classified in two ways: Physisorption and Chemisorption. If the interaction has a physical in nature, the process called the physisorption and if the interaction of adsorbate molecule and adsorbent due to chemical bonding or electrostatics interaction, the adsorption process called the chemisorption.

According to the adsorption procedure, adsorption equilibrium of adsorbate between the solution and adsorbent is attaining. The adsorption amount (q_e, mg/g) of the adsorbate at the equilibrium step was determined according to the following equation:

$$q_e = \frac{V(C_0 - Ce)}{M} \quad (1)$$

Where V is the solution volume (L), M is the mass of monolithic adsorbents (g), and Co and Ce are the initial and equilibrium adsorbate concentrations [17].

III. Adsorption Isotherms

Adsorption isotherms are the statically explication of a thermodynamic equilibrium relationship between the quantity of adsorbed adsorbate by a unit mass of adsorbent and the amount of adsorbate remaining in the bulk fluid phase at a constant temperature and pH [18].

(I) Langmuir isotherm

It is simple and still the most useful isotherm and valid for monolayer. This mode assumes the only single layer formed on the adsorption surface. Once on the adsorbent site occupied by adsorbate than no further adsorption takes place, thus adsorption of adsorbate on adsorbent at specific sites and adsorption is limited to specific sites [19]. The significant preferred standpoint of the Langmuir equation is the fact that it provides an estimate for the adsorption maximum of the adsorbent and a coefficient that is a measure of the adsorbate–adsorbent bonding energy [20]. The monolayer is form when the strength of the bond between adsorbate-adsorbent molecules is stronger than other interaction in adsorbent-adsorbate-solvent system. The first adsorbed molecule act as the initiator at the site of adsorption for second molecule will attached on the neighboring site [21]. Langmuir isotherm can be written as in eq. (2).

$$\frac{q_e}{q_m} = \frac{bC_e}{1+bC_e} \quad (2)$$

(II) Freundlich isotherm

The expression depicting of isothermal variation of adsorption of an amount of gas adsorbed by unit mass of solid adsorbent with pressure offered by herbert Freundlich in 1909. It is another form of Langmuir isotherm and used for the multilayer adsorption. The Freundlich isotherm used to describe the adsorption on the heterogeneous surface. According to isotherm, adsorption on the surface increases with increases the concentration of adsorbate [22]. Freundlich isotherm can be expressed in terms of an equation:

$$q_e = k_f C_e^{1/n} \quad (3)$$

K_f = Freundlich isotherm constant (mg/g) which indicate the adsorption capacity, 1/n =adsorption intensity, C_e = the equilibrium concentration of adsorbate (mg/L),q_e = The amount of metal adsorbed per gram of the adsorbent at equilibrium (mg/g).

If a value of n is equal to 1, then the distribution between the two phases are independent of the concentration. In a favourable adsorption, value of 1/n is below one but in a cooperative (unfavourable) adsorption, value of 1/n being above 1 [23-25].

(III) BET isotherm

Langmuir derived the equation for the isotherm of multilayer but it was complex after that Brunauer, Emmett, and Teller were developed a theory to account for multilayer adsorption [26]. BET is a special form of the Langmuir isotherm. In this isotherm pretended that the surface is energetically homogeneous and there is no interaction among adsorbed molecules [27]. BET equation can be expressed for liquid phase [28]:

$$q = q_m \frac{b_s C_e}{(1 - C_e b_L)[1 - - b_L C_e + b_s C_e]} \quad (4)$$

q_m(mg/g) is the amount of adsorbate per unit weight of adsorbent, C_e is the solute equilibrium concentration, b_s is the equilibrium constant of adsorption on the first layer, b is the equilibrium adsorption–desorption constant for the upper layers of adsorbate on the adsorbent.

IV. Wastewater treatment by hydrogel as an adsorbent

Cross linked structure of hydrogels are able to absorb the large quantity of water. Polymer chains covalently joined with each other and form cross linked structure. Some specific properties of hydrogels like the hydrophilicity and porous structure networks which make them swell quickly in the aqueous solution, which is beneficial for a reduction in the time to reach the adsorption equilibrium [29]. Surface of hydrogel provides floor for the adsorption phenomena. Adsorption is surface phenomena that take place on the surface of adsorbent. Hydrogel, Hydrogel-adsorbate-solvent system and Hydrogel-adsorbate-solvent system after adsorption are described in Fig. 1.

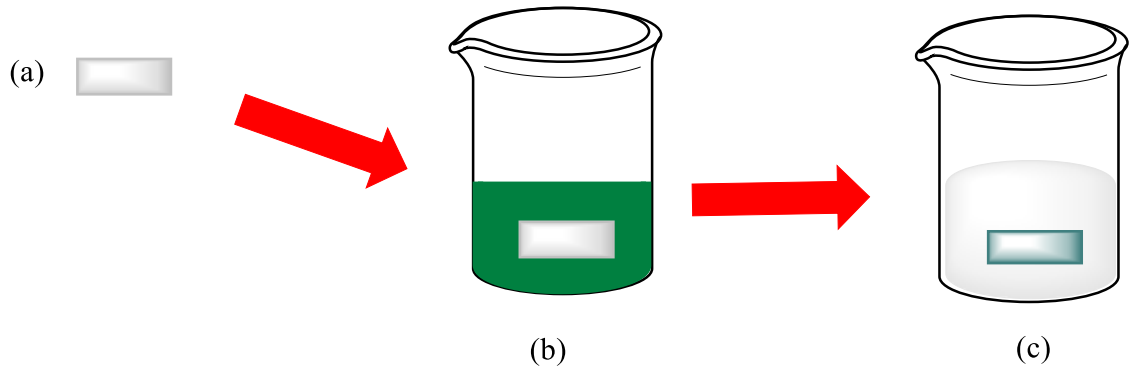

Fig. 1. (a) Hydrogel (b) Hydrogel-adsorbate-solvent system (c) Surface of hydrogel after adsorption.

Hydrogels comprise amide, amine, carboxylic acid, hydroxyl, and ammonium types of functional groups can bind metal ions and be good polychelatogens for water remediation applications [30]. Therefore, hydrogels offer potential as an adsorbent for removal of the pollutant from aqueous environment. In past, number of publication published on the applications of hydrogels as an adsorbent for removal of metal ions, dyes, and pesticides, inorganic and organic pollutant by adsorption process [31-35].

Factors effect on adsorption by hydrogels

(I) Effect of hydrogel structure and composition

Hydrogel composition has an influence on the selectivity and quantitative differentiation of pollutant. Adsorption of a pollutant from solution by hydrogel also depends on the presence of the functional group in hydrogel network, which play the anchor role in interaction with water. The chelating ability of hydrogels also depends on the functional group present in it. Adsorption of dyes investigated with a different changing composition of hydrogels based on the acrylamide, acrylic acid, and polyvinyl alcohol. Results indicated that the removal of methylene blue increased with increased the concentration of acrylamide up to the molar ratio of 1.5 but further increased the concentration of acrylamide then decreased removal efficiency. Hydrogel with 2% concentrations of acrylic acid had high removal ability [36].

The presence of more electron donor atoms such as N, O, P, S in the hydrogel network influences on the chelating ability of hydrogels. N-vinyl-2-pyrrolidone based homo polymer hydrogel and copolymer hydrogel based on N-vinyl-2-pyrrolidone and methylacrylate were crosslinked with macroinimer have prepared and investigated their ability to adsorb metal ion from solutions. Copolymer hydrogel showed higher metal ion binding capacity than homopolymer hydrogels because the copolymer hydrogel had the additional oxygen atom than homopolymer hydrogels [37]. Another example, Chitosan based polyacrylamide graft polymerized hydrogels investigated for adsorption of crude oil.

From **Fig. 2**, it can be observed that the higher removal efficiency of hydrogels observed with 20% and 30% an amount of acrylamide while lower removal percentage of oil showed in the hydrogel with 50% amount of acrylamide [38].

(II) Effect of contact time

From an economic point of view, the contact time is very crucial. The efficiency of hydrogels in wastewater treatment can be determined from the time required to adsorb the maximum amount of metal ions.

CMC-g-PAA/APT hydrogel composites applied for the adsorption of lead ion. At the beginning, adsorption rapidly increased and reached equilibrium within 60 min [39]. The highly hydrophilic nature of hydrogel reduced the contact time. CTS-g-PAA/UVMT hydrogel developed for the removal of ammonium-nitrogen. When water comes in contact with hydrogel, the ionization of poly acrylic acid takes place and which expands the hydrogel network and increase the porosity of hydrogel [40]. Poly (dimethyl diallyl ammonium chloride) and polyacrylamide based on reported hydrogels for the removal of nitrate aqueous solution. In the hydrogels, adsorption equilibrium reached at 60 min [41].

(III) Effect pH of feed solution

The pH of feed solution has great impact on the adsorption properties of hydrogel. Alteration in pH, change occurred in surface charge and the functional group present in the hydrogels. If hydrogel contain carboxylic groups, then at lower pH value, deprotonation of carboxylic group occurred which influence the adsorption capacity of hydrogels [42]. The graft copolymer type of composite hydrogels based on the Cellulose/polyacrylamide/hydroxyapatite reported for the removal of Cu^{+2}. The removal efficiency of hydrogel was high at PH 7 but decreased at above high pH due to precipitation of metal ion in hydroxide form. At lower pH value, the proton competitive mechanism plays vital role in the metal ion adsorption [43]. The pH is also a critical factor for adsorption of dyes. Mainly ionization of dyes is dependon the pH of feed solution. 3-acrylamidopropyl)-trimethylammoniumchloride and N, N-dimethylacrylamide monomer based copolymer hydrogel reported for the removal of azo dyes. The removal efficiency of hydrogel was higher at higher pH 11. In basic pH, the interaction occurred between amine groups in hydrogel and sulphate group in the dye molecule and so removal percentages of dyes increase [44].

(IV) Initial concentration of pollutant in feed solution

Initial concentration of pollutant has played significant role in the adsorption process and removal percentages of pollutant. Initial concentration of pollutant is proportional relationship with the adsorption capacity but inversely proportional relationship with the removal percentages. Hemicellulose, poly (methacrylic acid)/Carbon nanotube composite hydrogel constructed for the removal of the methylene blue. The higher initial concentration dye that reduce removal percentage slightly but adsorption amount increased [45]. Initial concentration of metal ion also plays decisive role in their adsorption. Metal ion adsorption on hydrogel takes place by physical and chemical interactions. Hydrogel based on the carboxymehylcellulose/2-acrylamido-2-methyl propane sulfonic acid reported for removal of metal ions. Author claimed that when the initial concentration of metal ion increased from 25 to 200 ppm then Adsorption capacity increased due to physical adsorption. When the concentration increased up to 1000 ppm, then the adsorption capacity decreased. In the presence of higher amount of metal ion then chemisorption take place [46].

(V) The temperature of feed solution

emperature mainly effects on the interaction between the adsorbate-adsorbent. HEA/AMPS monomer based hydrogel reported for removal of heavy metals. Effect of temperature on the adsorption of the metal ion by hydrogel investigated and it was found that the adsorption of divalent metal decreased as the temperature increased from 15 to 400C [47]. Another example, Poly (N-isopropylacrylamide)/Chitosan Semi-IPN based hydrogel used for the removal of radioactive Uranium (VI) ions. In thermo sensitive monomer based hydrogel at a lower temperature than LCST, Polymer network stretched and higher amount of adsorption of metal ion takes place but at higher temperature adsorption decreased [48].

(IV) Co-sorption

Since the past decade, researchers have investigated the influence of other chemically active species on adsorption process. Chitosan-g-poly (acrylic acid) /vermiculite hydrogel investigated for the adsorption of dyes in the presence of anionic surfactants. It was found that the limited concentration of surfactant increased adsorption of dyes. When surfactant in higher concentration form micelles, dissolves the dye molecule, and averts the adsorption of dye [49].

V. Recent developments in hydrogel and hydrogel composite for wastewater treatment

(I) Removal of metal ion and radioactive element

Industrial effluent contains larger amounts of hazardous heavy metals such as Cu, Pb, Hg, Ni, As, Cd, Cr. If they enter in higher proportion in aquatic system then they cause the serious problems for human health and aquatic living organisms. Therefore, it is

imperative to evacuate the heavy metals from wastewater. Jae-Woo Choi and his co-workers developed spherical hydrogel beads from alginates and examined for the competitive adsorption of Pb^{2+}, Cu^{2+}, and Ni^{2+} in batch and column study. In competitive adsorption study, it was found that the maximum uptake capacity of hydrogel beads was higher for the Pb^{2+} thanCu^{2+},Ni^{2+} because of Pb^{2+} has the maximum affinity to words carboxylic acid group as compare to Cu^{2+}, and Ni^{2+} [50]. Rajesh kumar et al. [51] developed 3-mercaptopropyl functionalized silica entrapped in the poly acrylamide hydrogel for the adsorption of arsenic. Adsorption equilibrium achieved within 30 min and maximum uptake of arsenic was 92.5µg/g obtained by hydrogel at an initial concentration of 100 µg/L.

M. G. Hamed et al. [52] synthesized hydrogel based on 2-acrylamido 2-methyl propane sulphonic acid, vinyl pyrilidone) monomer and silica as inorganic filler by utilizing radiation induced template method. Composite hydrogel investigated for the sorption of Cu^{+2}, Pb^{2+}, and rare earth element. Silica contain composite hydrogel has higher crosslinked structure and displayed the maximum sorption capacity compare to plain hydrogel. The composite hydrogel displayed a higher sorption capacity for Cu2+ was 99.8 mg/g and for $La^{3+,}$ Ce^{3+}, Nd^{3+}, Eu^{3+} and Pb^{2+} were 116, 103, 92, 76, 74 mg/gm. Nilanjana das et al. [53] developed hydrogel by using Chitosan, gum arabic, zinc oxide nanoparticles, montomorillonite and hydrogel nanobiocomposites modified by utilizing ß-cyclodextrin and triethylene tetra amine. In the modified systems, surface of structure changed by the interaction of cyclodextrin and triethylene tetra amine with crosslinking agent. The interaction of triethylene tetra amine with crosslinking agents creates the significant rough surface while interaction of cyclodextrin with crosslinking agents produces smooth surface. The availability of functional group and rough surface influeces on the adsorption of Hg(II). Therefore, TETA-NBC hydrogel showed the higher adsorption of Hg(II) as compared to CD-NBC hydrogel. Zhengkui li et al. [54] developed nano size hydrous manganese dioxide supported on (HMO-P(HMAm/HEA)) based hydrogel and applied for the removal for Pb^{2+}, Cu^{2+}, Cd^{2+} and Ni^{2+}. Adsorption of metal ion occurred due to the ion exchange between the –OH groups in hydrogel and metal ions. Among the all the metal ions, adsorption of Pb^{2+} was higher and adsorption of Ni^{2+} was lower. Reuse of hydrogel after consecutive cycles is key issue for wastewater treatment. T. Jamnongkan et al. [55] reported hydrogel based on PVA/chitosan blend type hydrogel and applied for the adsorption of Cu(II). Adsorption of Cu(II) mainly depended on the composition of hydrogel. Adsorption of Cu(II) increased by increasing the chitosan content in hydrogel network. Yianzheng et al. [56] reported magnetic macro porous chitosan grafted with poly (acrylic acid) hydrogel fabricated with the Pickering high internal emulsions template stabilized by modified Fe_3O_4 nanoparticles.A summary of the hydrogel and their application and regeneration of hydrogels is provided in Table 1.

Table 1. Hydrogels with different monomers, application of metal ion removal, and regeneration solutions.

Sr. No.	Hydrogel	Application	Regeneration	Ref.
1.	Alginate hydrogel beads	Removal of divalent metal ion (Pb^{2+}, Cu^{2+}, and Ni^{2+})	4% (v/v) of HCl	[50]
2.	Magnetic macroporous chitosan-g-poly (acrylic acid)	Removal of Pb^{2+} and Cd^{2+}.	0.5 mol/L of HCl	[56]
3	Poly (sodium acrylate-co-acrylamide)/ graphene oxide	Removal of Pb^{2+} and $Cd^{2+.}$	2%(v/v) of HCl	58]
4.	CTS/PAM double network hydrogel	Removal of Pb^{2+}, $Cu^{2+}$and Cd^{2+}.	1 M of HCl	[60]
5.	CTS-g-PAA monolith	Removal of Pb^{2+} and Cu^{2+}.	0.5 mol/L of HCl	[63]
6.	CTS-g-(AA-co-SS)/ISC hybrid hydrogel	Removal of Ce(III) and Gd(III).	0.5 mol/L of HCl, 0.1 mol/L of NaOH	[68]

Fe_3O_4 nanoparticles were modified by using 3-aminopropyl trimethoxysilane and methacryloxy-propyltrimethoxysilane. Pickering-HIPEs prepared by using P-xylene as the disperse phase, Chitosan aqueous solution as the continuous phase and Fe3O4-MNPs-M as the stable particle and acrylic acid as a monomer and small amount of co-surfactant. Macro porous structure and introduction of carboxyl group in hydrogels network responsible for the fast adsorption of metal ion. The maximum adsorption capacities for Cd^{2+} and Pb^{2+} were 308.84 mg/g and 695.22 mg/g.

S. N. Basri et al. [57] developed etherified sago starch and lactic acid based hydrogel prepared by irradiation technique for the removal of Pb, Cu, and Cd ions in their aqueous solutions. The ionic interaction between functional groups presents in the hydrogel play vital role in the removal of the metal ions. The removal capacity of hydrogels showed a maximum for lead ion while minimum for cadmium ion. Fan Zhang et al. [58] developed the hydrogel based on the acrylamide, sodium acrylate and graphene based hydrogel prepared by free radical polymerization and applied for the removal of the Pb^{2+} and Cd^{2+}. GO sheets introduced in the hydrogel network through hydrogen bonding. Adsorption of metal ions take place by chemisorption process and hydrogel had high affinity for Pb^{2+} then Cd^{2+}. The chelation and ion exchange mechanism takes part in the expulsion of the metal ion.

Removal of metal ions from the mixed contaminants still challenge for scientist. Zuliangchen et al. [59] prepared agrose-Fe nanoparticle based hydrogel for the removal of Cr(II). Hydrogel examined for the removal of Cr (II) individual also in presence of trichloroethylene. The results of the study suggested, Approx. 94.1% of Cr(VI) removed individually and 84.9% of Cr(VI) removed in presence of trichloroethylene. Y. Wai et al. [60] reported a novel concept of double network hydrogel for the use of hydrogel as an adsorbent. Chitosan crosslinked by EDTA and NMBA crosslinked polyacrylamide based double network hydrogel developed and scrutinized for the removal of heavy metals such as Cd(II), Cu(II), and Pb(II). The maximum uptake capacity of double network hydrogel for Cd(II), Cu(II) and Pb(II) were 86.00, 99.44 and 138.41 mg/g. Mousaghaemy et al. [61] developed magnetic hydrogel beads based on the tragacanth gum modified by (AMPS-co-VI) through grafting, graphene oxide and Fe3O4 nanoparticles hydrogel composites investigated for the removal of Pb(II), Cu(II). The surface topology of hydrogel changed into the porous and rough after the inclusion of graphene oxide which boost the adsorption of metal ion. Magnetic hydrogel beads have the maximum adsorption of Pb(II), Cu(II) were found 81.78 and 69.67 mgg-1. Meng-Jiywang et al. [62] reported environmental friendly PVA/CMC based porous hydrogel prepared by repeated freeze-thawing process and investigated the adsorption of metal ions. Functional group present in the PVA and CMC provides a binding site for adsorption of metal ions. Adsorption studies of metal ions revealed that among the all of these metal ions, adsorption of monovalent metal ion (Ag^{+}) was higher as compared to other divalent (Ni^{2+}, Cu^{2+}, and Zn^{2+}) metal ions. Yianzheng et al. [63] reported high internal phase emulsion technique used for the development of monolithic super macroporous hydrogel with 3D structure of the polymeric

network based on grafting of acrylic acid (AA) chains onto the backbone of chitosan and applied for the adsorption of Cu^{2+} and Pb^{2+}. From the experimental data, it was found that monolithic hydrogel was effective adsorbent for the removal of Cu^{2+} and Pb^{2+} and adsorbent was fast due to porous network. After complete five cycles of adsorption–desorption, the hydrogel still exhibited the highest adsorption capacity for metal ion removal.

Wei yang et al. [64] developed chitosan/reduced graphene oxide, montmorillonite hydrogel and investigated for the removal of Cr(VI). Surface area and surface property are critical criteria for an adsorbent. When rGO and montmorillonite introduced in the chitosan hydrogel, hydrogel became porous and rough surface formed and surface area becomes higher. Hydrogel composites had maximum uptake of Cr(VI) of composite was 87.03 mg/g and sorption increased with increasing temperature. Yun zhang et al. [65] prepared multifunctional polymer hydrogels based on inorganic polymer such as amino-modified zirconium oxide and natural organic polymer such tannic acid/alginate for the removal of the metal ions such as Pb^{2+}, Hg^{2+}, and CrO_4^{2-}. To study conducted to examine the synergistic effect of functional group presents in the hydrogel network on the adsorption of metal ions. The presence of reactive functional groups and structured network of hydrogel offers good an adsorbent for the metal ion. The ionic exchange and complexation take in the account for adsorption of Pb^{2+} and Hg^{2+} but adsorption of CrO_4^{2-} occurs by the electrostatic attraction and specific adsorption. Chengbinliu et al. [66] reported polyampholyte hydrogel with highly reactive amine and carboxyl group inserted by a radical polymerization procedure and applied for the removal of Pb(II) and Cd(II). Hydrogels have a good mechanical property, high adsorption rate and easily regenerated and reuse. The maximum uptake capacities of hydrogel was 216.1 mg/g for Pb(II) and 153.8 mg/g for Cd(II). Lina ma et al. [67] developed the polyacrylamide-phytic acid-polydopamine conducting porous hydrogel by three different interactions and applied for the removal of Cu^{2+}. In the hydrogel formation, AAm polymerise by free radical polymerisation, poly dopamine by coupling reaction and phytic acid crosslinks the poly dopamine and poly acrylamide. Hydrogel displayed the good capacity to remove of the Cu^{2+}. Wang Wenbo et al. [68] and his co-workers prepared composite hydrogel based on chitosan, acrylic acid sodium p-styrene sulfonate and semecite clay as an organic additive material by free radical graft polymerization and applied for the adsorption of Ce+3and Gd+3. Hydrogel had the maximum adsorption of $Ce^{+3}$and Gd^{+3} reached 174.05 and 223.79 mg/g. The hydrophilic functional groups such as $–NH_2$, –OH, –COOH, $–SO_3–$ are assisting in the adsorption process. Gautam Sen et al. [69] reported microwave assisted graft polymerisation and crosslink network of polyvinyl pyrrolidone and acrylamide based hydrogel and used for removal of chromium Cr(VI) in presence of a nutritional element of calcium and magnesium.

Nuclear energy is under development area to counter the increasing energy demand of the world. It has also taken into alternative energy sources by undertaking the earth's environment and shortage of conventional energy sources. At that time, a large amount of radioactive waste released into an environment which threats to human society and the environment. For safety of human community and the environment, removal of radioactive element from wastewater is important [70]. Jun Waang et al. prepared hydrogel based on graphene oxide/amidoxime by mixing the dispersions of both components and explored its application as an adsorbent for uranium (VI). The selective sorption of uranium examined in the presence of cations such as Mg^{2+}, Ca^{2+}, Ba^{2+} and Sr^{2+} and selective sorption taking in part because of available chelating group in the network [71]. K.S.V. Krishnarao et al. [72] reported construction of multi-chelating hydrogels by utilization of N-(2-aminothiozolyl) maleamic acid and acrylamide, hydroxyethyl acrylate and polymerization by using free radical polymerization. N-(2-aminothiozolyl) maleamic acid was formed by reaction of maleamic acid with 2-amino thiazole. The maximum uptake capacity for Uranium (VI) ion was 288mgg-1. 137Cs is hazardous radioactive element and having a half-life of 30.17 years. J.L. Wang et al. [73] prepared the magnetic chitosan beads crosslinked with glutaraldehyde and investigated for the removal of cesium. Nitrogen atoms present in the magnetic beads responsible for the removal of Cs^{+} because the strong electrostatic interaction.

(II) Removal of dyes

Dyes are used as in the different technology such as rubber, plastics, printing, leather tanning, paper production, cosmetics, etc. Around more than 10,000 dyes are commercially accessible and over 7 x 105 tonnes of dyes formulated annually. It was assessed that 2% colors delivered yearly are released in effluents from associated industries [74,75]. Discharge of wastewater containing dyes in natural water bodies without any treatment and it causes the problem for human health, aquatic animal, and the environment. It is basic to treat dye effluents before released into the earth environment. Dendritic polymers are able to trap the small molecule and hyper branched polyglycerol (HPG) is a typical dendritic polymer, retains the aliphatic backbone. Wenjun Fang et al. [76] reported hydrogel based on the methacrylated hyper branched polyglycerol and poly acrylic acid prepared by free radical polymerization and explored its application for the removal of cationic dye. HPG/PAA hydrogels were highly hydrophilic and stable at high temperature and wide range of pH 1-11. HPG/PAA hydrogels were removed 98.33% of cationic dye at 50 mg/L initial concentration of dye. Adsorption and photocatalysis mechanism are responsible for the removal of dyes by hydrogel technology. A summary of hydrogel and their application and mechanism of removal listed below in table 2.

J. J. Feng et al. [77] used the green assembly method to develop the graphene with glutathione and surface modified by nitrogen and sulphur doping. A glutathione is a natural antioxidant and possess multi-functional groups. A glutathione acts as a binder and reducing agent. Glutathione enhanced the distance between the reduced graphene sheets, assemble in a parallel manner and form of three-dimensional porous network. Graphene hydrogel investigated for the adsorption of three dyes, malachite green, crystal violet, and methylene blue.

Table 2. Hydrogels, Application of removal of dye, and Mechanism of removal.

Sr. No	Hydrogel	Application of removal of dye	Mechanism of removal	Ref.
1.	HPG/Poly acrylic acid	Removal of Methyl violet	Adsorption	[75]
2.	Chitosan/montmorillonite hydrogel	Removal of nitrazine dye	Adsorption	[76]
3.	Starch/poly(alginic acid-cl-acrylamide) nanohydrogel	Removal of coomassie brilliant blue R-250 dye	Adsorption	[77]
4.	TiO_2/calcium alginate	Methyl orange	Adsorption and photocatalysis	[78]
5.	Iron-doped titania/silane	Victoria blue	Adsorption and photocatalysis	[79]

6.	Reducing graphene/polyaniline/cuprous oxide	Congo red	Adsorption and photocatalysis	[80]
7.	TiO_2/Chitosan	Azo and Anthraquinone Dye	Adsorption and photocatalysis	[81]
8.	Titanium dioxide/polyacrylamide	Removal of methylene blue	Photocatalysis	[83]

Hydrogels displayed the adsorption of all three dyes due to porous structure and surface modification with heteroatoms. The adsorption capacity of hydrogel was stronger for all the dyes with in an increasing order: malachite green, methylene blue, crystal violet. Run fang et al. [78] was prepared cationic hydrogel by two methods. First, solution polycondensation method used to form the water-soluble cationic prepolymer by utilizing the triethylenetetramine, acetone and formaldehyde and crosslinked with glutaraldehyde to form the hydrogel. The contribution of surface adsorption to the total adsorption capacity was minor because internal diffusion plays an even more important role. The high cationic charge density and consuming abundant functional groups in structured network are responsible for the high adsorption capacity.

Hydrogel-clay composite received considerable attention in separation technology. Mu. Ufukyildiz et al. [79] prepared chitosan/MMT based hydrogel composite beads and crosslinked with sodium tripolyphosphateas a coagulant for ionic crosslinking and chemically crosslinked with glutaraldehyde. Hydrogel contain the functional group like $–NH_2$ and –OH groups that provide the specific sites for the adsorption of nitrazine dyes. Temperature also effect on the adsorption, when temperature increasing from 293 K to 313 K then adsorption decreased. The adsorption capacity was 185.69 mg/g for dye. Mu. Naushad et al.[80] prepared nano hydrogel using starch, poly alginic acid, poly acrylamide and investigated for the removal of anionic dye coomassie brilliant (CB) blue R-250. The adsorption of dye is depends on the pH and temperature types of variable. The higher pH value is retard the interaction of metal ion then reduced the adsorption of dye molecule and The higher adsorption observed at 55 0C. Foadkazemi et al. [81] reported hydrogel prepared with inorganic metal nanoparticles as an initiator. Hydrogel developed with an acrylamide monomer and TiO_2 P25 NPs as an initiator, and N, N, methylene bisacrylamide as a crosslinker in aqueous media under sunlight. The hydrogel displayed the photocatalytic degradation performance with 95.00% degradation of methylene blue after 5 h. D. Chen et al. [82] prepared hydrogel based on the 3D structure of Bi_2WO_6/graphene by one-step hydrothermal method and applied to the environment remediation. The amount of graphene has an influence on the adsorption and photocatalytic performance. Both the adsorption and photocatalytic improved with increasing the the percentage of graphene in the hydrogels. The hydrogel has capacity to remove 36.1 % of methylene blue.Yongfazhu et al. [83] reported hybrid gel with carbon nitride and SiO_2. Hybrid hydrogel investigated dually adsorption and in-situ photocatalytic degradation of coking wastewater, phenol, and methylene blue. Hybrid structures constructed with the carbon nitride/SiO2. Hybrid hydrogel prepared by alkali-solution and acid-gel process. Hybrid gel reduced 33% of total organic carbon content and exhibited good removal efficiency for phenol and methylene blue.

To avoid the separation of catalyst and its regeneration, Y. Zhu et al. [86] reported hydrogel based on the polyaniline/carbon nitride nanosheets composite hydrogel and investigated for the photocatalysis degradation of methylene blue. In situ polymerization technique used to form 3D hierarchical nanostructure of polyaniline/carbon nitride hydrogel. Carbon nitride nano sheets put an impact on the photocatalysis of pollutant because it has high surface area, interfacial charge separation, and longer lifetime. In removal mechanism of methylene blue, first they adsorbed on the hydrogel then in situ photocatalysis of methylene blue occurred. polyaniline/carbon nitride removed 92.5% of methylene blue at 4 h. F. Khan et al. [87] developed hydrogels nanocomposite hydrogels of alginate/carboxymethyl cellulose cross linked with the encapulation of TiO_2-NPs by using dissipative convective method followed by freeze-drying. The photocatalytic performance of hybrid examined with congo red dye under direct sunlight. The nano sized material incorporated hydrogel displayed 91.5 % of removal efficiency.

Hydrogels, application of photocatalysis of pollutant, source of light and their removal capacity are given in table 3.

Table 3. Hydrogels, Photocatalysis of pollutant, Source of light, and removal capacity.

Hydrogels	Pollutant	Source of light	Removal capacity	Ref.
Chitosan/poly(vinylalcohol)/ TiO_2	Acid red 14	40 W fluorescent light source	83.8%.	[84]
TiO_2-rGO-PDMAA	Methylene blue	300 W Xe lamp	100 %	[85]
Titanate–Graphene oxide	Methyle orange	375 W high-pressure mercury lamp	100%	[90]
Polyaniline /TiO_2	Bisphenol A	500-W Mercury lamp	100 %	[91]
Ag_2O / Sodium alginate	Methylene blue and malachite green	250 W high pressure mercury lamp	93% and 93 %	[92]
AgBr@rGO	Bisphenol A	400-W metal halide lamp	100 %	[93]
TiO_2/agarose	Methylene blue	6 W Vilber Lourmat lamp	100 %	[94]
Silica hydrogel/ α-Fe_2O_3 nanosheets	Bisphenol S	300W xenon lamp	91 %	[95]

Zhang et al. [88] developed of titanium dioxide nanoparticle and grapheme based hydrogel composite and applied for the removal of low-concentration bisphenol A. Mainly, adsorption and photocatalysis are phenomena responsible for the removal of the bisphenol A. The removal efficiency of the TiO_2-rGO electrode was higher than thin film electrode. The removal efficiency of the TiO_2-rGO hydrogel electrode was 96% at the end of 4 h with 20 mg/L concentration of bisphenol. A.R. Balasubramanian et al. [89] investigated the effect of doping on the graphene hydrogel and efficiency to remove pollutant. Doped graphene hydrogel prepared by using nitrogen as dopant with different concentration through a simple one-pot hydrothermal reaction and evaluated their efficiency for the adsorption and photocatalysis of Acridine orange. Author claimed that they were successfully introduced nitrogen atom in graphene hydrogel. It observed that there was any direct effect of nitrogen on the photocatalysis of Acridine orange but hydrogel with 6.78 % of a nitrogen content exhibited the higher surface area and displayed the highest photocatalytic activity due to higher adsorption and after photocatalysis of Acridine orange.

D. Chen et al. [99] reported establishment of hydrogel with new approach based on ethylene glycol, Fe-octacarboxylic acid phthalocyanine, nanomagnetic attapulgite. The Rhodamine B dye decomposition carried out by using hydrogel nanocomposite investigated for photodegradation of Rhodamine B dye. Means while incorporation of Fe-octacarboxylic acid phthalocyanine/magnetic attapulgite into hydrogel matrix that facilitates the interaction between them and provides new crosslinking junctions. However, temperature, pH, and magnetic responsive additive incorporated in the hydrogel matrix has an effect on the removal of Rhodamine B. At lower pH values of feed solution, hydrogel displayed the higher removal of dye and the photodegradation efficiency increases with higher magnetic field. In lower initial concentration, photodegradation process was faster and removal efficiency of 95% achieved at the end of 3 h. G. Sharma et al. [100] reported hydrogel based on the starch/poly(alginic acid-cl-acrylamide)/Fe/Zn nanocomposite hydrogel and applied for the photocatalytic degradation of mixture of dyes. Hydrogels composite prepared by polymerization/co-precipitation method and degradation of a mixture of dyes takes place with adsorption and adsorption photocatalysis. The hydrogel exhibited the 91% of malachite green and 82% of the fast green dye were degraded at the end of 5 h.

W. Cui et al. and his co-workers [101] reported reduced graphene oxide core shell with Ag and AgCl nanoparticles and nanoparticle containing core-shell incorporated in reduced graphene hydrogel and explored its ability to remove bisphenol A. The synergistic mechanism of adsorption and photocatalysis involved in the removal of bisaphenol. The removal efficiency of hydrogel was 93.7% for bisaphenol A. Yongfazhuet al. [102] demonstrated the silver phosphate and graphene based hydrogel for efficient degradation of bisphenol A under visible-light irradiation with the synergy of adsorption and photocatalysis. Silver phosphate/graphene based hydrogel displayed the good removal efficiency of bisphenol A, 100% removal of bisphenol-A achieved at the end of 60h.

(III) Removal of anions

When anionic species present above a certain level in the water and considered as the pollutants and their bioaccumulation become an environmental concern. At the point when their fixations increment in the earth start to influence the human wellbeing and ecosystem [103]. Liang Ni et al. [104] prepared the cationic hydrogel based on the 1-allyl-3-methylimidazolium chloride and acrylamide by using the solution polymerisation and applied for the removal of the permanganate anion. The cationic character of hydrogel is due to the acrylamide. Hydrogel with 10 wt.% of AMIMCl exhibited the higher adsorption capacity of permanganate anion and equilibrium adsorption achieved in 5 min. Rahini et al. [105] reported the removal of phosphate by chemically modification bentonite derived parent hydrogel. The modification of hydrogel executed with surfactant and ion and mixed ion solutions. The mixed ion solution modified hydrogel showed the maximum adsorption capacity and the rewoquate surfactant modified hydrogel showed the lower adsorption capacity of phosphate ion.

Su et al. [106] reported semi-IPNs hydrogel prepared based on the feather protein modified by potassium acrylate and poly vinyl by graft copolymerization technique and investigated the adsorption of NH^{+4} and PO_3^{-4}. Adsorption was pH dependent and maximum adsorption was found at pH 6. Zhu and his coworkers [107] reported the allylamine hydrochloride-based porous hydrogel for reducing the serum phosphorus level to control hyperphosphatemia without calcium and aluminium preparations. Allylamine hydrochloride and N,N-methylene bis-acrylamide as monomer based hydrogel was prepared by free radical polymerization and primary and secondary amine both possess the binding ability with phosphate but primary amine possess the strong binding ability than secondary amine.

Genc et al. [108] investigated the adsorption of phosphate by hydrogel based on poly N-isopropylacrylamide/aluminium alginate IPN structure. Result of adsorption study revealed that at pH 3, maximum adsorption occurred and above pH 3, adsorption was lower. Adsorption of phosphate also investigated in presence of SO_4^{2-}, Cl- and NO_3^- ion and it was observed that SO_4^{2-} ions effects on the adsorption phosphate. The phosphate ions removal capacity of hydrogel remains with five consecutive cycles. Zhang et al. [109] reported quaternized hydrogel based on 1-vinylimidazole for adsorption of anions. The polymerisation of 1-vinylimidazole carried out by utilizing the free radical mechanism and quaternized by using the dimethylsulfate. The adsorption of anions such as fluoride, nitrite, bromide, phosphate, sulphate investigated and among all of these ions, maximum adsorption of phosphate ions takes place around 98.2% and minimum adsorption of fluoride around 24.7%. Hanna Siwek et al. [110] prepared alginate/FeCl3 based hydrogel microcapsule and studied the adsorption of phosphate from its solution and natural water. Hydrogel microcapsules have formed by using polyelectrolyte complex-gelation method with droplets of aquatic solution of polysaccharide (alginate) with gelation salt. The rate of adsorption of phosphate ions by the alginate/Fe (III) capsules higher in natural water than from their solution in distilled water. The phosphate ions removed by a combination of two processes, first Adsorption of phosphate ion on a hydrogel and another precipitation of phosphorous with Fe^{+3}. The average removal of phosphate ion was 87.5%. Dong et al. [111] reported the lanthanum-loaded magnetic cationic hydrogel and scrutinized for the de-fluoridation of water. The loading the lanthanide carried out by impregnating the hydrogel in the saturated lanthanum nitrate solution. The process of de-fluoridation was highly pH depended and electrostatic interaction, ligand exchange, and Lewis acid-base interaction play vital role in the adsorption. The maximum adsorption capacity of hydrogel was 93% of fluoride.

(IV) Removal of pesticides and ciprofloxacin

Tiwarialka et al. [112] prepared the super paramagnetic nanoiron oxide loaded copolymer hydrogel based on the styrene and acrylic acid as a monomer. The nanocomposites hydrogel reconnoiter for the removal of dichlorvos. The higher adsorption of pesticide observed higher at pH 4 but reduction in adsorption observed at higher pH because at higher pH electrostatic repulsion between adsorbent and adsorbate. The maximum uptake up to 80.16% achieved in 60 minutes. C. Dwivedi et al. [113] reported

the gold nanoparticle embedded on hydrogel beads constructed by cross-linked chitosan for the removal of methyl parathion. Composites beads were prepared in the spherical form to avoid the irregular size and lower mechanical strength. Chitosan served the as stabilizer for gold nanoparticle and binding matrix for hydrogel beads. Surface area and pore radius of composite hydrogel was $2.67 m^2 g^{-1}$ and 17.25 nm. Surface area of nanoparticle composite hydrogel was higher than blank chitosan gel, which enhanced the adsorption of methyl parathion on nanoparticle composite hydrogel bead as compared to blank chitosan hydrogel.
Antibiotic received consideration as a pollutant in aquatic system and they have a direct natural activity on organisms, unlike many other pollutants [114]. Ciprofloxacin hydrochloride, one of the commonly used fluoroquinolones, it was found in wastewater and surface water [115]. It is very toxic to some organism even at low concentrations. Thus, removal of ciprofloxacin is very important. Yuan Zhuang and his co-workers [116] reported TiO2 nanotube/reduced graphene oxide based hydrogel for the removal of ciprofloxacin. Adsorption of ciprofloxacin occurred by physical and chemical mechanism. The maximum adsorption capacity of hydrogel composite for the ciprofloxacin was 181.8 mg/g. J. Ma et al. [117] prepared three-dimensional porous graphene hydrogel by using hydrothermal reduction method with ascorbic acid as a reducing agent and explored its application for the removal of ciprofloxacin. The porous hydrogel is able to adsorb maximum amount of ciprofloxacin at adsorption of at higher pH 8. The hydrogel consuming the positive charged at pH 8 and cationic form of ciprofloxacin attract towards the hydrogel. Therefore, the higher the electrostatic repulsion effect repelled the adsorption of ciprofloxacin. The maximum adsorption of ciprofloxacin was 235.6 mg/g.

VI. Conclusion

This review provides up to date account of recent developments in the field of hydrogel and hydrogel composites used for wastewater treatment. Classification, methods of synthesis, and recent development in hydrogels for wastewater treatment reviewed. In the last few years, hydrogels received the greater attention due to their exceptional properties for wastewater treatment. To increase the efficiency of hydrogels for removal of pollutant, development takes place in the hydrogels preparation and new strategies applied to introduce new chelating functional groups in hydrogel for adsorption of pollutant. The wealth of research into hydrogel composite formulations for wastewater treatment applications demonstrates the great potential. Compared to the conventional hydrogels, composite and nano-composites hydrogel has remarkable properties. Still, hydrogel technology needs to develop in terms of selectivity of pollutant and reuse of hydrogels.

VII. References

[1] Bora, T., and Dutta, J. 2014 Applications of nanotechnology in wastewater treatment—A Review, Journal of Nanoscience and Nanotechnology. 14(1):613–626.

[2] Seow, T.W., Lim, C.K., Md Nor, M.H., Mubarak, M.F.M., Lam, C.Y., Yahya, A., and Ibrahim, Z. 2016 Review on wastewater treatment technologies,International Journal of Applied Environmental Sciences,11(1): 111-126,.

[3] Yildiz, U., Kemik, O.F, and Hazer, B. 2010 The removal of heavy metal ions from aqueous solutions by novel pH-sensitive hydrogels. Journal of Hazardous Materials, 183(1):521–532.

[4] Zohuriaan-Mehr, M.J.,. Kabiri, K. 2008 Superabsorbent polymer materials: A Review, Iranian Polymer Journal, 17(6): 451-477.

[5] Hennink, W.E., and van Nostrum, C.F. 2002 Novel crosslinking methods to design hydrogels, Advanced Drug Delivery Reviews, 54: 13–36.

[6] Solpan, D., Duran, Saraydin, S.D., and Guven, O. 2003 Adsorption of methyl violet in aqueous solutions by poly(acrylamide-co-acrylic acid) hydrogels, Radiation Physics and Chemistry, 66:117–127.

[7] Ozkahraman, B., Acar, I., and Emik, S. 2011 Removal of cationic dyes from aqueous solutions with poly (N-isopropylacrylamide-co-itaconic acid) hydrogels, Polymer Bulletin, 66:551–570.

[8] Abdel-Aal, S.E., Hegazy, E.A., AbouTaleb, M.F.,and Dessouki, A.M. 2008. Radiation synthesis and characterization of 2-hydroxyethyl-methacrylate-based hydrogels containing di- and tri-protic acid and its application on wastewater treatment, Journal of Applied Polymer Science, 107:1759–1776.

[9] Shawky, H.A., El-Sayed, M.H., El-Hag Ali, A., Abdel Mottale, M.S. 2006. Treatment of polluted water resources using reactive polymeric hydrogel, Journal of Applied Polymer Science. 100:3966 –3973.

[10] Zheng, Y., Liua, Y., and Wang, A. 2011.Fast removal of ammonium ion using a hydrogel optimized with response surface methodology, Chemical Engineering Journal, 17:1201–1208.

[11] Li, Y., and Wang, L., 2016. Removing organic dyes by using a small peptide hydrogel, Chemistry letters, 45:1253–1255.

[12] Kurdtabar, M., Kermani, Z.P., and Marandi, G.B. 2015. Synthesis and characterization of collagen-based hydrogel nanocomposites for adsorption of Cd^{2+}, Pb^{2+}, methylene green and crystal violet, Iranian Polymer Journal, 24:791–803.

[13] Ekebafe, L.O., Ogbeifun D. E., and Okieimen, F.E. 2015. Removal of heavy metals from aqueous media using native cassava starch hydrogel, African Journal of Environmental Science and Technology, 6:275-282.

[14] Inal, M., and Erduran, N. 2015. Removal of various anionic dyes using sodium alginates/poly(N-vinyl-2-pyrrolidone) blend hydrogel beads, Polymer Bulletin, 72:1735-1752.

[15] Ali, I., and Gupta, V. K. 2007 Advances in water treatment by adsorption technology, Nature Protocols, 1:2661-2667.

[16] Fu, F., and Wang, Q. 2011 Removal of heavy metal ions from wastewaters: A review, Journal of Environmental Management, 92:407-418.

[17] Ringot, D., Lerzy, B., Chaplain, K., Bonhoure, J.P., Auclair, E., and Larondelle, Y. 2007. In vitro absorption of ochratoxin A on the yeast industry by-products: Comparison of isotherm models, Bioresource Technology, 98:1812–1821.

[18] Dabrowsk, A. 2001 Adsorption from theory to practice, Advances in Colloid and Interface Science, 93: 135-224.

[19] Saadi, R., Saadi, Z., R. Fazaeli, R., Fared, N.E. 2015. Monolayer and multilayer adsorption isotherm models for sorption from aqueous media, Korean Journal of Chemical Engineering, 32(5):787-799.

[20] Vandenbruwane, J., Neve, S.D., Qualls, R.G., Sleutel, S., and Hofman, G. 2007 Comparison of different isotherm models of dissolved organic carbon (DOC) and nitrogen (DON) sorption to mineral soil, Geoderma, 139:144–153.

[21] Patiha, Heraldy, E., Hidayat, Y., and Firdaus, M., 2016. The Langmuir isotherm adsorption equation: The monolayer approach, Materials Science and Engineering, 107 (1): 012067.

[22] Dehmolaei, A., Vadi, M., 2014. Comparative study of isotherms adsorption of B_{12} by single-wall carbon nanotube and multi-wall carbon nanotube, Orient. J. Chem, 30:265-269.

[23] Kono, H. 2015. Preparation and Characterization of Amphoteric Cellulose Hydrogels as Adsorbents for the Anionic Dyes in Aqueous Solutions, Gels, 1:94-116.

[24] Fytianos, K., Voudrias, E., and Kokkalis, E.2000. Sorption-desorption behavior of 2,4-dichlorophenol by marine sediments, Chemosphere, 40:3-6.

[25] Rauf, M.A., Bukallah, S.B., Hamour, F.A., and Nasir, A.S. 2008 Adsorption of dyes from aqueous solutions onto sand and their kinetic behavior, Chemical Engineering Journal, 137:238–243.

[26] Gregg, S.J., Sing, K.S.W., 1982 Adsorption, surface area, and porosity, Academic Press, London.

[27] Do, D.D., 1998. Adsorption analysis: equilibria and kinetics. Imperial College Press London.

[28] Coutrin, N.P., Altenor, S., and Gaspard, S. 2009 Assessment of the surface area occupied by molecules on activated carbon from liquid phase adsorption data from a combination of the BET and the Freundlich theories, Journal of Colloid and Interface Science, 332:515–519.

[29] Kasgoz, H., Durmus¸ A., Kasgoz, A. 2008. Enhanced swelling and adsorption properties of AAm-AMPSNa/clay hydrogel nanocomposites for heavy metal ion removal, Polymer for advance technologies, 19:213–220.

[30] Gad, Y.H. 2008 Preparation and characterization of poly(2-acrylamido-2-methylpropanesulfonic acid)/Chitosan hydrogel using gamma irradiation and its application in wastewater treatment, Radiation Physics and Chemistry, 77:1101– 1107.

[31] Ozay, O., Ekici, S., Baran, Y., Aktas, N., Sahiner, N. 2009 Removal of toxic metal ions with magnetic hydrogels, Water research, 43:4403–4411.

[32] Zhou, Y., Fu, S., Liu, H., Yang, S., and Zhan, H. 2011. Removal of methylene blue dyes From wastewater using cellulose-based superadsorbenthydrogels, Polymer Engineering And Science, 51(12):2417–2424.

[33] Aouad, F.A., Pan, Z., Orts, W.J., and Mattoso, L.H.C. 2009. Removal of paraquat pesticide from aqueous solutions using a novel adsorbent material based on polyacrylamide and methylcellulose hydrogels, Journal of Applied Polymer Science, 114:2139–2148.

[34] Kioussis, D.R., Smith, D.F., and kofinas, P. 2001 Ammonium perchlorate–binding poly(allylamine hydrochloride) hydrogels for wastewater remediation, Journal of Applied Polymer Science, 80:2073–2083.

[35] Pan, G., Kurumada, K.I., and Yamada, Y. 2008. Application of hydrogel for the removal of pollutant phenol in water, Journal of the Chinese Institute of Chemical Engineers, 39:361–366.

[36] Zendehdel, M., Barati, A., Alikhani, H., and Hekmat, A. 2010. Removal of methylene blue dye from wastewater by adsorption onto semi-inpenetrating polymer network hydrogels composed of acrylamide and acrylic acid copolymer and polyvinyl Alcohol, Iranian Journal of Environmental Health Science & Engineering, 7(5): 423-428.

[37] Yildiz, U., Kemika, O.F., and Hazer, B. 2010. The removal of heavy metal ions from aqueous solutions by novel pH-sensitive hydrogels, Journal of Hazardous Materials, 183 (2010) 521–532.

[38] Sokker, H.H., El-Sawy, N.M., Hassan, M.A., and El-Anadouli, B.E. 2011. Adsorption of crude oil from aqueous solution by hydrogel of chitosan based polyacrylamide prepared by radiation induced graft polymerization, Journal of Hazardous Materials, 190:359–365.

[39] Liu, Y., Wang, W., and Wang, A. 2010. Adsorption of lead ions from aqueous solution by using carboxymethyl cellulose-g-poly (acrylic acid)/attapulgite hydrogel composites, Desalination, 259: 258–264.

[40] Zheng, Y., Xie, Y., Wang, A. 2012. Rapid and wide pH-independent ammonium-nitrogen removal using a composite hydrogel with three-dimensional networks, Chemical Engineering Journal, 179:90– 98.

[41] Zheng, Y.,and Wang, A. 2010. Nitrate Adsorption using poly(dimethyl diallyl ammonium chloride)/polyacrylamide hydrogel, Journal of Chemical & Engineering Data, 55:3494–3500.

[42] Maziad, N. A., Mohsen, M., Gomaa, E., and Mohammed, R. 2015. Radiation copolymerization of hydrogels based in polyacrylicacid/polyvinyl alcohol applied in water treatment Processes, Journal of Materials Science and Engineering A, 5 (11-12):381-390.

[43] Samandari, S.S., and Gazi, M. 2013. Cellulose-graft-polyacrylamide/ hydroxyapatite composite hydrogel with Possible Application in Removal of Cu (II) Ions, Reactive & Functional Polymers, 73:1523-1530.

[44] Patel, Y.N. and Patel, M.P. 2013. Adsorption of azo dyes from water by new poly (3-acrylamidopropyl)-trimethylammonium chloride-co-N,N-dimethylacrylamide superabsorbent hydrogel—Equilibrium and kinetic studies, Journal of Environmental Chemical Engineering, 1:1368–1374.

[45] Sun, X.F., Ye, Q., Jing, Z., and Li, Y. 2014. Preparation of hemicellulose-g-Poly(methacrylic acid)/ Carbon Nanotube Composite Hydrogel and Adsorption Properties, Polymer Composites, 35(1):45–52.

[46] Ali, A.E. 2012. Removal of heavy metals from model wastewater by using carboxymehylcellulose/2-Acrylamido-2-methyl Propane Sulfonic Acid Hydrogels, Journal of Applied Polymer Science, 123:763–769.

[47] Li, Z., Wang, Y., Wu, N., Chen, Q., and Wu, K. 2013. Removal of heavy metal ions from wastewater by a novel HEA/AMPS copolymer hydrogel: preparation, characterization, and mechanism, Environmental Science and Pollution Research, 20:1511–1525.

[48] Liu, Y., Liu, Y., Cao, X., Hua, R., Wang, Y., and Li, Q. 2014. Temperature swing adsorption of uranium (VI) ions on Poly(N-isopropylacry-lamide)/chitosan Semi-IPN, International Journal of Nuclear Energy Science and Engineering, 1(1):8-14.

[49] Liu, Y., Zheng, Y., and Wang, A. 2010. Enhanced adsorption of Methylene Blue from aqueous solution by chitosan-g-poly (acrylic acid)/vermiculite hydrogel composites, Journal of Environmental Sciences, 22(4):486–493.

[50] An B., Lee, H., Lee, S., Lee, S.H., and Choi, J.W. 2015. Determining the selectivity of divalent metal cations for the carboxylgroup of alginate hydrogel beads during competitive sorption, Journal of Hazardous Materials, 298:11–18.

[51] Kumar, R., Jain, S.K., Verma, S., and Malodia, P. 2015. Mercapto functionalized silica entrapped polyacrylamide hydrogel: Arsenic adsorption behaviour from aqueous solution, Journal of Colloid and Interface Science, 456:241–245.

[52] Borai, E.H., Hamed, M.G., El-kamash, A.M., Siyam, T., and El-Sayed, G.O. 2015. Template polymerization synthesis of hydrogel and silica composite for sorption of some rare earth elements, Journal of Colloid and Interface Science, 456:228–240.

[53] Varghese, L.R., and Das, N. 2015 Removal of Hg (II) ions from aqueous environment using glutaraldehyde cross linked nano biocomposite hydrogel modified by TETA and ß-cyclodextrin: Optimization, equilibrium, kinetic and ex situ studies, Ecological Engineering. 85: 201–211.

[54] Zhu, Q., and Li, Z. (2015) Hydrogel-supported nanosized hydrous manganese dioxide: Synthesis, characterization, and adsorption behavior study for Pb^{2+}, Cu^{2+}, Cd^{2+} and Ni^{2+} removal from water, Chemical Engineering Journal, 281: 69–80.

[55] Jamnongkan, T., and Singcharoen, K. 2016. Towards novel adsorbents: the ratio of PVA/chitosan blended hydrogels on the copper (II) ion adsorption, Energy Procedia, 89: 299 – 306.

[56] Zhu, Y., Zheng, Y. Wang, F., and Wang, A. 2016. Fabrication of magnetic macroporous chitosan-g-poly (acrylic acid) hydrogel for removal of $Cd^{2+}$and Pb^{2+} International Journal of Biological Macromolecules, 93:483–492.

[57] Basri, S.N., Zainuddin, N., Hashim, K., and Yusof, N.A. 2016. Preparation and characterization of irradiated carboxymethyl sago starch-acid hydrogel and its application as metal scavenger in aqueous solution, Carbohydrate Polymers, 138:34–40.

[58] He, S., Zhang, F., Cheng, S., and Wang, W. 2016. Synthesis of sodium acrylate and acrylamide copolymer/ GO hydrogels and their effective adsorption for Pb^{2+} and Cd^{2+}, ACS Sustainable Chemistry & Engineering, 4(7):3948–3959.

[59] Luo, F., Chen, Z., Megharaj, M., and Naidu, R., Simultaneous removal of trichloroethylene and hexavalent chromium by green synthesized agarose-Fe nanoparticles hydrogel, Chemical Engineering Journal, 294: 290–297.

[60] Ma, J., Zhou, G., Chu, L., Liu, Y., Liu, C., Luo, S., and Wei, Y. 2017 Efficient removal of heavy metal ions with EDTA functionalized chitosan/polyacrylamide double network hydrogel, ACS Sustainable Chemistry & Engineering, 5(1):843–851.

[61] Sahraei, R., Pour, Z.S., and Ghaemy, M. 2017. Novel magnetic bio-sorbent hydrogel beads based on modified gum tragacanth/graphene oxide: Removal of heavy metals and dyes from water, Journal of Cleaner Production, 142:2973-2984.

[62] Wang, L.Y., and Wang, M.J. 2016. Removal of heavy metal ions by poly(vinyl alcohol) and carboxymethyl cellulose composite hydrogels prepared by a freeze−thaw method, ACS Sustainable Chemistry & Engineering, 4(5): 2830–2837.

[63] Zhu, Y., Zheng, Y., Wang, F., and Wang, A. 2016 Monolithic supermacroporous hydrogel prepared from high internal phase emulsions (HIPEs) for fast removal of Cu^{2+} and Pb^{2+},Chemical Engineering Journal, 284: 422–430.

[64] Yu, P., Wang, H.Q., Bao, R.Y., Liu, Z., Yang, W., Xie, B.H., and Yang, M.B. 2017 Self-assembled sponge-like chitosan/reduced graphene oxide /montmorillonite composite hydrogels without crosslinking of chitosan for effective cr(vi) sorption, Acs Sustainable Chemistry & Engineering, 5(2):1557–1566.

[65] Zhang, Y., He, F., and Li, X. 2016. Three-dimensional composite hydrogel based on polyamine zirconium oxide, alginate and tannic acid with high performance for Pb(II), Hg(II) and Cr(VI) trapping, Journal of the Taiwan Institute of Chemical Engineers, 00 (2016) 1–8.

[66] Zhou, G., Luo, J., Liu, C., Chu, L., Ma, J., Tang, Y., Zeng, Z., Luo, S. 2016. A highly efficient polyampholyte hydrogel sorbent based fixed-bed process for heavy metal removal in actual industrial effluent, Water Research, 89:151-160.

[67] Zhao, Z., Chen, H., Zhang, H., Ma, H., Wang, Z. 2017 Polyacrylamide-phytic acid-polydopamine conducting porous hydrogel for rapid detection and removal of copper (II) ions, Biosensors and Bioelectronics, 91:306–312.

[68]Feng, W., Wenbo, W., Yongfeng, Z., Aiqin, W. 2017. Evaluation of Ce(III) and Gd(III) adsorption from aqueous solution using CTS-g-(AA-co-SS)/ISC hybrid hydrogel adsorbent, Journal of Rare Earths, 35(7): 697-708.

[69] Pal, P., Pandey, J.P., Sen, G., Modified PVP based hydrogel: Synthesis, characterization and application in selective abstraction of metal ions from water, Materials Chemistry and Physics, 194: 261-273.

[70] Yu, S., Wang, X., Tan, X., and Wang, X. 2015. Sorption of radionuclides from aqueous system onto graphene oxide-based materials: A review, Inorganic Chemistry Frontiers, 2:593-612.

[71] Wang, F., Li, H., Liu, Q., Li, Z., Li, R., Zhang, H., Liu, L., Emelchenko, G. A., Wang, J. 2016. A graphene oxide/amidoxime hydrogel for enhanced uranium capture, Scientific Reports. 6:19367(1-8).

[72] Reddy, N.S., Rao, K.S.V.K., Rao, K.M., Parambadath, S., Ha, C.S. 2016. Aminothiozolyl maleamic acid based multi chelating hydrogels for the separation of uranium (VI) ions from aqueous environment, Polymers for Advanced Technologies, 27(10):1317-1324.

[73] Chen, Y.W., and Wang, J.L. 2016. Removal of cesium from radioactive wastewater using magnetic chitosan beads cross-linked with glutaraldehyde ,NuclScience and Techniques, 27(2): 43.

[74] Allen, S.J., Koumanova, B. 2005. Decolourisation of water/wastewater using adsorption**,** Journal of the University of Chemical Technology and Metallurgy 40(3):175-192.

[75] Bhatti, K.S., and Ramesh, S.T. 2013. Removal of dyes using waste as low-cost adsorbents: a review, Applied Water Science, 3(4):733-790.

[76] Ying, H., He, G., Zhang, L., Lei, Q., Guo, Y., and Fang, W. 2016. Hyper branch edpolyglycerol/poly(acrylic acid) hydrogel for the efficient removal of methyl violet from aqueous solutions, Journal of Applied Polymer Science, 133(5) (2016) 1-11.

[77] Shi, Y.C., Wang, A.J., Wu, X.L., Chen, J.R., and Feng, J.J. 2016. Green-assembly of three-dimensional porous graphene hydrogels for efficient removal of organic dyes, Journal of Colloid and Interface Science, 484:254–262.

[78] Fang, R., He, W., Xue, H., and Chen, W. 2016. Synthesis and characterization of a high-capacity cationic hydrogel adsorbent and its application in the removal of Acid Black 1 from aqueous solution, Reactive and Functional Polymers, 102:1–10.

[79] Ngwabebhoh, F.A., Erdem, A., and Yildiz, U. 2016. Synergistic removal of Cu(II) and nitrazine yellow dye using an eco-friendly chitosan-montmorillonite hydrogel: Optimization by response surface methodology, Journal applied polymer science, 133(29):1-4.

[80] Sharma, G., Naushad, M., Kumar, A., Rana, S., Sharma, S., Bhatnagar, A., Stadlerd, F.J., Ghfar, A.A., and Khan, M.R. 2017. Efficient removal of coomassie brilliant blue R-250 dye using starch/poly(alginic acid-*cl*-acrylamide) nanohydrogel, Process Safety and Environmental Protection, 109:301–310.

[81] Zhao, K., Feng, L., Lin, H., Fu, Y., Lin, B., Cui, W., Li, S., and Wei, J. 2014. Adsorption and photocatalytic degradation of methyl orange imprinted composite membranes using TiO_2/calcium alginate hydrogel as matrix, Catalysis Today, 236(1):127–134.

[82] Anirudhan, T.S., Divya, P.L., Nima, J., and Sandeep, S. 2014. Synthesis and evaluation of Iron-doped titania/silane based hydrogel for the adsorptional photocatalytic degradation of Victoria blue under visible light, Journal of Colloid and Interface Science, 434:48–58.

[83] Miao, J., Xie, A., Li, S., Huang, F., Cao, J., and Shen, Y. 2015. A Novel reducing graphene/polyaniline/cuprous oxide Composite Hydrogel with Unexcepted Photocatalytic Activity for the Degradation of Congo red, Applied Surface Science, http://dx.doi.org/10.1016/j.apsusc.2015.11.005

[84] Dhanya, A., and Aparna, K. 2016. Synthesis and Evaluation of TiO_2/Chitosan based hydrogel for the Adsorptional Photocatalytic Degradation of Azo and Anthraquinone Dye under UV Light Irradiation, Procedia Technology, 24:611 – 618.

[85] Kazemi, F., Mohamadnia, Z., Kaboudin, B., and Karimi, Z. 2016. Photodegradation of methylene blue with a titanium dioxide/polyacrylamide photocatalyst under sunlight, Journal of Applied polymer Science, 133(19):(2016) 433386 (1-9).

[86] Yang, J., Chen, D., Zhu, Y., Zhang, Y., and Zhu, Y. 2016. 3D-3D Porous Bi_2WO_6/Graphene Hydrogel Composite with Excellent Synergistic Effect of Adsorption-Enrichment and Photocatalytic Degradation, Applied Catalysis B, Environmental, http://dx.doi.org/10.1016/j.apcatb.2016.12.035

[87] Zhang, M., Luo, W., Wei, Z., Jiang, W., Liu, D., and Zhu, Y. 2016 Separation free C_3N_4/SiO_2hybrid hydrogels as high active photocatalysts for TOC removal, Applied Catalysis B: Environmental, 194:105–110.

[88] Rasoulifard, M.H., Dorraji, M.S.S., and Mozafari, V. 2016. Visible Light Photocatalytic activity of Chitosan/ Poly(vinyl alcohol)/TiO_2Nanocomposite for Dye Removal: Taguchi-based Optimization, Environmental Progress & Sustainable Energy, 36(1):66-72.

[89] Wu, D., Yi, M., Duan, H., Xu, J., and Wang, Q. 2016. Tough TiO_2-rGO-PDMAA nanocomposite hydrogel via one-pot UV polymerization and reduction for photodegradation of methylene blue, Carbon, 108: 394-403.

[90] Jiang, W., Luo, W., Zong, W., Yao, W., Li, Z., and Zhu, Y. 2016 Polyaniline/Carbon Nitride Nanosheets Composite Hydrogel: A Separation-Free and High-Efficient Photocatalyst with 3D Hierarchical Structure, Small, 12(32): 4370-4378.

[91] Thomas, M., Naikoo, G.A., Sheikh, M.U., Bano, M., and Khan, F. 2016. Effective photocatalytic degradation of Congo red dye using alginate/ carboxymethyl cellulose/TiO_2 nanocomposite hydrogel under direct sunlight irradiation, Journal of Photochemistry and Photobiology A: Chemistry, 327:33–43.

[92] Zhang, Y., Cui, W., An, W., Liu, L., Liang, Y., and Zhu, Y. 2017. Combination of photoelectrocatalysis and adsorption for removal of bisphenolAoverTiO_2-graphene hydrogel with 3D network structure, Applied Catalysis B, Environmental, http://dx.doi.org/10.1016/j.apcatb.2017.08.076

[93] Jiang, Y., Chowdhury, S., and Balasubramanian, R. 2017. Nitrogen-doped graphene hydrogels as potential adsorbents and photocatalysts for environmental remediation, Chemical Engineering Journal, doi: http://dx.doi.org/10.1016/j.cej.2017.06.156

[94] Chen, F., An, W., Li, Y. Y. Liang, Cui, W. 2017 Fabricating 3D porous PANI/TiO_2-graphene hydrogel for the enhanced UV-light photocatalytic degradation of BPA, Applied Surface Science, http://dx.doi.org/10.1016/j.apsusc.2017.08.146

[95] Ma, Y., Wang, J., Xu, S., Zheng, Z., Du, J., and Feng, S. 2017 Ag_2O/sodium alginate supramolecular hydrogel as a film photocatalyst for removal of organic dyes in wastewater, RSC Advances,7:15077–15083.

[96] Chen, F., An, W., Liu, L., Liang, Y., Cui, W. Highly efficient removal of bisphenol A by a three-dimensional graphene hydrogel-AgBr@rGO exhibiting adsorption/photocatalysis synergy, Applied Catalysis B, Environmental .http://dx.doi.org/10.1016/j.apcatb.2017.05.078

[97] Mai, N.X.D., Bae, J., Kim, T., Park, S.H., Lee, G.W., Kim, J.H., Lee, D., Son, H.B., Lee, Y.C., and Hur, J. 2017. Recyclable, Recoverable, and Reformable Hydrogel-Based Smart Photocatalyst, Environmental. Science: Nano, 4(4):955-966.

[98] Shao, P., Ren, Z., Tian, J., Gao, S., Luo, X., Shi, W., Yan, B., Li, J., and Cui, F. 2017. Silica hydrogel-mediated dissolution-recrystallization strategy for synthesis of ultrathin α-Fe_2O_3 nanosheets with highly exposed (110) facets: A superior photocatalyst for degradation of bisphenol S, Chemical Engineering Journal, 323:64–73.

[99] Yuan, Z. and Chen, D., Fabrication and photocatalytic behavior of a novel nanocomposite hydrogels containing Fe-octacarboxylic acid phthalocyanine, Journal of Applied Polymer Science,134(42):45428 (1-7).

[100] Sharma, G., Alothman, Z.A., Kumar, A., Sharma, S., Ponnusamy, S.K., and Naushad, M. 2017. Fabrication and characterization of a nanocomposite hydrogel for combined photocatalytic degradation of a mixture of malachite green and fast green dye, Nanotechnology for Environmental Engineering, 2(1):1-4.

[101] Chen, F., Zhao, J., An, W., Hu, J., Liang Y., and Cui, W. 2017. Inserting AgCl@rGO into graphene hydrogel 3D structure: synergy of adsorption and photocatalysis for efficient removal of bisphenol A RSC Advances, 7 (63): 39814-39823.

[102] Mu, C., Zhang, Y., Cui, W., Liang, Y., and Zhu, Y., Removal of bisphenol A over a separation free 3D Ag_3PO_4-graphene hydrogel via an adsorption-photocatalysis synergy, Applied Catalysis B: Environmental, 212:(2017) 41–49.

[103] Lito, P.F., Aniceto, J.P.S., Silva, and C.M. 2012 Removal of Anionic Pollutants from Waters and Wastewaters and Materials Perspective for Their Selective Sorption, Water Air Soil Pollution, 223:6133–6155.

[104] Jiang, H., Qi, X.X., Zhao, J.Z., Ni, L., and Chen, Z.S. 2014 Synthesis of AMIMCl/AM copolymer hydrogel and its adsorption effect on permanganate anion, Separation Science and Technology, 49(6):915-923.

[105] Rahni, S.Y., Mirghaffari, N., Rezaei, B., Ghaziaskar, H.S. 2014 Removal of phosphate from aqueous solutions using a new modified bentonite-derived hydrogel, Water Air Soil Pollution, 225:1916.

[106] Su, Y., Liu, J., Yue, Q., Li, Q., and Gao, 2014. B. Adsorption of ammonium and phosphate by feather protein based semi-interpenetrating polymer networks hydrogel as a controlled-release fertilizer, Environmental Technology, 35(4):446–455.

[107] Wan, J., Tao, T., Zhang, Y., Liang, X., Zhou, A., and Zhu, C. 2016. Phosphate adsorption on novel hydrogel beads with interpenetrating network (IPN) structure in aqueous solutions: kinetics, isotherms and regeneration, RSC Advances, 6:23233-23241.

[108] Genc, F., Uzun, C., and Guven, O. 2016. Quaternized poly(1-vinylimidazole) hydrogel for anion adsorption, Polymer Bulletin, 73(1):179-190.

[109] Zhang, H., Yu, M., Zhang, H., and Bai, L. 2016. Facile preparation and evaluation of allyl amine hydrochloride-based porous hydrogel without calcium and aluminum: an alternative candidate of phosphate binder, Polymer Bulletin,73(12):3371–3384.

[110] Siwek, H., Bartkowiak, A., Włodarczyk, M., and Sobecka, K. 2016. Removal of phosphate from aqueous solution using alginate/ iron (iii) chloride capsules: a Laboratory Study, Water Air and Soil Pollution, 227(11):427-436.

[111] Dong, S., and Wang, Y. 2016. Characterization and adsorption properties of a lanthanum-loaded magnetic cationic hydrogel composite for fluoride removal, Water Research, 88 (2016) 852-860.

[112] Alka, T., and Anita, B. 2014. Effective Removal of Pesticide (Dichlorvos) by Adsorption onto Super Paramagnetic Poly (styrene-co-acrylic acid) Hydrogel from Water, International Research Journal of Environment Science, 3(11):41-46.

[113] Dwivedi, C., Gupta, A., Chaudhary, A., and Nandi, C.K., 2014 Gold nanoparticle chitosan composite hydrogel beads show efficient removal of methyl parathion from waste water, RSC Advances, 4:39830-39838.

[114] Zhuang, Y., Yu, F., Ma, J., and Chen, J. 2015 Adsorption of ciprofloxacin onto graphene–soy protein biocomposites, New Journal of Chemistry,39:3333-3336.

[115] Genc, N., Dogan, E.C., and Yurtsever, M., Bentonite for ciprofloxacin removal from aqueous solution, Water and Science Technology, 68(4):848-855.

[116] Zhuang, Y., Yu, F., and Ma, J. 2015 Enhanced adsorption and removal of ciprofloxacin on regenerable long TiO_2 nanotube/graphene oxide hydrogel adsorbents, Journal of Nanomaterials, 2015:1-8.

[117] Ma, J., Yang, M., Yu, F., and Zheng, J. 2015. Water-enhanced removal of ciprofloxacin from water by porous graphene hydrogel, Scientific Reports, 5:13578.