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REVIEW

Alginate Hydrogel Adsorbents in Adsorption of Inorganic and Organic Pollutants: A Review

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The present review discusses various alginate hydrogel adsorbents with unique adsorption performance in environmental remediation. Novel alginate composites were developed with, high swelling capacity and capable of adsorbing toxic inorganic and organic pollutants. Alginate hydrogel adsorbents were developed with a single network and double network structure with excellent adsorption ability in removal of toxic inorganic and/or organic pollutants. Alginate with single or double network composite hydrogels were developed when alginate was combined with graphene/chitosan/polymer to get superior adsorbents in removal of toxic pollutants. Acrylic acid/alginate hydrogel in recent studies are efficient in the elimination of inorganic and organic contaminants. This review will generate interest to researchers to develop novel alginate composite hydrogels with unique properties in the adsorption of toxic inorganic, organic contaminants. This work provides a worthy challenge and the future possibility of designing novel alginate materials for various applications.

Keywords: Alginate hydrogel, Dye, Heavy metals, Adsorption, Adsorption capacity.

INTRODUCTION

The present worldwide concern on the toxicity of water pollutants such as, toxic inorganic elements, organic dyes, pharmaceutical and other industrial organic wastes is a great challenge in developing countries [1-7]. The agricultural and industrial discharges with toxic inorganic and organic pollutants are non-biodegradable water contaminants that lead to serious health hazards in human beings through the food chain [8-11]. Some organic and inorganic pollutants were encountered as worst contaminants in water bodies with persistent nature that adversely affect the aquatic environment [12]. Hence there is more interest to explore an efficient methodology for pollutant removal from contaminated water. A few remediation techniques were utilized for the evacuation of harmful toxins, for example photocatalytic degradation [13], chemical precipitation [14], reverse osmosis [15], adsorption [16-18], *etc.*, Some skill methods used in treatment are costly, difficult for operation and time consuming, however an advantageous simple and high efficient treatment method in the removal of pollutants is adsorption. In literature, preparation of alginate hydrogel was used as efficient adsorbents with good adsorption performance in removal of dyes, heavy metals, pharmaceutical, other organic wastes. Also some alginate hydrogel composites prove to be promising materials in removal of toxic organic and inorganic contaminants and this has created interest to various researchers and scientists to develop novel alginate hydrogel adsorbents with unique properties and superior adsorption performance in treating contaminated water.

Alginate hydrogel adsorbents: Sodium alginate is a nontoxic and inexpensive natural polysaccharide [19-21] having hydroxyl and carboxyl groups that can be effectively crosslinked with Ca^{2+}, Fe^{3+}, Y^{3+} ions. Alginate is usually modified using varying physico-chemical process to increase its mechanical properties and adsorption performance [22]. The structure of sodium alignate is given in Fig. 1 [23]. Hydrogels are characterized as three-dimensional (3D) networks that has special structure, which allows the diffusion of solute that results in swelling in aqueous solution. Some novel hydrogels were prepared by mixing chemically cross linked network with physically cross linked network. Alginate based composites were developed with a single and double network structure with high swelling capacity and adsorption performance. When alginate was combined with other materials (graphene/chitosan/polymer), then the adsorption efficiency of the hydrogel, in the adsorption of inorganic and organic pollutants increases. Double network hydrogels have higher strength and has good mechanical strength than the single network hydrogel [24]. When carboxylated cellulose (CC) was incorporated into the alginate hydrogel then the number of negatively charged carboxylate ions in the hydrogel increases and this is evident from zeta potential analysis [25].

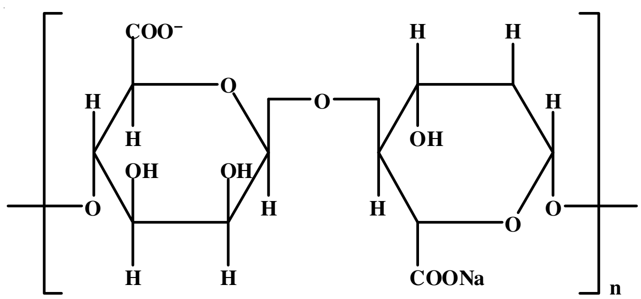

Fig. 1. Chemical structure of sodium alignate

Adsorption using calcium alginate hydrogel has gained significant attention due to high specific surface area [26], low cost and its composite forming tendency with other materials [27]. In preparing novel composite hydrogels with high adsorption performance, alginate hydrogels were modified to get more porous and more functional groups that are capable of adsorbing toxic inorganic and organic pollutants. In the literature, alginate hydrogel adsorbents have gained significant attention due to their high adsorption performance [28]. Thus, alginate hydrogel adsorbents with unique surface properties were prepared and reported as efficient adsorbents in environmental remediation.

Alginate hydrogel adsorbents in adsorption

Alginate/graphene hydrogel adsorbents in environmental remediation: In past decade, several alginate/graphene hydrogel adsorbents were prepared and reported to have wide applications in environmental remediation by various researchers [28-38]. Alginate/graphene adsorbents were prepared by using sodium/calcium alginate mixed with graphene and other crosslining agents, *etc.* to prepare novel alginate/graphene adsorbents [28-31] and their adsorption in removal of toxic dyes and heavy metals and pharmaceutical compounds has been tested. Zhuang *et al.* [28] synthesized double-network hydrogel composed of GO and alginate (GAD) and GAS, investigated the adsorption of methylene blue through batch, column adsorption. The thermal stability of GAD was better than GAS. The BET surface area analysis of GAD and GAS were 37.6 m^2/g and 3.4 m^2/g, respectively. The microporous nature of GAD is the reason for the difference in the adsorption performance difference between GAD and GAS and the adsorption capacity was reported as 1.84 g/g for GAS and 2.30 g/g for GAD. Verma *et al.* [29] prepared sodium alginate crosslinked acrylic acid/graphite powder (NaA-cl-AAc/GP hydrogel) and studied the adsorption of malachite green dye. The NaA-cl-AAc/GP posses' good thermal stability due to the positive influence of GP as inferred from TGA analysis. The NaA-cl-AAc/GP hydrogel's active surface was reported to show negative surface charge at pH 7. The adsorption capability of malachite green onto NaA-cl-AAc/GP was due to distinctive surface morphology, large surface area and strong functional groups. Analysis of isotherm data of NaA-cl-AAc/GP show that Langmuir model fit well for malachite green dye adsorption and the adsorption capacity was recorded to be 628.93 mg/g. Polyethyleneimine functionalized, graphene oxide embedded calcium alginate (fGOCA) hydrogel was synthesized by Arshad *et al.* [30] and tested the adsorption of Pb(II), Hg(II) and Cd(II) from aqueous solution. The surface functional groups of fGOCA, the $-NH_2$ functional group of polyethyleneimine favours the adsorption of metal ions onto fGOCA. The adsorption of heavy metals onto fGOCA was studied at varying experimental conditions and the adsorption capacity for Pb(II), Hg(II) and Cd(II) removal was reported as 602, 374 and 181 mg/g respectively. He *et al.* [31] synthesized novel yttrium-immobilized-graphene oxide-alginate hydrogel (Y-GO-SA) and tested the adsorption of arsenic and tetracycline (TC). The adsorption of Y-GO-SA was highly pH dependent and the optimum pH for As(V) and TC was 5.0 and 7.0, respectively. The adsorption of As(V) and TC onto the Y-GO-SA hydrogel was due to chemisorptions. The isotherm studies on the adsorption of As(V) and TC adsorption onto Y-GO-SA was carried out and the adsorption capacity was reported as 273.39 and 477.9 mg/g. Graphene oxide/alginate hydrogel membrane (GAHMs) was tested as an adsorbent by Bai *et al.* [26] for the evacuation of Cr(III) and Pb(II) ions. The GAHMs adsorption of metal ions before and after adsorption was portrayed by FTIR, SEM, EDAX and XPS and the results indicate that hydrophilic groups were involved in the adsorption process. The optimum pH for GAHMs adsorption of Cr(III) and Pb(II) was 6.0 and 5.0. The adsorption mechanism of GAHMs involves electrostatic attraction, chelate reaction and ion exchange process. The carboxyl and hydroxyl groups in the GAHMs were primarily involved in the adsorption process. The isotherm data in adsorption of both metal ions by GAHMs was best described by Langmuir model and the adsorption capacity was 327.9 mg/g [Pb(II)) and 118.6 mg/g (Cr(III)], respectively. The adsorption of ciproflaxin was investigated using NH_2DN [32], CNTs/L-Cyst@GO/SA [33], GAD hydrogel [34], GSA [35] and the adsorption capacity was reported as 301.36, 181, 344.83 and 100 mg/g, respectively. In NH_2DN [32], the thermal and mechanical properties were superior due to the double network structure.

In CNTs/L-Cyst@GO/SA [33], the functional groups present on the ciproflaxin adsorbs the hydrogel by electrostatic interaction. Sodium alginate grafted polyacrylamide/graphene oxide hydrogel (SA-PAM/GO) was synthesized by Jiang *et al.* [36] and studied the removal of Cu(II) and Pb(II) ions from aqueous solution. The SA-PAM/GO adsorption of Cu(II) and Pb(II) was affected by pH, ionic strength, equilibrium time and concentration. The optimum pH for Cu(II) and Pb(II) adsorption was pH-5 and pH-5.5. The adsorption kinetic results of SA-PAM-GO reveal that chemical adsorption mechanism controls the adsorption of heavy metal ions. The adsorption capacity was reported as 68.76 and 240.69 mg/g for Cu(II) and Pb(II), respectively. Yi *et al.* [37] prepared graphene oxide encapsulated polyvinyl alcohol/sodium alginate hydrogel (SPG hydrogel) and studied the removal of Cu(II) ions in batch mode adsorption experiments. The content of Cu(II) ions adsorbed onto SPG hydrogel was dependent on pH, competing ions and initial concentration of metal ion. The adsorption of Cu(II) on the surface of SPG was by ion-exchange (or) complexation process. The isotherm data was best described by Langmuir model and the monolayer adsorption capacity of Cu(II) was reported as 247.16 mg/g, respectively. Feng *et al.* [38] prepared black-pearl reduced rGO oxide-sodium alginate (rGO (50)-SA hydrogel) and the adsorption of phenol was tested by changing the contact time, initial phenol concentration and temperature and rGO (50)-SA dosage. The optimum rGO content in the composite hydrogel was 50% with favourable adsorption for phenol and the adsorption capacity was reported as 49.358 mg/g.

Table-1 presents the adsorption studies of several alginate/graphene hydrogel adsorbents accessed as promising materials in removal of organic and inorganic pollutants in aqueous phase. If the analyze the adsorption performance of all the graphene/alginate adsorbents in environmental remediation graphene/alginate double network hydrogel [28,32,34] was reported to have high adsorption capacity in removal of dyes, heavy metal and pharmaceutical compounds. Also alginate/acrylic acid composite hydrogel [29] is quite promising adsorbent in removal of methylene blue. Thus, the adsorption potential of alginate/graphene hydrogel adsorbent is promising in environmental remediation. The FTIR spectrum of calcium alginate beads, magnetic calcium alginate beads, methyl violet-loaded calcium alginate beads and methyl violet-loaded magnetic calcium alginate beads are shown in Fig. 2.

TABLE-1
ALGINATE/GRAPHENE HYDROGEL ADSORBENTS IN ADSORPTION OF POLLUTANTS

Alginate hydrogel	Pollutant name	Adsorption capacity (mg/g)	Ref.
GAD hydrogel	MB	2.3 g/g	[28]
GAS hydrogel	MB	0.6 g/g	[28]
NaA-cl-AAc/GP	MG	628.93	[29]
fGOCA	Pb(II)	602.00	[30]
	Hg(II)	374.00	[30]
	Cd(II)	181.00	[30]
Y-GO-SA	TC	477.90	[31]
	As(V)	273.39	[31]
GAHMs	Pb(II)	327.90	[26]
	Cr(III)	118.60	[26]
NH_2-DN	CIP	301.36	[32]
	Cu(II)	153.91	[32]
CNTs/L-cys@GO/SA	CIP	181.00	[33]
SA-PAM-GO	Pb(II)	240.69	[36]
	Cu(II)	68.76	[36]
GAD hydrogel	CIP	344.83	[34]
	TC	290.74	[34]
SPG	Cu(II)	247.16	[37]
GSA	CPX	100.00	[35]
rGO (50)-SA hydrogel	Phenol	49.358	[38]

Alginate/polymer hydrogel adsorbents in removal of pollutants: Several researchers prepared alginate/polymer hydrogel adsorbents [39-48] and investigeated the adsorption performance in removal of organic and inorganic pollutants. Single network alginate/polyvinyl alcohol hydrogel (SAP) and double network alginate/polyvinyl alcohol hydrogel (DAP) was prepared by Kong *et al.* [39] for the adsorption of methylene blue. Batch experiments were conducted for methylene blue removal using SAP and DAP adsorbents and the effect of pH, contact time, temperature and initial methylene blue dye concentrations were investigated. The adsorption isotherm data of SAP and DAP fitted well to Langmuir model and the adsorption capacity was found to be 1255.75 mg/g and 1437.48 mg/g, respectively. Makhado *et al.* [40] synthesized sodium alginate poly(acrylic acid) (SA-poly(AA)) hydrogel and sodium alginate poly(acrylic acid)/zinc oxide (SA-poly(AA/ZnO) hydrogel and tested the adsorption of methylene blue dye. The incorporation of zinc oxide nanoparticles onto SA-poly(AA) hydrogel matrix enhanced the water uptake. The SA-poly(AA)/ZnO was reported

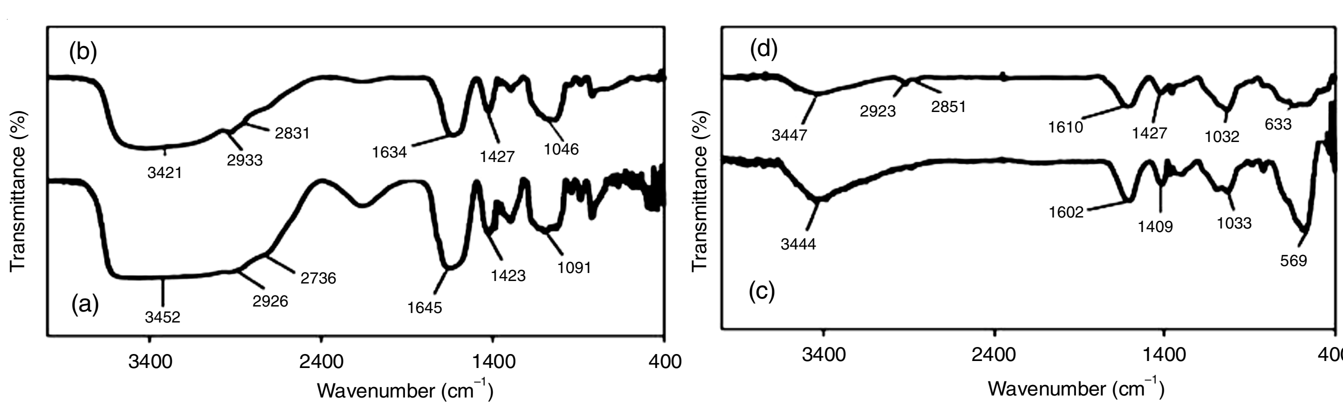

Fig. 2. FTIR spectra of: (a) calcium alginate hydrogel beads, (b) methyl violet-loaded calcium alginate hydrogel beads, (c) magnetic hydrogel beads and (d) methyl violet-loaded magnetic hydrogel beads

to have good adsorption for methylene blue on comparing with SA-poly(AA) hydrogel and the adsorption capacity was found to be 1529.6 mg/g and 1129.0 mg/g, respectively. Bhattacharyya & Ray [41] prepared semi-ipn hydrogel of acrylic copolymers and sodium alginate (CPSA4) and tested the hydrogel with optimum swelling characteristics in removal of basic fuschine and methyl violet from aqueous solution. The adsorption of basic fuschine dye was more than methyl violet dye due to their structural difference and the adsorption was found to increase with temperature. The basic fuschine and methyl violet dye adsorption data fitted well to Langmuir isotherm model and the adsorption capacity was found to be 763 and 550 mg/g for basic fuschine and methyl violet, respectively. The adsorption of Cu(II) ions on PAA-SA NFHs [42], ALG/PEI hydrogel [43] was carried out and the adsorption capacity was reported as 591.7 and 322.6 mg/g, respectively. The PAA-SA NFHs surface characteristics and adsorption performance was analyzed based on the PAA/SA precursor solution concentration and optimized (4:1 ratio) to get PAA-SA NFHs with intrinsic fibrous morphology and good adsorption behaviour. The ALG/PEI hydrogel exhibits a porous structure with various pore size and achieved a high Pb(II) ion adsorption capacity of 344.8 mg/g.

Jiang *et al.* [43] & Godiya [44] prepared versatile core/shell like alginate@polyethylenimine composite (Alginate@PEI-1.5) and studied the removal of Cr(VI), Pb(II) and Cu(II) ions. The strong chelation between the Pb(II) and Cu(II) ions and the abundantly deprotonated amino groups facilitate the rapid adsorption as pH was adjusted to 5.5 and 4.5, respectively. The PEI modification resulted rapid immobilization of amino groups into the alginate beads and thus the protonated amino group at pH-2 exhibits electrostatic interaction with Cr(VI) that resulted in enhanced adsorption. The Alginate@PEI-1.5 shows high adsorption for the tested metal ions and the adsorption capacity was found to be 497.1, 535.6 and 163.7 for Cr(VI), Pb(II) and Cu(II), respectively. Bahrami *et al.* [45] prepared, double network hydrogel of sodium alginate/polyacrylamide cross linked with octaaminopropyl polyhedral oligomeric silsesquioxane hydrochloride salt (OA-POSS) (Alg/PAAm/OA-POSS). The swelling and dye adsorption behaviour of Alg/PAAm/OA-POSS hydrogel was investigated at varying conditions. The isotherm study shows that the Langmuir model fits well for the adsorption of methylene blue onto Alg/PAAm/OA-POSS hydrogel and the adsorption capacity was 75.4117 mg/g. Mandal & Ray [46] synthesized poly(acrylic acid-*co*-hydroxy ethyl methacrylate) and sodium alginate (SCPIPN) and tested the removal of Congo red and methyl violet from aqueous medium. The SCPIPN was found to have high affinity to Congo red and methyl violet and the adsorption capacity was reported as 172 and 120 mg/g, respectively. Mahdavinia *et al.* [47] synthesized κ-carrageenan (Car) and sodium alginate (Alg) biopolymer (CarAlg-MMt) and tested the adsorption of crystal violet. The experimental data of CarAlg-MMt was analyzed using the Langmuir model and the adsorption capacity was reported as 88.8 mg/g respectively. Sodium alginate/gelatin based ZnS nanocomposite hydrogel (ZnS/NaAla-Gel-cl-polyAAm) was synthesized by Priya *et al.* [48] and studied the removal of Biebrich scarlet and crystal violet. The adsorption of dye by nanocomposite was optimized and under optimum conditions, the Biebrich scarlet and crystal violet showed 97.37% and 94.45% dye removal. The isotherm study of ZnS/NaAla-Gel-cl-polyAAm adsorption suggests monolayer adsorption and the adsorption capacity was found to be 1.96 and 9.32 mg/g for Biebrich scarlet and crystal violet, respectively.

Table-2 presents the adsorption performance of alginate/polymer hydrogel adsorbents in removal of organic and inorganic pollutants. If we examine the adsorption performance it is obvious that polyvinyl alcohol/alginate single and double network hydrogel [39] has excellent adsorption for methylene blue. Also acrylic acid/alginate composites [40-42] were hydrogels with unique properties and significantly high adsorption performance in removal of inorganic and organic pollutants. Thus, acrylic acid/polymer hydrogels are excellent adsorbents in environmental remediation.

TABLE-2
ALGINATE/POLYMER HYDROGEL
ADSORBENTS IN ADSORPTION OF POLLUTANTS

Alginate hydrogel	Pollutant name	Adsorption capacity (mg/g)	Ref.
DAP hydrogel	MB	1437.48	[39]
SAP hydrogel	MB	1255.75	[39]
SA-Poly(AA) hydrogel	MB	1529.60	[40]
SA-Poly(AA)/ZnO hydrogel	MB	1129.00	[40]
CPSA4	BF	763.00	[41]
	MV	550.00	[41]
PAA-SANFHs	Cu(II)	591.70	[42]
ALG/PEI	Cu(II)	322.60	[43]
	Pb(II)	344.80	[43]
Alginate@PEI-1.5	Pb(II)	535.60	[44]
	Cr(VI)	497.10	[44]
	Cu(II)	163.70	[44]
Alg/PAAm/OA-POSS	MB	75.4117	[45]
SCPIPN	CR	172.00	[46]
	MV	120.00	[46]
CarAlg/MMt	CV	88.80	[47]
ZnS/NaAla-Gel-cl-PolyAAm)	BS	1.96	[48]
	CV	9.32	[48]

Alginate/chitosan adsorbent in removal of pollutants: Studies on various alginate/chitosan adsorbents was carried out in the past decade by several researchers and tested in removal of organic and inorganic pollutants [49-51]. Tang *et al.* [49] prepared a novel physically cross linked double network hydrogel of chitosan/sodium alginate/calcium ion (CTS/SA/Ca^{2+} PCDNH) and investigated the removal of Pb(II), Cu(II) and Cd(II) ions. The FTIR and ^{13}C-SSNMR analysis of PCDNH revealed a structure composed of physically crosslinked CTS/SA and SA/Ca^{2+} networks. The porous structure of PCDNH provides a large surface area and binding sites for the swelling and adsorption of Pb(II), Cu(II) and Cd(II) ions. The isotherm study of metal ions adsorption onto PCDNH show that the adsorption is a chemo-physical adsorption and the adsorption capacity calculated from Langmuir model was found to be 215.20-400.90 mg/g (Pb^{2+}), 57.94-71.10 mg/g (Cu^{2+}) and 83.95-110.69 mg/g (Cd^{2+}), respectively. The Pb(II) adsorption on CHT/ALG/Fe_3O_4@SiO_2 composite hydrogel [50] and

WSC-SA hydrogel [51] gave adsorption capacities of 234.77 mg/g and 66 mg/g, respectively. The CHT/ALG/Fe_3O_4@SiO_2 composite hydrogel [50] was non-toxic as there is no polyvalent metals or organic compounds utilized as crosslinking agents in preparing the novel adsorbent. The adsorption of Cu(II) by CCM hydrogel [52], WSC-SA hydrogel [51] and Mag-Ben/CCS/Alg [53] was carried out and the adsorption capacity was reported as 143.276, 88.2 and 56.79 mg/g, respectively. The WSC-SA hydrogel has, good binding capacity for Pb^{2+} and Cu^{2+} due to the presence of plenty of inherent functional sites on the adsorbent. The adsorption of Cu(II) onto Mag-Ben/CCS/Alg attained equilibrium in 90 min at pH 5 with Cu(II) % removal of 92.62 ± 0.39%. The Mag-Ben/CCS/Alg composite hydrogel was easily separated from the solution after adsorption due to its magnetic nature. Hydrogel beads of poly(vinyl alcohol) sodium alginate-chitosan-montmorillionite (MMTNs) was prepared by Zhang *et al.* [54] and tested the adsorption capacity of methylene blue from aqueous solution. The stability and microstructure of MMTNs were studied by FTIR, TG and SEM. The process parameters in the adsorption of methylene blue by MMTNs were optimized. The MMTNs was reported to have high affinity to methylene blue and the adsorption capacity was reported as 137.15 mg/g, respectively. Shehzad *et al.* [55] prepared novel hybrid biocomposite based on aminothiocarbamate derivative of alginate/carboxymethyl chitosan/TiO_2(TiO_2/TSC-CMC) and studied the adsorption of Ni(II) ions. The optimized adsorbent 3TiO_2/TSC-CMC hydrogel was found to be mesoporous, stable and integrated in highly acidic or basic media. The isotherm study on the adsorption of Ni(II) onto 3TiO_2/TSC-CMC shows that the Langmuir model fitted well with a monolayer adsorption capacity of 172 mg/g at pH 6.0 and 298K. The authors concluded that 3TiO_2/TSC-CMC is a cost effective and efficient adsorbent in the remediation of Ni(II) ions from aqueous solution.

Table-3 presents some of the alginate/chitosan adsorbents studied in removal of inorganic and organic contaminants. Among all the alginate/chitosan adsorbents alginate/chitosan/alginate double network hydrogel [49] has comparatively good adsorption in metal ion adsorption. Hence, alginate based composite developed with a network structure proved to be efficient adsorbents in environmental remediation.

Some other alginate hydrogel in removal of pollutants: Several authors in the recent past synthesized alginate hydrogel

TABLE-3
ALGINATE/CHITOSAN HYDROGEL ADSORBENTS IN ADSORPTION OF POLLUTANTS

Alginate hydrogel	Pollutant name	Adsorption capacity (mg/g)	Ref.
CS/SA/Ca^{2+}PCDNH	Pb(II)	215.20-400.90	[49]
	Cu(II)	57.94-71.10	[49]
	Cd(II)	83.95-110.69	[49]
CHT/ALG/Fe_3O_4@SiO_2	Pb(II)	234.77	[50]
WSC-SA hydrogel	Cu(II)	88.20	[51]
	Pb(II)	66.00	[51]
CCM hydrogel	Cu(II)	143.276	[52]
Mag-Ben/CCS/Alg	Cu(II)	56.79	[53]
MMTNs	MB	137.15	[54]
TiO_2/TSC-CMC	Ni(II)	172.00	[55]

adsorbents [56-60] with significant interest and tested its adsorption performance in removal of inorganic and organic contaminants. The methylene blue adsorption on some of the high adsorption capacity alginate hydrogel adsorbents include SA-cl-poly(AA-TiO_2) [56], SCFA hydrogel [57], MSH-AB hydrogel [58], DAA-Ge-Ag hydrogel [59]) and SA/PEI hydrogel [60] with adsorption capacities 2257.36, 1335, 785.45, 625 and 400 mg/g, respectively. The adsorption of methylene blue onto SA-cl-poly(AA-TiO_2 is due to the functional groups on the surface that increased the affinity for methylene blue and resulted in facile and fast adsorption. The highly porous network and the abundant oxygen containing groups of SCFA hydrogel provided efficient adsorption performance of methylene blue dye.

Alginate/polyaspartate (Alg/PASP) hydrogel was prepared by Jeon *et al.* [61] and studied the removal of methylene blue and methylene green dyes. Batch mode adsorption studies on Alg/PASP were carried out at 25 ºC. The authors concluded that the ionic interaction between the Alg/PASP hydrogel and dye molecule facilitates the rapid adsorption of cationic dyes methylene blue and methylene green with adsorption capacity as much as 600-700 mg/g for methylene blue and 300-350 mg/g for methylene green dye. Shen *et al.* [62] prepared modified graphitic carbon nitride composite hydrogel (*g*-C_3N_4/SA) and characterized using FTIR, SEM, EDS and XPS analysis. The authors tested the adsorption of *g*-C_3N_4/SA in removal of Pb(II), Ni(II) and Cu(II) through batch mode adsorption experiments. The adsorption of *g*-C_3N_4/SA was pH dependent and the isotherm analysis using Langmuir model was reported to have high adsorption capacity of 384.4, 306.3 and 168.2 mg/g for Pb(II), Ni(II) and Cu(II), respectively. Carboxylated cellulose nanocrystal/sodium alginate hydrogel (CCN-Alg) was prepared by Hu *et al.* [63] and studied the adsorption of Pb(II) ions. The CCN-Alg characterization results reveal that the hydroxyl and carboxyl group present on the surface favours the complexation of lead ions. The adsorption isotherm data of CCN-Alg fitted well to Langmuir model and the adsorption capacity was 338.98 mg/g. Shehzad *et al.* [64] prepared aminocarbamate moiety grafted calcium alginate hydrogel (PSC-CA) and tested the adsorption of Ag(I) ions. The characterization results of PSC-CA proved the successful modification and loading of Ag(I) ions into the material. Adsorption studies of PSC-CA showed that the pH, adsorbent dose, contact time and Ag(I) concentration influenced the adsorption process. The isotherm data of PSC-CA fitted well to Langmuir model and the adsorption capacity calculated at 298 K was reported as 210 mg/g. Yu *et al.* [65] κ-carrageenan/sodium alginate double network hydrogel (κ-Car/SA) and studied the removal of antibiotics. As the κ-Carrageenan concentration increased, the intermolecular interction and viscosity increased. The κ-Car/SA hydrogel was analyzed using FTIR and zeta potential test and this revealed that there is electrostatic interaction and hydrogen bonding between κ-Car/SA and ciproflaxin. The adsorption of ciproflaxin onto κ-Car/SA was optimum at pH 4 and the adsorption capacity was reported as 229 mg/g, respectively. The adsorption of methylene blue on CNC-ALG hydrogel [66] and PC@Fe_3O_4-NPs@Alginate [71] gave adsorption capacities of 255.5

mg/g and 180.42 mg/g, respectively. Djelad *et al.* [67] prepared alginate-whey composite hydrogel (ALG-AW) and assertained using FT-IR analysis. The adsorption of crystal violet dye onto ALG-AW was investiagted in batch adsorption experiments and the optimum adsorbent dose was 0.4g/L at pH 6. The ALG-AW adsorption isotherm data fitted well to Langmuir model and the adsorption capacity was reported as 220 mg/g. Varaprasad *et al.* [68] prepared *Laminaria digitata* brown sea weed based alginate hydrogel (LDBS hydrogel) and studied the adsorption of Pb(II) ions. The results of optical microscope image shows that the porous network of the LDBS hydrogel increased on increasing the alginate content in the hydrogel.

The LDBS hydrogel containing high alginate content was reported to have highest swelling capacity (109.3 g/g). The adsorption of Pb(II) onto LDBS hydrogel was rapid and the adsorption capacity was reported to be 110 mg/g. Zhang *et al.* [69] prepared novel biochar/pectin/alginate hydrogel beads (BPA) and investigated the adsorption of Cu(II) from aqueous solution. The BPA adsorption was studied in batch mode and the adsorption parameters were optimized. The isotherm data fitted well to Freuendlich model and the adsorption capacity of BPA was reported as ~ 80.6 mg/g. Dai *et al.* [70] prepared alginate/starch ether composite hydrogel, tested the adsorption of Cu(II) ions. The impct of pH and initial Cu(II) concentration was studied for HIPS/SA. The adsorption isotherm data of HIPS/SA fitted well to Langmuir model and the adsorption capacity was 25.81 mg/g.

Wang *et al.* [72] prepared TAL hydrogel for phosphate adsorption. The TAL hydrogel adsorption was optimized (1% talc, 0.15 M La^{3+}) in the pH of 4-6. The adsorption of phosphate onto TAL hydrogel at optimum pH 4 arrived at a most extreme adsorption limit of 16.4 mg P/g. Pan *et al.* [73] prepared NIPAM and TBP reinforced with calcium alginate (NIPAM/TBP/CA hydrogel) and tested the adsorption of phenol from aqueous solution. The calcium alginate present in NIPAM/TBP hydrogel improved the mechanical sturdiness of the hydrogel. Iron nanoparticles-calcium alginate hydrogel membrane (FeNPs-CaAlg hydrogel) was prepared by Liu *et al.* [74] and tested the removal of Cr(VI) ions. The composite hydrogel at 23 ºC can remove 99.5% of 1 mg/L Cr(VI) (50 mL) in 10 min for an FeNPs-CaAlg dose of 0.6g at pH 5.41.

Table-4 presents the alginate hydrogel adsorbents with varying adsorption performance in the adsorption of inorganic and organic pollutants. Among all the other alginate hydrogel adsorbents, (SA-cl-poly(AA-TiO_2) had a superior adsorption performance (2257.36 mg/g) in dye removal.

The literature review summarizes the various alginate hydrogel adsorbents that were synthesized and proved to be good adsorbents in the removal of inorganic and organic pollutants [56-74]. On comparing the various alginate hydrogel adsorbents, it is observed that alginate/double network hydrogel [24,32,39,49]. Also, some recent studies on adsorption show that when acrylic acid was present in the alginate hydrogel adsorbent then the materials proved to be promising in the removal of inorganic and organic pollutants [29,40,52,56]. Hence, alginate hydrogels prepared by combining alginate with graphene/chitosan/polymer gave hydrogels with high adsorption performance in evacuation of organic, inorganic pollutants. This review will guide researchers to utilize alginate hydrogels in preparing novel composite materials in environmental remediation.

TABLE-4
OTHER ALGINATE HYDROGEL
ADSORBENTS IN ADSORPTION OF POLLUTANTS

Alginate hydrogel	Pollutant name	Adsorption capacity (mg/g)	Ref.
SA-cl-poly(AA)-TiO_2	MB	2257.36	[56]
SCFA hydrogel	MB	1335.00	[57]
MSH-AB hydrogel	MB	785.45	[58]
DAA/Ge/Ag hydrogel	MB	625.00	[59]
SA/PEI hydrogel	MB	400.00	[60]
Alg/PASP	MB	600-700	[61]
	MG	300-350	[61]
g-C_3N_4/SA	Pb(II)	383.40	[62]
	Ni(II)	306.30	[62]
	Cu(II)	168.20	[62]
CCN-Alg hydrogel	Pb(II)	338.98	[63]
PSC-CA	Ag(I)	210.00	[64]
k-Car/SA	CIP	229.00	[65]
CNC-ALG	MB	255.50	[66]
ALG-Aw	CV	220.00	[67]
LDBS hydrogel	Pb(II)	110.00	[68]
BPA	Cu(II)	80.60	[69]
HIPS/SA hydrogel	Cu(II)	25.81	[70]
PC@Fe_3O_4-NPs@Alginate	MB	49.66	[71]
TAL hydrogel	Phosphate	16.40	[72]
NIPAM/TBP/CA hydrogel	Phenol	–	[73]
FeNPs-CaAlg hydrogel	Cr(VI)	–	[74]

Conclusion

There are more systematic studies in preparing novel alginate hydrogel adsorbents in the removal of inorganic and organic pollutants. It's obvious that toxicity dyes, heavy metals, pharmaceutical compounds and other toxic compounds on the environment leads to adverse effect and is of concern in to preserve the environment. Hence, alginate hydrogel adsorbents have gained attention in the adsorption of toxic inorganic, organic contaminants. The literature suggests that novel alginate double network hydrogels provided efficient adsorption in the removal of pollutants. Also, some alginate/acrylic acid hydrogels were promising materials with superior adsorption performance in environmental remediation. A positive insight to prepare novel alginate hydrogel composite materials with good surface properties in the future may be used as unique materials in environmental remediation.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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